MONTAN
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LEOBEN

Chair of Process Technology and Industrial Environmental Protection

Doctoral Thesis

Techno-economic assessment of emerging power-to-gas technologies using advanced generic methods

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April 2022

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AFFIDAVIT

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I declare that I have read, understood, and complied with the guidelines of the senate of the Montanuniversität Leoben for "Good Scientific Practice".

Furthermore, I declare that the electronic and printed version of the submitted thesis are identical, both, formally and with regard to content.

Date 05.04.2022

Signature Author
Hans Böhm

"It is difficult to make predictions, especially about the future."

— *A Danish politician*

Acknowledgment

I would like to thank Univ.-Prof. Dipl.-Ing. Dr.-Ing. Markus Lehner for providing me the opportunity to conduct this thesis under his supervision and guidance. Furthermore, I appreciate the mentoring of him and Univ.-Prof. Dipl.-Ing. Dr.techn. Thomas Kienberger, as well as their valuable feedback and availability for discussions of energy topics regarding this work and beyond.

Moreover, I greatly appreciate the support of the association *Energieinstitut an der Johannes Kepler Universität Linz* currently under the guidance of Dr. Robert Tichler and Prof. Dr. Horst Steinmüller previously, on the elaboration of this thesis. The diversity of the projects I was able to contribute to and the corresponding insights into interdisciplinary energy topics had a significant impact on the holistic view incorporated in this work. In this context, I would like to thank the entire *Energieinstitut* team for the constant exchange of expertise and knowledge and always helpful teamwork. Special thanks go to Andreas and Stefan for being such great office mates and always being open to fruitful discussions on all kinds of topics.

Finally, I thank my family, including my parents Monika and Hans, and friends for their everlasting support, along with their constant questioning about when I will finally finish. In addition, not to be forgotten, my beloved companion in life, Romana, for her patience and appreciation over all the years we have been together now.

Hans Böhm, April 2022

Abstract

The efforts required to achieve the climate targets set in the Paris Agreement and the associated transition of energy generation and supply necessitate the implementation of new energy technologies. In addition, the remaining carbon budgets involved in reaching these targets require early and effective action. Therefore, policy- and decision-makers must rely on comprehensive assessments to identify the economic viability and effectiveness of climate change mitigation of emerging energy technologies. Thus, they must be able to set supportive measures and regulations or decide on corresponding investments.

In terms of the energy transition, apart from electrification, the demand for renewable gases, such as hydrogen or synthetic natural gas (SNG), is substantial. Therefore, power-to-gas (PtG) is a fundamental cornerstone of future renewable and sustainable energy systems. However, the corresponding technologies are still in a relatively early stage of technological maturity, especially regarding implementations at industrial scale. On one hand, this leads to hesitancy in their implementation, while on the other, the hydrogen demand of >1500 TWh/a identified for the EU suggests an early and rapid expansion of capacities. Hence, this thesis provides a prospective techno-economic assessment (TEA) of today's most promising and mature PtG technologies to estimate their short- and long-term competitiveness, allowing for the identification of the required measures.

To estimate the development of technology costs, economies of scale were considered by implementing a disaggregated experience curve model. This model allows for an effective assessment of scaling effects over all investigated technologies. In addition, it shows the importance of considering spillover learning effects between technologies to avoid overestimating the individual effects of technological learning. According to these investigations, the technology costs for PtG applications are expected to decrease by 30–75 % solely through technological learning induced by the non-energetic industrial demand for hydrogen by 2050. With the additional consideration of increasing system scales above 50 MW, overall cost reductions for all technologies are calculated with >75 %. Consequently, the product generation costs for hydrogen and SNG from PtG were found to decrease significantly for corresponding large-scale implementations. Depending on the source of electricity, hydrogen production costs are evaluated to reach values well below $100\,€/\mathrm{MWh}_{\mathrm{H_2}}$ in the long term. Owing to the additional efforts required for the methanation process, the identified general production costs for SNG relate to approximately $150\,€/\mathrm{MWh}_{\mathrm{SNG}}$. However, in that context, the elaborated assessments show that significantly better performance can be achieved if synergistic effects between the processes are appropriately utilized. Therefore, an integrated system within an industrial application scenario can achieve an effective product cost of $<50\,€/\mathrm{MWh}_{\mathrm{SNG}}$. Furthermore, studies have shown that the competitiveness of PtG is widely affected by its consideration as an integral part of future energy systems, and thus, its capabilities regarding sector coupling. The utilization of byproducts, namely oxygen and waste heat, not only contributes to the economic viability of the process but can also have a significant impact on systemic energy efficiency by reducing diverse supply efforts.

Finally, the elaborated assessment methods and performed analysis also represent a generic outline of the capabilities of prospective techno-economic methods to identify the potential of early-stage technologies to contribute to the energy transition. Therefore, these methods allow for early identification of the technical and economic risks involved, as well as potential bottlenecks regarding resource and demand potentials, thus enabling the establishment of effective measures.

Kurzfassung

Die zur Erreichung der im Übereinkommen von Paris festgelegten Klimaziele notwendigen Maßnahmen und die damit verbundene Energiewende erfordern den Einsatz neuer Energietechnologien. Weiters erfordert das für diese Zielerreichung verbleibende CO_2-Budget eine rasche und effektive Umsetzung entsprechender Maßnahmen. Daher müssen politische und unternehmerische Entscheidungsträger auf nachvollziehbare Bewertungen zurückgreifen, um die ökonomische Eignung, aber auch die Effektivität hinsichtlich einer Eindämmung des Klimawandels, dieser neuen Technologien einschätzen zu können. Nur so können Förder- und Regulierungsmaßnahmen gesetzt bzw. entsprechende Investitionsentscheidungen zugunsten dieser Technologien getroffen werden.

In Bezug auf die Energiewende ist, abgesehen von einer weitgehenden Elektrifizierung, der Bedarf für erneuerbare Gase, wie Wasserstoff oder Erdgassubstitute (SNG), als hoch eingeschätzt. In diesem Zusammenhang wird daher Power-to-Gas (PtG) als ein wesentlicher Bestandteil zukünftiger erneuerbarer und nachhaltiger Energiesysteme angesehen. Allerdings befinden sich die damit verbundenen Technologien noch in einem relativ frühen Technologiestadium, insbesondere hinsichtlich einer breiten Implementierung im industriellen Maßstab. Dies führt einerseits dazu, dass diesbezügliche Umsetzungen nur zögerlich erfolgen. Andererseits erfordert ein Wasserstoffbedarf von >1500 TWh/a, wie er für die EU identifiziert wurde, eine frühe und schnelle Erweiterung entsprechender Kapazitäten. Aus diesem Umstand bildet die vorliegende Dissertation eine prospektive techno-ökonomische Bewertung der aktuell vielversprechendsten und ausgereiftesten PtG-Technologien, um deren kurz- und langfristige Wettbewerbsfähigkeit abzuschätzen und die Identifizierung geeigneter Maßnahmen zu ermöglichen.

Um die zukünftige Entwicklung von Technologiekosten abschätzen zu können, wurden Skaleneffekte mithilfe eines disaggregierten Modells für Lernkurven analysiert. Dieses Modell erlaubt eine effektive Bewertung dieser Skaleneffekte über alle betrachteten Technologien hinweg und zeigt insbesondere die Wichtigkeit der Berücksichtigung von technologieübergreifenden Lerneffekten, die ansonsten zu einer Überschätzung von individuellen Effekten von technologischem Lernen führen können. Den durchgeführten Analysen zufolge, wird für PtG-Technologien eine Reduktion der kapital-bezogenen Kosten um 30–75 % erwartet, alleine durch Lernkurveneffekte ausgelöst vom Einsatz der Technologien zur Deckung des nicht-energetischen industriellen Wasserstoffbedarfs bis 2050. Bei einer zusätzlichen Berücksichtigung steigender individueller Anlagenkapazitäten >50 MW wurden die gesamten Kostenreduktionen als >75 % ermittelt. Daraus lässt sich auch eine signifikante Senkung der Produktgestehungskosten für Wasserstoff und SNG aus PtG-Prozessen ableiten. Abhängig von der betrachteten Quelle für die elektrische Strombereitstellung lassen sich die zu erwartenden Wasserstoffgestehungskosten in der Langzeitbetrachtung mit deutlich unter 100 €/$\mathrm{MWh}_{\mathrm{H_2}}$ abschätzen. Hinsichtlich SNG führt der zusätzliche Aufwand der Methanisierung zu Gestehungskosten im Bereich von 150 €/$\mathrm{MWh}_{\mathrm{SNG}}$. Allerdings haben die durchgeführten Studien auch gezeigt, dass eine entsprechende Nutzung von Synergieeffekten zwischen den einzelnen Prozessen zu einer deutlichen Steigerung der techno-ökonomischen Gesamteffizienz der Technologien führen kann. So kann ein hochintegriertes System in einer industriellen Anwendung potenziell effektive Gestehungskosten von <50 €/MWh$_{\mathrm{SNG}}$ erzielen. Des Weiteren zeigen die durchgeführten Analysen, dass die Wettbewerbsfähigkeit von PtG-Prozessen auch weitgehend von deren Betrachtung als integraler Bestandteil zukünftiger Energiesysteme und Einsatzmöglichkeiten im Sinne der Sektorkopplung abhängig ist. Die Verwertung von Nebenprodukten, wie Sauerstoff oder Abwärme, dient dabei nicht nur der Steigerung der individuellen Wirtschaftlichkeit der Anlage, sondern stellt auch einen wesentlichen Einfluss auf die Gesamteffizienz im Energiesystem dar durch die Verminderung separater Aufwände, um diese zur Verfügung zu stellen.

Zu guter Letzt stellen die eingesetzten Bewertungsmethoden und durchgeführten Analysen auch einen generischen Überblick dar, welche Möglichkeiten von prospektiven techno-ökonomischen Methoden zur Identifizierung der Potenziale von neuen Technologien hinsichtlich deren Beitrag zur Energiewende bestehen. Dabei erlauben sie eine frühe Identifikation der mit ihnen verbundenen technischen und ökonomischen Risiken sowie potenzieller Einschränkungen hinsichtlich Ressourcen- und Bedarfspotenziale und erlauben damit entsprechend effektive Maßnahmen zu setzen.

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Abbreviations

AEC	alkaline electrolysis cell.
AEM	anion exchange membrane.
AFC	alkaline fuel cell.
BF	blast furnace.
BoP	balance of plant.
CAAGR	compound average annual growth rate.
CAPEX	capital expenditures.
CAS	cryogenic air separation.
CCU	carbon capture and utilization.
CHP	combined heat and power.
CS	crude steel.
CSTR	continuous-stirred tank reactor.
DAC	direct air capture.
DH	district heating.
DRI	direct reduced iron.
EC	European Commission.
ETS	Emissions Trading System.
EU	European Union.
FC	fuel cell.
FLH	full-load hours.
GHG	greenhouse gas.
H2020	Horizon 2020.
HHV	higher heating value.
HVC	high value chemicals.
IEA	International Energy Agency.
LCoE	levelized costs of energy.
LCoH	levelized costs of hydrogen.
LCoP	levelized costs of product.
LHV	lower heating value.
LR	learning rate.
MFR	methane formation rate.
NASA	National Aeronautics and Space Administration.
NECP	national energy and climate plan.
OFLC	one-factor learning curve.
OPEX	operational expenditures.
PEM	proton exchange membrane.
PEMEC	PEM electrolysis cell.
PEMFC	PEM fuel cell.
PR	progress ratio.
PtG	power-to-gas.
PtH_2	power-to-hydrogen.
PtM	power-to-methane.
PtX	power-to-x.
PV	photovoltaics.
RES	renewable energy sources.
RWGS	reverse-water-gas-shift.
SMR	steam methane reforming.
SNG	synthetic natural gas.
SOEC	solid oxide electrolysis cell.
SOFC	solid oxide fuel cell.
SWOT	strengths, weaknesses, opportunities, threats.
TEA	techno-economic assessment.
TFLC	two-factor learning curve.
TRL	technology readiness level.

Glossary

closed carbon cycle A self-contained system in which carbon emissions do not exit the defined system boundaries and are re-utilized or stored within them.

co-electrolysis Simultaneous reduction of CO_2 alongside with H_2O in the electrolysis process.

economies of scale The collective term for reduction of technology costs through technological learning and upscaling.

EU-27 The European Union consisting of its 27 member states as per the year 2021.

experience curve The representation of technological learning as the correlation between overall production costs and underlying cumulative production volume. See also learning curve.

high value chemicals All ethylene and associated by-products, other than hydrogen, that result from the ethylene production process, including propylene, butadiene and benzene, without limitation.

Horizon 2020 An EU Research and Innovation Programme (2014–2020) and the financial instrument implementing the Innovation Union.

learning curve The representation of technological learning as the correlation between labor costs and the underlying cumulative production volume. It is partially used synonymously with experience curve.

learning investment The cumulative investments required to promote the experience curve of an entrant technology until cost parity with incumbent competitors is reached.

national energy and climate plan Introduced by EU as a part of the *Clean energy for all Europeans* package and outline how EU countries intend to address energy efficiency, renewables, greenhouse gas emissions reductions, interconnections, and research and innovation.

power-to-gas The production of gaseous energy carriers (usually H_2 or SNG) from electric power through electrocatalytic processes (e.g., water electrolysis).

power-to-hydrogen The production of hydrogen from electric power through electrolysis.

power-to-methane The aggregated process of producing methane (or SNG) using electrolysis and methanation technologies.

power-to-x The production of chemicals (e.g., hydrocarbons) directly from electrochemical processes or based on hydrogen produced via electrolysis.

spillover effects With relation to technological learning, spillover effects describe learning effects that are not directly related to the experience gained from the production of the investigated technology itself but from concurrent processes that induce indirect learning effects.

techno-economic assessment A comprehensive method of analyzing the technological characteristics and economic performance of an industrial process or a technology and its products.

technological learning The change in technology costs, more precisely technology production costs, by experience gained through increasing cumulative production volume. This is also described as *economies of manufacturing scale*.

upscaling The reduction in technology costs by simplification and size effects through increasing capacities per installed unit. Therefore, it is also described as *economies of unit scale*.

1 Introduction

Limiting the effects of climate change and keeping global warming well below 2 °C, preferably to 1.5 °C, compared to pre-industrial levels, as outlined and committed to in the Paris Agreement [1], is one of the major global challenges today. Therefore, numerous national and international climate targets and actions have been proposed to significantly reduce greenhouse gas (GHG) emissions and reach climate neutrality by the mid-century. In this context, after setting a target to reduce GHG emissions by 80–95 % until 2050 in 2009, the European Union (EU) also aimed to be climate neutral by 2050, as stated in their *strategic long-term vision for a prosperous, modern, competitive and climate neutral economy* [2]. Along with this 2050 long-term strategy, the European Commission (EC) agreed on a climate and energy framework for 2030 with the following key targets:

- $\geq$40 % cuts in greenhouse gas emissions (from 1990 levels)
- $\geq$32 % share for renewable energy
- $\geq$32.5 % improvement in energy efficiency

Meanwhile, the target of GHG emission cuts has already been adjusted and raised to at least 55 % compared to the 1990 levels as part of the Green Deal [3].

Looking at the distribution of GHG emissions in the EU-27 across sectors, with about 24 % caused by energy industries, 22 % by transport, and 21 % by the industry sector (energetic and non-energetic use) [4] representing the three main source sectors of GHG emissions, it becomes obvious that achieving these climate targets is a cross-sectoral challenge. At the same time, it shows that even though a massive expansion of renewable electricity production is inevitable and the electrification of certain sectors and applications is consequential from an efficiency perspective, the reduction of CO_2 emissions affects processes that still depend on specific energy carriers or feedstock materials, which at present are fossil-based and require a climate neutral replacement. In this context, H_2 from renewable sources has emerged as a potential carbon-free alternative in many sectors and applications.

Hydrogen is also applicable as an alternative fuel in combustion engines or fuel cells – be it for stationary or mobile applications – and a major demand for renewable H_2 is seen in industrial use. First, there is the production of iron and steel, where the direct reduced iron (DRI) route for producing iron sponge from iron ore may be a successor to the current coke-based reduction process. Second, there is the chemical sector, where the vast majority of today's H_2 demand is utilized for fertilizer production, and an additional H_2 demand is expected as a feedstock material for currently fossil-based base and high value chemicals (HVC) [5]. Moreover, the generation of higher-value hydrocarbons with long usage periods and appropriate recycling mechanisms can provide potential carbon sinks and the long-term fixation of carbon emissions as part of a carbon capture and utilization (CCU) process. In this regard, closed carbon cycles, as part of circular economies, act as an additional measure towards climate neutrality.

In addition to the direct energy or feedstock use of hydrogen, its production from renewable electricity sources, such as wind or photovoltaics (PV), offers additional opportunities in combination with these intermittent energy sources. Gaseous or even liquified forms of hydrogen as an energy carrier provide high energy densities with appropriate advantages compared to more efficient storage types, such as batteries, when it comes to the transportation of renewable energy over extended distances or seasonal storage to temporally decouple the energy supply and demand (see Figure 1.1).

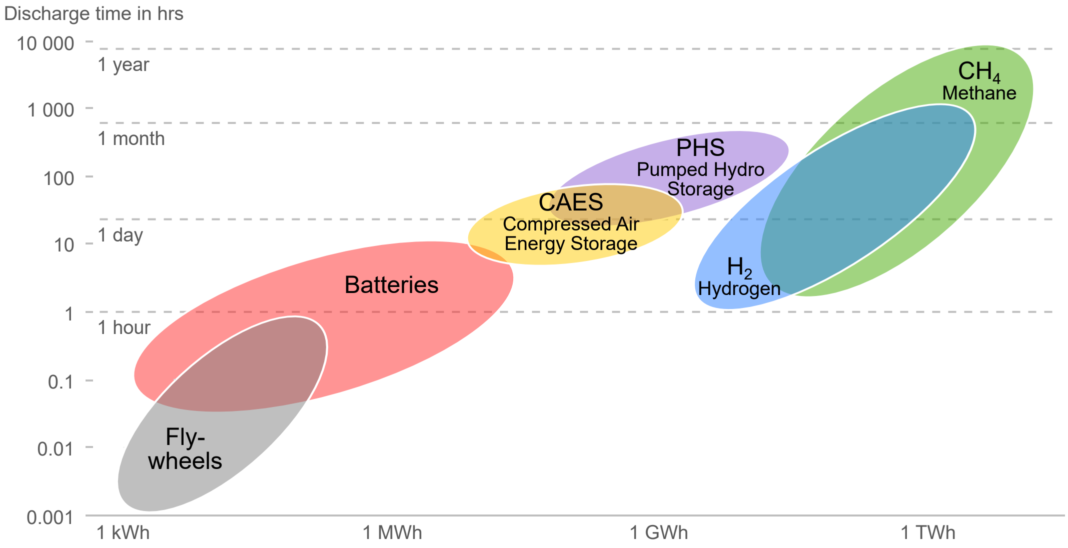

Figure 1.1: Storage capacity of different energy storage applications. Based on Teske *et al.* [6]

This projected importance of hydrogen has led to a significant number of national hydrogen strategies for governments worldwide [7]. In addition, the EU outlined a hydrogen roadmap in 2020, targeting an installed electrolysis capacity of 6 GW_{el} in 2024 and 40 GW_{el} in 2030 within the EU borders. An additional capacity of 40 GW_{el} outside the EU is planned until 2030 to provide the appropriate import capacities [8]. However, simulations of the EU energy systems for a 95 % reduction target revealed demands for electrolysis capacities from 70 to 1000 GW_{el} [9, 10], depending on the permission to store fossil CO_2, presumably making additional efforts within the EU necessary. This can also be deduced from the 1.5 °C scenario in the EU strategic long-term vision [2], leading to a hydrogen demand of $>$1500 TWh a^{-1}, and thus electricity-based production capacities of more than 615 GW_{el}, as discussed by Lux and Pfluger [11] more recently.

It becomes apparent that the efforts that have to be made to reach these ambitious goals are tremendous. Concurrently, actions must be taken early to allow for the consistent implementation of technologies and measures, rather than making disruptive changes that occur if relevant decisions are delayed (see also Figure 1.2). Therefore, policy- and decision-makers must rely on robust analyses and projections to identify relevant technologies concerning their long-term feasibility according to the defined objectives. Thus, the prospective techno-economic assessment (TEA) of energy technologies for individual applications, but even more so in a holistic and cross-sectoral manner, is an important measure for the effective implementation of climate change mitigation strategies.

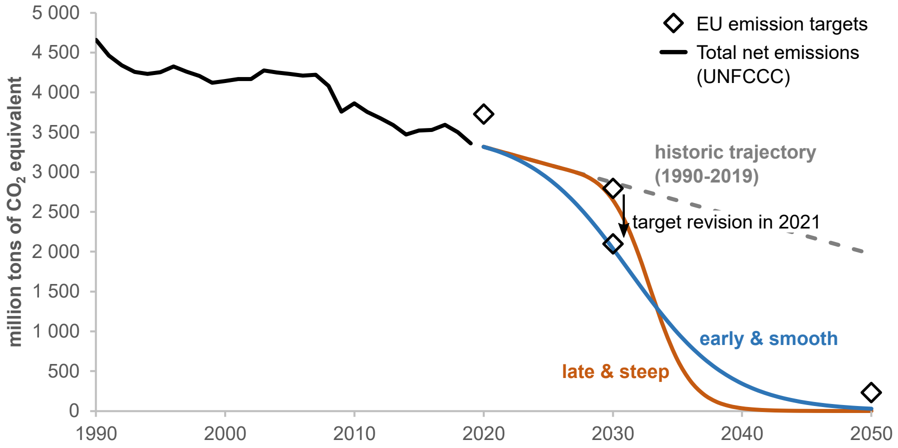

Figure 1.2: Potential reduction pathways for EU-27 emissions in relation to historical trends. Based on Victoria *et al.* [12].

Emission data from EEA [13]. Transition pathways are based on the remaining CO_{2eq} budget of 700 Gt to avoid human-induced global warming above 1.7 °C with a probability of >66 % [14], using a population-based share (6 %) for the EU [15].

This thesis examines the fast and effective transition toward the set climate goals based on the importance of TEA and the expected role played by power-to-gas (PtG) in that context. Therefore, it outlines generic methods for the prospective techno-economic evaluation of emerging energy technologies, using PtG technologies and their applications as a representative example. This includes the implementation of a disaggregated cost model to evaluate the scaling effects induced by technological upscaling in terms of produced units and individual unit scales, as well as the identification of current and future synergy potentials from an overall systemic perspective. Furthermore, the boundary conditions for the economic competitiveness of PtG products as renewable energy carriers were assessed for different application scenarios in mid- and long-term scenarios. Finally, this study aims to provide a prospective evaluation of the potential future role of PtG technologies as part of de-fossilized future energy systems and the corresponding costs involved. It is, thus, meant to reveal appropriate measures that have to be taken – regarding early and total learning investments, but also overall resource efficiency and product utilization – to implement renewable hydrogen and synthetic natural gas (SNG) as a cornerstone of the energy transition.

2 Research and thesis approach

Restricting the anthropogenic greenhouse effect and the associated reduction in emissions that damage the climate is an important aspect of the current climate and energy politics. However, the reduction or avoidance of fossil energy carriers is not limited to the transition of electricity generation and supply to renewable sources, such as wind and solar power. Instead, it implies other forms of energy supply, energy storage and transport, and the provision of renewable feedstock for the industry. Consequently, this has resulted in the development of various new energy technologies that serve the different sectors of such a transformation to a widely renewable energy system.

Owing to the timeframe defined for fulfilling the set climate targets, the required technologies are temporally restricted from becoming established in their respective fields of application, at least when compared to the extent available for state of the art and maximally efficient mature processes. Thus, it is necessary to identify and evaluate the suitability of these new technologies for the intended application fields at an early stage of development. In this context, *technical feasibility* is as important as *long-term economic viability*.

2.1 Research objective

To assess *technical feasibility*, it is important to identify technical risks, options for integration in existing process chains, and utilizable synergies already on a low technology readiness level (TRL). In addition, in an energy system completely based on renewable energy sources (RES), the attainable total efficiency, both energetic and exergetic, plays a decisive role in choosing suitable energy carriers, transformation technologies, and processes. In this regard, the variety of different strategies and development paths, in combination with low TRL, makes investment decisions significantly more difficult compared to established competitive applications. Thus, to ensure *long-term economic viability*, identifying relevant value chains is crucial for the selection of novel technologies. Therefore, the required investment costs and appropriate product generation costs need to be estimated by considering the overall process. This applies to current as well as future costs, considering the learning curve and scaling effects, and allows for a mid- and long-term evaluation of the economic competitiveness of the technology.

This thesis' main objective is to address the issues outlined by the development and application of generic methods for the TEA of different technologies and their applications for the transformation and storage of renewable energy carriers. Therefore, the following three research targets were defined:

- Analysis of technological developments and description of potential process chains and transformation processes.
- Evaluation of future cost developments involved with technology roll-out and upscaling to industry-relevant scales.

- Application of the developed assessment tools based on cross-sectoral usage applications for a quantitative characterization.

While these objectives are elaborated with a close focus on hydrogen and PtG technologies within this thesis, the corresponding methods are aimed at being applicable to all types of technologies, processes, and applications related to energy storage and transition.

2.2 Relevant publications

To reach the outlined research targets, this study used a cumulative approach. The individual stages are addressed in consecutive research topics that cover the entire evaluation procedure – from the definition and application of a projection model, evaluation of resource and demand potentials, to the final techno-economic process assessment for a relevant application. Therefore, this thesis is based on the following peer-reviewed articles published in international journals.

(i) H. Böhm, S. Goers, and A. Zauner, “Estimating future costs of power-to-gas – a component-based approach for technological learning”, *International Journal of Hydrogen Energy*, vol. 44, no. 59, pp. 30 789–30 805, 2019. DOI: `10.1016/j.ijhydene.2019.09.230`

(ii) H. Böhm, A. Zauner, D. C. Rosenfeld, and R. Tichler, “Projecting cost development for future large-scale power-to-gas implementations by scaling effects”, *Applied Energy*, vol. 264, p. 114 780, 2020. DOI: `10.1016/j.apenergy.2020.114780`

(iii) V. Rodin, J. Lindorfer, H. Böhm, and L. Vieira, “Assessing the potential of carbon dioxide valorisation in Europe with focus on biogenic CO_2”, *Journal of CO2 Utilization*, vol. 41, p. 101 219, 2020. DOI: `10.1016/j.jcou.2020.101219`

(iv) H. Böhm, S. Moser, S. Puschnigg, and A. Zauner, “Power-to-hydrogen & district heating: Technology-based and infrastructure-oriented analysis of (future) sector coupling potentials”, *International Journal of Hydrogen Energy*, vol. 46, no. 63, pp. 31 938–31 951, 2021. DOI: `10.1016/j.ijhydene.2021.06.233`

(v) H. Böhm, M. Lehner, and T. Kienberger, “Techno-economic assessment of thermally integrated co-electrolysis and methanation for industrial closed carbon cycles”, *Frontiers in Sustainability*, vol. 2, p. 77, 2021. DOI: `10.3389/frsus.2021.726332`

As illustrated in Figure 2.1, even though they have been elaborated widely independently and with different focus, these articles represent a coherent structure within this thesis. Therefore, article (i) includes the general definition and characterization of the disaggregated learning curve model used for the evaluation of future technology cost developments. Article (ii) expands this learning curve model by component-related unit-scaling effects, and applies the model to power-to-gas applications for a prospective evaluation of cost reduction potentials for large-scale implementations. The elaborations in article (iii) provide a general analysis of CO_2 utilization potentials from different industrial and biogenic sources, along with costs and volumes involved. The obtained results serve as an input for the techno-economic assessments performed in article (v), which also utilizes the models defined in (i) and (ii) to evaluate the cost aspects of integrated power-to-gas systems in industrial scenarios. Furthermore, article (iv) presents an overarching analysis of potential synergies of power-to-hydrogen (PtH_2) technologies – with a special focus on district heating – to point out the potential implications of a broad implementation of PtG technologies as part of the energy transition, thereby impacting its techno-economic performance from a systemic perspective.

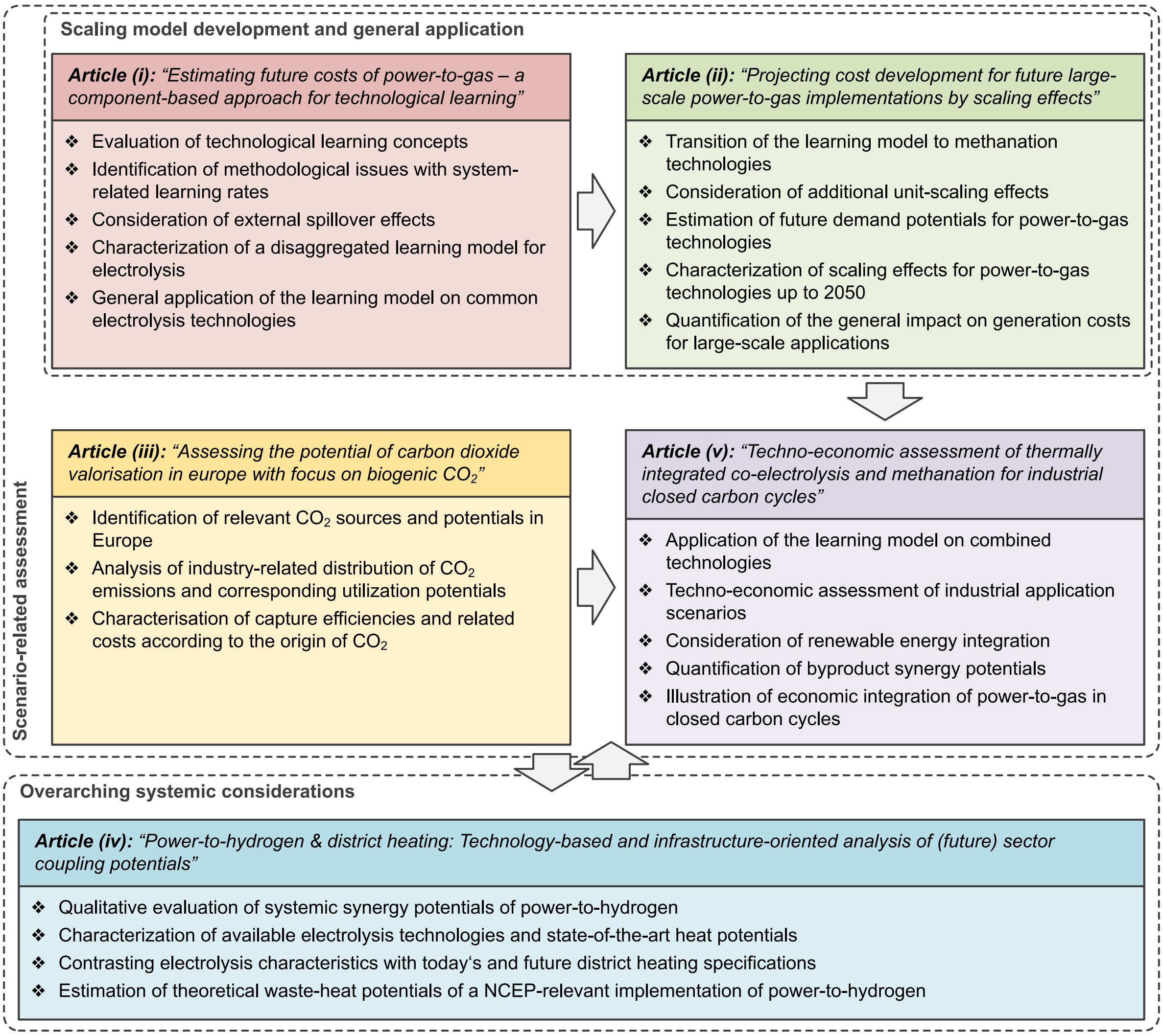

Figure 2.1: Illustration of the main contents and relations between the articles included in the thesis

2.2.1 Author contributions

Regarding the main publications, I was the principal author for the articles (i), (ii), and (v), thus, being responsible for the conceptualization and methodology of the paper. Paper (iii) was mainly authored by *Valerie Rodin*, where I contributed to the collection and preparation of data on fossil CO_2 sources as well as data on the efficiency and costs of CO_2 capture. In addition, I share the main authorship for the article (iv) with *Simon Moser*, whose contribution was focused on the elicitation of expert and stakeholder knowledge, while I mainly provided an evaluation of PtH_2 technology status and theoretical waste heat potential. *Stefan Puschnigg* contributed with an initial literature review and provided review and editing work together with *Andreas Zauner*. *Andreas Zauner* also contributed significantly by collecting economic data and validating the results of the project tasks and deliverables that led to the articles (i) and (ii). *Sebastian Goers* mainly outlined the theoretical concept of technological learning in paper (i). *Daniel C. Rosenfeld* contributed with data for the economic database to validate the learning curve and scaling model, as well as editing the paper (ii). *Markus Lehner* was a project partner in the project leading to the results in article (v) and reviewed and edited the manuscript together with *Thomas Kienberger* prior to submission. *Johannes Lindorfer* and *Robert Tichler* acted as project leaders for the projects that led to the results discussed in the individual articles. *Luciana Vieira* contributed as an overall project manager to the project behind article (iii). Table 2.1 shows the percentage of own contributions to the main activities for the given articles.

Table 2.1: Overview of own contribution shares to the individual journal articles and activities

	Article (i)	Article (ii)	Article (iii)	Article (iv)	Article (v)
Conceptualization	90%	85%	25%	40%	100%
Methodology	100%	80%	15%	50%	100%
Validation	90%	75%	35%	70%	100%
Investigation	85%	70%	30%	50%	100%
Writing	85%	90%	25%	50%	100%
Review & Editing	75%	75%	25%	75%	90%

2.2.2 Additional related publications

Beyond the aforementioned main articles, the following scientific works with methodological or thematic relevance to this thesis have been published (ordered chronologically acc. to date of publication).

(vi) H. Böhm and J. Lindorfer, “Techno-economic assessment of seasonal heat storage in district heating with thermochemical materials”, *Energy*, vol. 179, pp. 1246–1264, 2019. DOI: `10.1016/j.energy.2019.04.177`

(vii) J. Lindorfer, D. C. Rosenfeld, and H. Böhm, “23 – Fuel Cells: Energy conversion technology”, in *Future Energy (Third Edition)*, T. M. Letcher, Ed., Third Edition, Elsevier, 2020, pp. 495–517, ISBN: 978-0-08-102886-5. DOI: `10.1016/B978-0-08-102886-5.00023-2`

(viii) F. Graf, M. Heneka, A. Zauner, H. Böhm, R. Tichler, J. Cohen, C. Friedl, and J. Reichl, “Ökologische, wirtschaftliche und sozio-ökonomische Bewertung von PtG-Technologien”, in *Power-to-Gas*, ser. Edition gwf Gas + Energie, F. Graf, R. Schoof, and M. Zdrallek, Eds., Essen: Vulkan Verlag, 2020, ISBN: 978-3-8356-7445-5

(ix) D. C. Rosenfeld, H. Böhm, J. Lindorfer, and M. Lehner, “Scenario analysis of implementing a power-to-gas and biomass gasification system in an integrated steel plant: A techno-economic and environmental study”, *Renewable Energy*, vol. 147, pp. 1511–1524, 2020. DOI: `10.1016/j.renene.2019.09.053`

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3 Background and state of the art

The following sections outline the current state of the art of PtG technologies with regard to available and common technologies, performance, and cost. Furthermore, common methods for the techno-economic assessment of energy technologies are discussed.

3.1 Power-to-gas

In general, the term power-to-gas (PtG) describes the aggregate of processes that produce gaseous energy carriers from electric power. Hence, this includes the mere production of hydrogen by water (or steam) electrolysis, also known as power-to-hydrogen (PtH$_2$), and the production of synthesis gas, by for example, using co-electrolysis of H_2O and CO_2, as well as the downstream synthesis of the electrolysis product to methane or SNG using methanation. The complete route from electrolysis to methanation is often referred to as power-to-methane (PtM). Even though electrolysis, at least for certain technologies, is considered mature and commercially available, PtM as an integrated system approach still represents a process with additional research potential and which has only seen single commercial implementations [16].

Apart from PtG or PtM, the production of higher-value hydrocarbons from electrochemical processes is summarized by the more general term power-to-x (PtX). However, the following sections focus on the elaboration of electrolysis and methanation technologies.

3.1.1 Electrolysis technology status

The core technology of the PtG process, as defined today, is the electrolysis cell used to split water (or steam) into hydrogen and oxygen.

$$\mathrm{H_2O} \longrightarrow \mathrm{H_2} + \frac{1}{2}\,\mathrm{O_2} \qquad \Delta H_r^\circ = 285.83\,\frac{\mathrm{kJ}}{\mathrm{mol}} \tag{3.1}$$

Although the total reaction (3.1) applies to all water electrolysis processes, the partial reactions for the process are dependent on the individual technology. Currently, three main electrolysis cell types are available and used for PtG: (i) alkaline, (ii) proton exchange membrane (PEM), and (iii) solid oxide electrolysis cells (see Figure 3.1). The former two, alkaline and PEM, are usually operated with liquid water at temperatures below 100 °C and are referred to as *low-temperature electrolysis*. In contrast, solid oxide electrolysis cell (SOEC) is often referred to as *high-temperature electrolysis* because it is usually fed with steam and operated at temperatures of 650–900 °C. The main characteristics of these three technologies are summarized in Table 3.1 and described in more detail below.

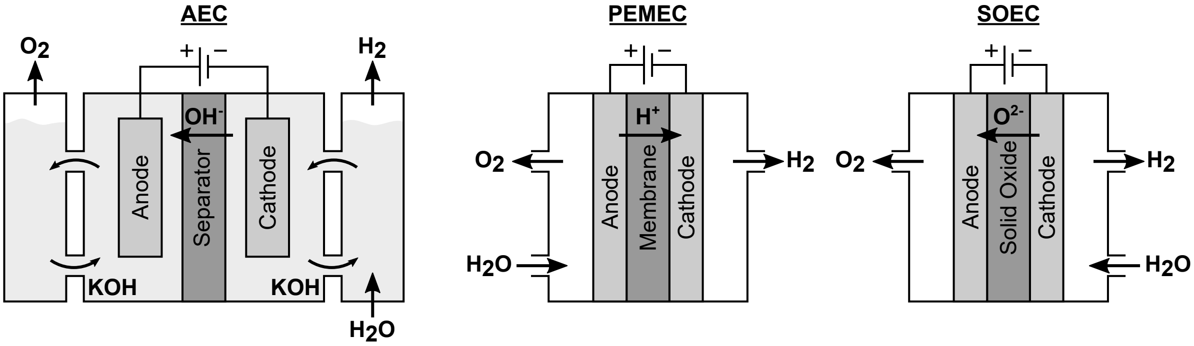

Figure 3.1: Schematic illustration of the three common water electrolysis cell types. Based on Steinmüller *et al.* [17]

Table 3.1: Key characteristics of state-of-the-art water electrolysis technologies. Source: Böhm *et al.* [18]

Technology	Alkaline (AEC)	Proton exchange membrane (PEMEC)	Solid oxide (SOEC)
Technology status	Commercial (mature) TRL 9	Commercial (mature) TRL 8–9	Pre-commercial (R&D) TRL 5–6
Operating temperature	60–90 °C	50–80 °C	650–900 °C
Typical stack capacity	<10 MW	<5 MW	<100 kW
Electric efficiency (LHV)	63–71 %	60–68 %	100 %[a]
Load flexibility	20–100 %	0–100 %	−100 % / +100 %
Cold start-up time	1–2 h	5–10 min	Hours
Warm start-up time	1–5 min	<10 s	15 min

[a] Operation at thermoneutral voltage

3.1.1.1 Alkaline electrolysis

The alkaline electrolysis cell (AEC) is the most mature among the described electrolysis technologies and has been used on an industrial scale since the early 1900s. As shown in Figure 3.1, an alkaline cell consists of a cathode and an anode area, which are filled with an electrolyte and separated by a diaphragm. The electrolyte commonly used in AEC is a 25–30 % solution of caustic potash (KOH) that limits the operating temperature to 60–90 °C. Electrodes usually consist of a nickel-based anode and an activated cathode [19]. Because it is well established and does not require any noble catalyst material, the investment costs (CAPEX) are relatively low compared to other technologies. However, current research focuses on increasing the current densities and operating pressures to improve the future performance of the AEC over concurrent technologies [19, 20]. The electrolytic process in AEC can be described by the following half-reactions at the electrodes [21]:

Anode reaction: $2\,\mathrm{OH^-} \longrightarrow \frac{1}{2}\,\mathrm{O_2} + \mathrm{H_2O} + 2\,\mathrm{e^-}$

Cathode reaction: $2\,\mathrm{H_2O} + 2\,\mathrm{e^-} \longrightarrow \mathrm{H_2} + 2\,\mathrm{OH^-}$

3.1.1.2 Proton exchange membrane electrolysis

PEM electrolysis, also described as polymer electrolyte membrane electrolysis, is currently the most used water electrolysis technology besides AEC. In comparison, PEM electrolysis cell (PEMEC) provides higher cell efficiencies and power densities. Furthermore, it allows for more flexible operating conditions and fast start-up times ranging from minutes (cold start) to seconds (warm start) [19]. This is a major advantage in grid service operations, such as the storage of electricity surpluses. However, the available system capacities are still lower than those of AEC. This and the use of expensive catalysts, such as platinum and iridium, and fluorinated membrane materials, along with increased system complexity due to high-pressure operation and required water purity, result in higher specific system costs. Hence, the optimization of material usage, reduction of system complexity, and scale-up of the technology are the current main topics in PEM electrolysis research activities [20]. While improvements in these areas would lead to a reduction of system CAPEX, another challenge for the reduction of hydrogen production costs is the increase of stack lifetimes to reduce replacement and maintenance costs.

As can be seen in Figure 3.1, the anode and cathode sides of PEMEC are separated by a proton-conducting membrane, which also functions as an electrolyte. Consequently, the cell operates according to the following partial reactions [22]:

Anode reaction: $2\,\mathrm{H_2O} \longrightarrow 4\,\mathrm{H^+} + \mathrm{O_2} + 4\,\mathrm{e^-}$

Cathode reaction: $4\,\mathrm{H^+} + 4\,\mathrm{e^-} \longrightarrow 2\,\mathrm{H_2}$

Currently, the polymer membrane, commonly a perfluorosulfonic acid polymer (e.g., Nafion™), represents a weak point in this technology. While the concept itself would allow for operations at elevated pressure and thus reduce downstream compression efforts, this currently leads to significantly reduced lifetimes of the membranes due to alternating pressure ratios, especially at start-stop cycles. Therefore, the common operating conditions for PEMEC systems are pressure levels up to 50 bar and temperatures of 50–80 °C [19].

3.1.1.3 Solid oxide electrolysis

The solid oxide electrolysis cell (SOEC) is operated at temperatures of 650–900 °C, and is thus categorized as high-temperature electrolysis. It uses solid ion-conducting ceramics as electrolytes (e.g., yttria stabilized zirconia) and does not rely on noble catalytic materials (see Figure 3.1). Although the high-temperature levels imply high demands on the used cell materials, they also allow SOEC to be supplied with steam instead of water, which reduces the total energy demand by the heat of evaporation ΔH_{evap} (see Figure 3.2). Thus, the required energy demand can be partially supplied in the form of heat instead of electricity, thereby increasing the electrical efficiency of the cell. Because SOEC is the least mature cell type under those discussed herein, current R&D tasks focus on materials that increase cell stability and lifetime, as well as scaling up the technology to commercial levels [20, 23].

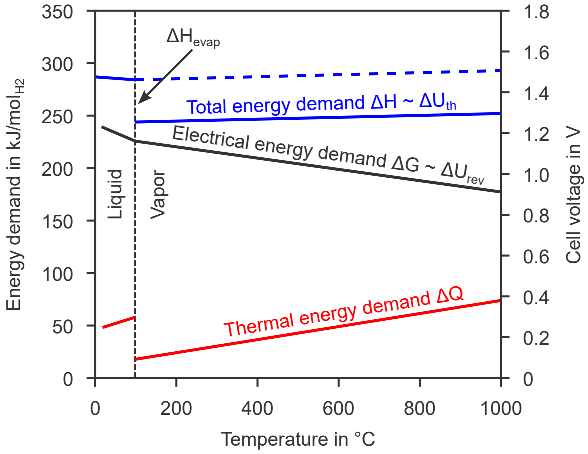

Figure 3.2: Total (ΔH), thermal (ΔQ), and electrical (ΔG) energy demand of an ideal electrolysis cell as function of the temperature. Source: Böhm *et al.* [18]

Steam electrolysis In relation to PtG, SOEC may be considered in different types of operations, either producing hydrogen from H_2O or being fed with H_2O and CO_2 to produce a synthesis gas (syngas). In the commonly used case for sole hydrogen production, as described for AEC and PEMEC, SOEC is fed with H_2O in the form of steam and produces H_2 and O_2 according to the following partial reactions:

$$\text{Anode reaction:} \qquad O^{2-} \longrightarrow \frac{1}{2}\,O_2 + 2\,e^-$$

$$\text{Cathode reaction:} \qquad H_2O + 2\,e^- \longrightarrow H_2 + O^{2-}$$

Co-electrolysis However, the high operating temperatures of SOEC also favor a reduction of CO_2 to CO and O_2 alongside steam, which is indicated as co-electrolysis. The partial reactions at the electrodes are identical to the steam electrolysis, apart from the additional reduction of CO_2 at the cathode [24]:

$$\text{Anode reaction:} \qquad 2\,O^{2-} \longrightarrow O_2 + 4\,e^-$$

$$\text{Cathode reactions:} \qquad H_2O + 2\,e^- \longrightarrow H_2 + O^{2-}$$

$$CO_2 + 2\,e^- \longrightarrow CO + O^{2-}$$

where the overall reaction and its products are dependent on the input composition:

$$\underbrace{n\,CO_2 + m\,H_2O \longrightarrow n\,CO + m\,H_2}_{\text{cathode side}} + \underbrace{\frac{m+n}{2}\,O_2}_{\text{anode side}} \qquad (3.2)$$

A major advantage of co-electrolysis arises in combination with downstream processes, such as methanation, because the syngas composition can be adapted according to the process requirements [25, 26]. Furthermore, the thermodynamically favorable reduction in the electrolyzer and the potential for the thermal integration of exothermic downstream processes allow for high overall system efficiencies [27, 28].

Recent research has also investigated co-electrolysis of CO_2 and H_2O for alkaline-based anion exchange membrane (AEM) electrolysis cells [29]. However, these technologies are still in their early stages of development [30, 31].

3.1.2 Methanation technology status

The methanation process involves the synthesis of SNG from hydrogen and carbon dioxide, and carbon monoxide, respectively. This synthesis can be performed in either a chemical (catalytic) or biological conversion process. For both pathways, there are different process and reactor concepts known, as shown in Figure 3.3. The basic characteristics of both are summarized in Table 3.2 and described below.

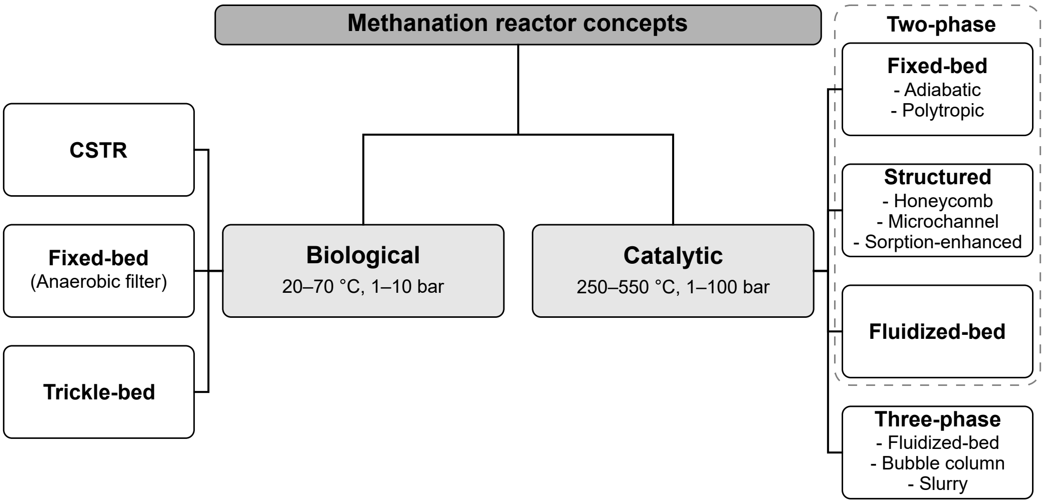

Figure 3.3: Different reactor concepts for methanation. Based on Götz *et al.* [32], Rönsch *et al.* [33], and Lecker *et al.* [34]

3.1.2.1 Chemical methanation

Chemical methanation uses the catalytic conversion processes of H_2 and CO (CO methanation) or CO_2 (CO_2 methanation). It can be differentiated by reactor types, such as structured, fixed-, or fluidized-bed, or three-phase reactors (see also Figure 3.3). The methanation reaction follows the following gross reactions (cf. [32, 33]), depending on the carbon source (CO or CO_2):

Table 3.2: Key characteristics of state-of-the-art methanation processes. Based on information from Götz *et al.* [32], Rönsch *et al.* [33], Lecker *et al.* [34], Grond, Schulze, and Holstein [35], Ghaib and Ben-Fares [36], and Graf, Krajete, and Schmack [37]

Parameter	Chemical methanation	Biological methanation
Operation		
Process temperature	200–700 °C	15 °C[a] to 98 °C[b]
Delivery pressure	<80 bar	>1 bar
GHSV	3000–6000 h^{-1}	<110 h^{-1}
Capacity		
Max. nominal production capacity	<500 MW_{SNG}	<15 MW_{SNG}
Efficiency		
Conversion efficiency	70–85 %	95–100 %
Durability		
Catalyst lifetime	24 kh	-
Availability	85 %	90 %
Tolerance to impurities (e.g., H_2S)	low	high
Flexibility		
Response characteristics[c]	<5 min	seconds
Cold start-up time	hours	minutes

[a] mesophilic methanogens
[b] thermophilic methanogens
[c] deployment time from standby

$$\text{CO methanation:} \quad \mathrm{CO\,(g) + 3\,H_2(g) \longleftrightarrow CH_4(g) + H_2O\,(g)} \quad \Delta H_r^\circ = -206.3\,\frac{\mathrm{kJ}}{\mathrm{mol}} \tag{3.3}$$

$$\text{CO}_2\text{ methanation:} \quad \mathrm{CO_2(g) + 4\,H_2(g) \longleftrightarrow CH_4(g) + 2\,H_2O\,(g)} \quad \Delta H_r^\circ = -165.1\,\frac{\mathrm{kJ}}{\mathrm{mol}} \tag{3.4}$$

The equilibrium of gas reactions can be influenced by the operating pressure; thus, a higher pressure tends to lead to higher methane yields. In contrast, because the reaction is exothermic, an increase in the operating temperature shifts the equilibrium toward the reactant side. Hence, higher temperatures may accelerate the methanation reaction but decrease methane yield [33]. However, both hydrogenation processes are accompanied by several side reactions, such as the reverse-water-gas-shift (RWGS) reaction (3.5). Basically, CO_2 methanation is a combination of CO methanation and the RWGS reaction [32]:

$$\mathrm{CO\,(g) + H_2O\,(g) \longleftrightarrow CO_2(g) + H_2(g)} \qquad \Delta H_r^\circ = -41.2\,\frac{\mathrm{kJ}}{\mathrm{mol}} \tag{3.5}$$

Another relevant side reaction is the Boudouard reaction (3.6), which promotes the formation of solid carbon and can, among other complex reaction paths, lead to the deactivation of the catalyst by covering it with a carbon layer. This is primarily a problem with CO methanation [33].

$$\mathrm{2\,CO\,(g) \longleftrightarrow C\,(s) + CO_2(g)} \qquad \Delta H_r^\circ = -172.5\,\frac{\mathrm{kJ}}{\mathrm{mol}} \tag{3.6}$$

The inactivation of catalysts by carbon or coke deposition as well as by temperature hotspots or catalyst poisons, which are often contained in industrial off-gases considered for synthesis, is a well-known problem in catalytic methanation processes. Thus, appropriate temperature management in the reactor is crucial, not only for the prevention of catalyst deactivation by sintering [38], but also for enhancing methane yields and the process performance [39]. Hence, the reduction in operating temperature [40] and improvement of heat transfer by structured catalyst carriers and micro-structured reactors have been the focus of recent research [33, 41]. Furthermore, current R&D activities investigate potential performance improvements by pressurized multi-stage methanation and intermediate gas compression and recycling [42].

Despite renewed research efforts and several implementations in the context of PtG [43], chemical methanation technology is well established and commercially available. Owing to its usage in coal-to-SNG processes, especially in China, it has also been proven to be effective in large-scale facilities with multi-MW to GW capacities [33]. Today's applications mostly use nickel catalysts, which provide high activity and selectivity for CH_4 while costs are relatively low compared to alternatives such as ruthenium, rhodium, or cobalt [32, 33]. Depending on the reactor technology and process management, operating temperatures of 250–700 °C and pressures of up to 100 bar are state of the art [39].

3.1.2.2 Biological methanation

Although biological methanation can be described by the Sabatier reaction (see Eq. (3.4)), it uses autotrophic hydrogenotrophic methanogens, which act as autocatalysts and perform conversion instead of a chemical catalyst. Since these methanogens are also present in the production of biogas using anaerobic digestion, two potential process implementations become apparent (see also Figure 3.4):

(a) In a combined process with anaerobic digestion, where hydrogen could be directly injected into the digester and utilize the internally produced CO_2 in an *in-situ* process.

(b) In a separate reactor by converting H_2 and CO_2 externally provided at a stoichiometric ratio, in an *ex-situ* process.

While the in-situ process avoids the cost and space requirements for an additional reactor and biogas upgrading equipment [44], methane formation rate (MFR) is limited by the rate of CO_2 production in the digestion process. Therefore, the possible MFR is low at $<0.1\,h^{-1}$. Furthermore, although an almost complete conversion of CO_2 (approx. 99 vol% CH_4) could be achieved using continuous-stirred tank reactor (CSTR) [45], usually a CO_2 content of >20 vol% remains in the product gas, which results in methane concentrations of about 75 vol%. Moreover, the dependence on the digestion process prevents optimization of the methanation process at elevated temperatures and pressures, which is favorable for hydrogenotrophic methanogens [32].

In contrast, an ex-situ process in an external reactor allows for the independent optimization of the process and methane yield. Therefore, methane concentrations of 75–98 % can be achieved in the product gas, which is eligible to replace natural gas, at least for the upper value [32]. Furthermore, a separate reactor capacity is not bound by that of the digester, and it facilitates the use of CO_2 sources apart from biogas production [44], including industrial carbon emissions.

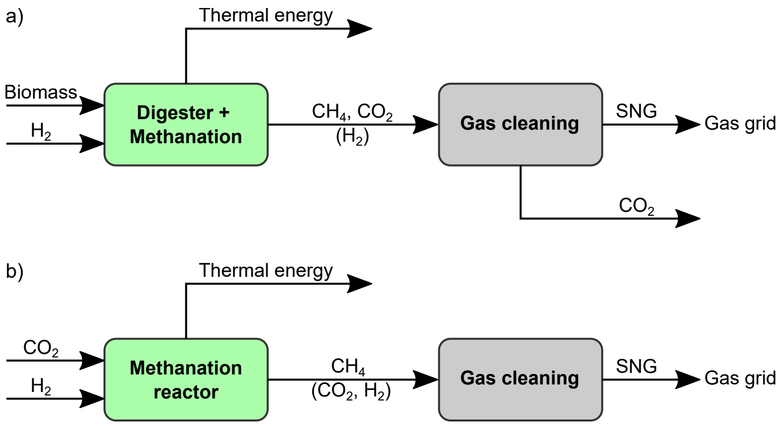

Figure 3.4: Schematic diagram of biological methanation in an (a) in-situ and an (b) ex-situ process

The most limiting factor for MFR is assumed to be the gas-liquid gas transfer, that is, the supply of H_2 to the microorganisms [34, 44]. Therefore, developmental efforts have focused on the specific delivery of hydrogen to methanogens. This may be achieved by optimized reactor concepts, such as trickle-bed reactors, which show higher specific CH_4 production than other configurations [44]. Additionally, the solubility of gases in the liquid is improved by increasing the operating pressure, which also reduces the size of the gas bubbles. Thus, the contact area between the gaseous substrates and methanogens is reduced, while the gas up-flow is slowed down. In addition, the increased partial pressures of the reactants improve the conversion efficiency [32]. Biological methanation reactors can reach conversion efficiencies of >95 % at operating pressures of 1–3 bar [32, 35, 37]. The operating temperatures are mainly dependent on the order of the methanogens used. The optimal temperature for microorganisms activity ranges between 15 °C for mesophilic methanogens and 98 °C for thermophilic methanogens [32, 34].

3.1.3 Power-to-gas technology costs

According to the presumed importance of PtG technologies in the transformation of the energy system and their relative novelty, accompanied investment costs and their expected development are important factors, not only for implementing parties but also for funding and legislative bodies. However, owing to the low number of actual implementations [46], the actual costs for commercial PtG plants are of limited availability, and must be extrapolated from established technologies, pilot plants, and expert estimations. As the review of available relevant literature, published as part of Böhm *et al.* [47], has shown, appropriate projections based on these data are not consistent.

The results for electrolysis, depicted in Figure 3.5, revealed a cost range for recent implementations at capacities <1 MW of approximately 1090–2000 €/kW_{el} for AEC systems [48–50], and approximately 1500 €/kW_{el} for PEMEC systems [49, 50]. For SOEC systems, no cost data for this dedicated capacity range were found, which is assumed to result from the early technology state (cf. Table 3.1). The International Energy Agency (IEA) hydrogen project database [46] only lists 17 registered projects using SOEC technology that were online before 2020 – compared to 219 AEC and PEMEC based projects – with only two of them not being categorized as *DEMO*. For plant scales in the range of 1–10 $\mathrm{MW_{el}}$, the investigated data generally shows a lower investment cost for AEC at 800–1400 €/$\mathrm{kW_{el}}$ [20, 48, 51–53], whereas for PEMEC no clear cost reduction could be identified, but rather, an extension of the cost range. The broad distribution of the data from 960 €/$\mathrm{kW_{el}}$ to 2100 €/$\mathrm{kW_{el}}$ [20, 48, 52, 53] revealed significant uncertainties, which may be explained by the fact that PEM electrolyzers with individual scales significantly greater than 1 $\mathrm{MW_{el}}$ were first implemented in the last decade [46]. For high-temperature electrolysis, only projections for multi-MW plants are available, owing to the lack of actual implementations, which are in the range of 2250–2500 €/$\mathrm{kW_{el}}$ for system scales of 5–10 $\mathrm{MW_{el}}$ [20, 48, 54], although significant outliers of <600 €/$\mathrm{kW_{el}}$ were excluded [51, 55, 56].

However, beyond this relatable set of data, the executed literature review also revealed some major issues in conjunction with publicly available data on electrolysis costs, as it often misses relevant information to allow for a serious comparison. Many of the gathered cost data do not specify a related system size for both the AEC and PEMEC findings [50, 51, 54, 57, 58]. Furthermore, the data may have differing or unspecified system boundaries, and thus include peripherals, or misses information regarding the capacity reference used (electric input, lower heating value (LHV) or higher heating value (HHV) of the product). These issues were also confirmed by a review of electrolysis cost studies of 30 years conducted by Saba *et al.* [49].

Since the publication of the initial literature study, further publications related to electrolysis cost data and development have become available. For instance, the recent *IEA Global Hydrogen Review 2021* [7] states that the costs of alkaline electrolysis systems in China are in the low range of 660–1150 €/$\mathrm{kW_{el}}$, including all peripherals, engineering, procurement and construction, with individual sources reporting costs as low as 500 €/$\mathrm{kW_{el}}$. In comparison, the global average is quoted with approximately 890–1240 €/$\mathrm{kW_{el}}$ for AEC and 1550 €/$\mathrm{kW_{el}}$ for PEM, both of which are covered by the ranges given above.[1] Though, most of the recently published data is based on the sources outlined above, or the results derived in Böhm *et al.* [47], rather than resulting from actual installations. These new data are not included in Figure 3.5.

A similar review was performed for chemical and biological methanation. However, the availability of recent cost data is sparse, which is why data from 2012 onward are included. In the capacity range of 1–30 $\mathrm{MW_{SNG}}$, chemical methanation showed a cost range of 160–1970 €/$\mathrm{kW_{SNG}}$, as confirmed by Schlautmann *et al.* [59]. By comparison, biological methanation is in the range of 100–1430 €/$\mathrm{kW_{SNG}}$ [35, 37, 60–64]. However, this generally lower cost range for biological methanation has not been confirmed by recent analyses of state of the art [65]. An overview of the data, including outliers in the system size from 0.1 $\mathrm{MW_{SNG}}$ to 1000 $\mathrm{MW_{SNG}}$, is provided in Figure 3.6.

3.2 Techno-economic assessment of energy technologies

The techno-economic assessment (TEA) is considered an important measure for achieving the present challenges of energy transition within the given time frame for implementation. It allows estimations of the technical and economic feasibility of potential technologies and identification of critical characteristics. Therefore, it represents a supportive measure for identifying and

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[1] Exchange rate USD/EUR=1.13

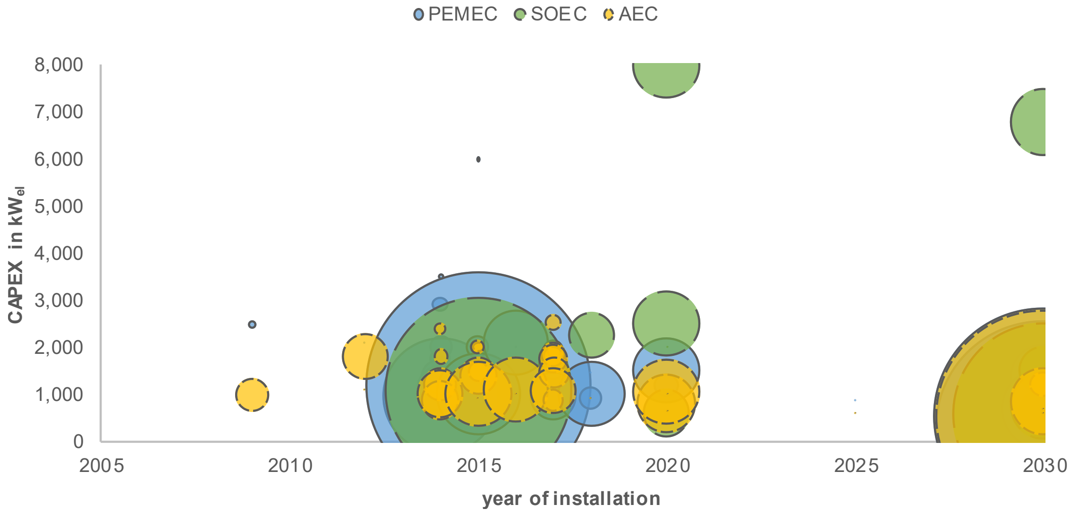


Figure 3.5: Collected data on specific investment costs for different electrolysis installations related to the year of installation (bubble area indicates the rated power from 0.1 to 100 $\mathrm{MW_{el}}$). Source: Böhm *et al.* [47]

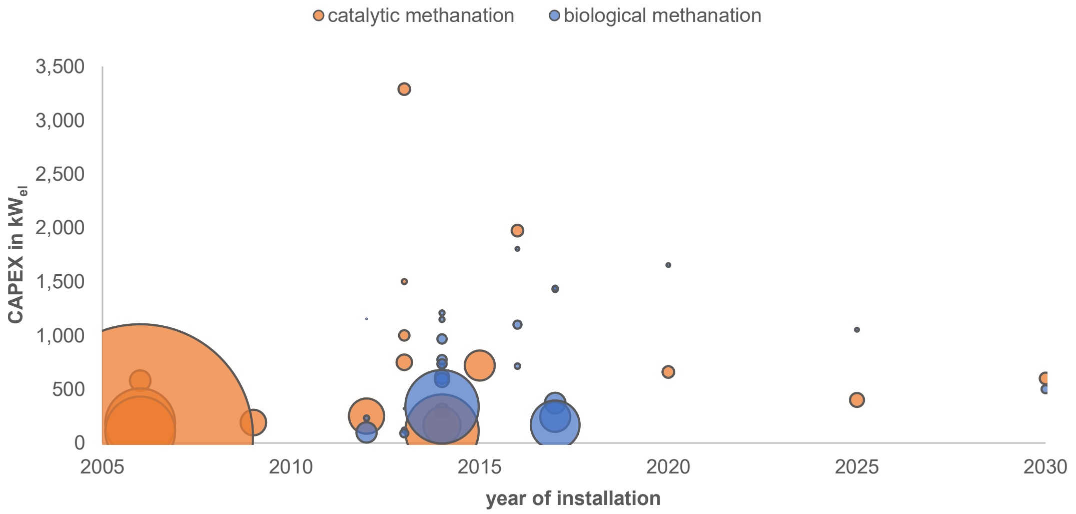


Figure 3.6: Collected data on specific investment costs for different methanation installations related to the year of installation (bubble area indicates the rated power from 0.1 to 1000 $\mathrm{MW_{SNG}}$). Source: Böhm *et al.* [47]

prioritizing R&D efforts as well as regulatory and funding necessities. With the increasing need for early evaluation of emerging technologies to cope with climate change, the standardization of TEA methods has become a topic of recent research [66, 67]. However, to date, TEA of energy technologies has aggregated various methodological approaches and indicators. The following sections provide an overview of the principles and fundamental contents of the TEA method.

3.2.1 Technology maturity

For the classification of a process or product in terms of its assessability, technological maturity is an important factor, since it implicitly sets limitations on the availability of reliable data and the accuracy of projections. In this context, in addition to other models, technology readiness level (TRL) is an established and commonly used scale to describe the readiness of emerging technologies. TRL was originally invented by National Aeronautics and Space Administration (NASA) in the 1970s and represents, in its current form, a nine-level scale [68]. The TRL scale is since been found, partially in adapted forms, in various applications and basically describes technology maturity from basic principles (TRL 1) to proven operational systems (TRL 9). It was also adopted by the EU to evaluate research projects under the EU Framework Programme for Research and Innovation *Horizon 2020 (H2020)* [69]. Table 3.3 provides an overview of the different levels mapped to different phases of maturity as specified for the chemical industry by Buchner *et al.* [70], which is used as a reference for this work.

Table 3.3: Technology readiness levels (TRLs) and characterization. Based on Zimmermann *et al.* [66] and Buchner *et al.* [70]

TRL	Phase	Title	EU H2020 definition based on [69]
1	Research	Idea	Basic principles observed
2		Concept	Technology concept formulated
3		Proof of concept	Experimental proof of concept
4	Development	Preliminary process development	Technology validated in lab
5		Detailed process development	Technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies)
6		Pilot trials	Technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies)
7	Deployment	Demonstration and full-scale engineering	System prototype demonstration in operational environment
8		Construction and start-up	System complete and qualified
9		Continuous operation	Actual system proven in operational environment (competitive manufacturing in the case of key enabling technologies; or in space)

However, while the NASA definition of *technology* is usually conceptualized at the level of a component technology featuring new materials, scale, or working principles [68], the scope of the term *technology*, and thus, the TRL classification in the H2020 context, is not clearly defined.

Therefore, in energy research, the TRL method is commonly used for single components, processes, and (sub)systems. This also includes the integration of different technologies in novel implementations; thus, even though the use of established components implies individually high TRLs of 7–9, their integration and combination in a novel process or system may result in a significantly lower TRL for the concept as a whole. Thus, the definition of a TRL for a certain technology always depends on the set system boundary and, consequently, on the definition of such boundaries.

With respect to TEA, the identification of the appropriate TRL for an investigated system, subsystem, or process already allows for a rough estimation of what data can theoretically be expected to be available for the TEA. Conversely, this also clarifies the data that must be approximated or extrapolated from previous experiences or comparable technologies [66]. Therefore, process or technology assessment often depends on a combination of both primary and calculated secondary data [71]. Additionally, a low TRL also means that assessments may have to be repeated or updated with the increasing maturity of the technology. Nonetheless, TEA can still be a valuable measure for evaluating and comparing different process routes at very low TRLs to identify preferable options and relevant indicators at an early stage of development. For instance, Böhm and Lindorfer [72] used this method to identify potential materials for seasonal thermal energy storage at TRL 2–4 using data for reasonable process routes.

As mentioned previously, TRLs can differ between the individual components of an investigated technology. This also accounts for the PtG, or more precisely, the PtM process. Although individual technologies within the PtG process route, such as low-temperature electrolysis (cf. Table 3.1), have already reached TRL 9, the concept of PtG as a whole, including the integration of methanation, is characterized by a TRL of 5–7 [16]. Therefore, techno-economic data of alkaline and PEM electrolysis, such as system efficiencies, implementation costs, and expected lifetimes, are available for commercial applications. This is similar to catalytic methanation, where experience from multiple GW of installed capacities for CO methanation from coal gasification is available [33]. However, an efficient combination of electrolysis and methanation within a PtM process is still under intense research to optimize the use of synergies and operating conditions. This is even more relevant for highly integrated processes, such as the thermal integration of endothermal co-electrolysis and exothermal methanation, for the highest process efficiency [27, 42]. Additionally, the achievement of commercial status (TRL 9) still indicates the availability of a functional and reliable implementation and does not describe techno-economic invariability. Therefore, a technological materiality, or n^{th} of its kind implementation, would have to be reached, which can be stated by a share of about 1 % in the global market or energy mix [73]. In particular, component lifetimes, efficiencies, and costs are expected to be subject to these learning effects and experience permanent improvements. The estimation of the future development of such parameters, and thus the techno-economic performance of the process as a whole, requires additional measures and methods. A common experience with commercialization is the reduction of technology costs with increasing numbers of produced units and implementation scales. These effects are further discussed using the term *economies of scale.*

3.2.2 Economies of scale

For an effective and sustainable transformation of today's energy systems, the relevant technologies also need to be economically viable compared to the currently predominant fossil competitors. However, corresponding cost reductions are presumed for promising technologies or are already seen and verified, such as, for example, for wind and PV power plants. An appropriate evaluation of potential future technology costs and the investments required to reach this competitiveness is not only relevant to the implementing parties. Especially for policy- and decision-makers in the field of energy politics, this information is of significant importance because it allows recommendations on regulatory frameworks and funding as driving factors for the enforcement of emerging technologies [74, 75]. However, for low TRLs, such estimations of the expected cost reduction must be based on known methods and empirical values, such as economies of scale.

In general, economies of scale subsume the different effects of cost reductions that come with increasing scales of product or technology deployment. Within the scope of this thesis, two different scaling effects are differentiated: (i) cost changes resulting from increasing cumulative production volumes, also referred to as *economies of number scale*, and (ii) nonlinear increases in costs for implementations of larger scales, referred to as *economies of unit scale.* The differences in their interpretation, application, and use are discussed below.

3.2.2.1 Economies of number scale – Technological learning

Technological learning describes the reduction in manufacturing costs of a product or technology through an increasing number of units produced. This cost reduction is known as *learning curve* or *experience curve.* Even though these terms are often used interchangeably in the literature, they originally describe two different concepts: the learning curve, which was first published by Wright [76], refers to the variation in labor costs by repeated production as the experience of workers and thus their efficiency increases with every unit. Therefore, the learning curve applies to a single production process or company, whereas the term experience curve, which was first characterized by Boston Consulting Group [77] in the 1960s, extends the scope of the effect to an entire industry [78]. Concerning today's global market economy and the energy transition being an overarching challenge, the technological learning used herein refers to the concept of the experience curve.

In contrast to the learning curve, the experience curve not only relates to individual input costs, resulting from manufacturing time or material usage, but also represents the evolution of the total costs of a production process. Thus, it also includes indirect costs, such as for R&D, distribution, and marketing [79]. However, the experience curve uses the same principle as the learning curve theory observed by Wright [76] – a decrease in production time or costs, respectively, by a constant percentage, which is denoted as learning rate (LR), with each doubling of cumulative production (see Eq. (3.7)). This correlation implies that entrant technologies with comparably low cumulative production volumes experience a faster decrease in costs for a certain absolute increase in produced units than incumbent technologies underlying the same learning rate. Consequently, early investment in novel technologies can lead to a rapid decrease in production costs. This *learning investment* can thus be supported by policy interventions, such as funding or regulatory measures, to attract investments. Therefore, the evaluation of learning rates, and thus the projection of cost reduction potentials, is an important part of the energy transition to identify and support long-term viable technologies in the early stages for rapid implementation. Consequently, in recent years, technological learning methods have been applied to various energy technologies to identify corresponding learning rates (cf. [80–82]).

The mathematical description of the experience curve can be represented as follows:

$$C_t = C_0 \left(\frac{X_t}{X_0} \right)^{-r} \tag{3.7}$$

with:

C_0 ... initial costs at time $t = 0$
C_t ... costs at a given time t
X_0 ... initial cumulative production at time $t = 0$
X_t ... cumulative production at a given time t
r ... learning/experience parameter

Consequently, in a twice-logarithmic plot, the experience curve is a linear function with a gradient of $-r$. Because the constant decrease by LR is observed every doubling of the cumulative production, the correlation of the learning parameter r and LR can be described as in Eq. (3.8), at which LR is often substituted by the progress ratio (PR).

$$LR = 1 - PR = 1 - 2^{-r} \tag{3.8}$$

Beyond the basic form of the experience curve, different adaptations and advancements are found in the literature, which are briefly discussed below. For consistency with relevant and prior publications [79, 83], the term *learning curve* is used synonymously with the *experience curve*, although primarily experience curves are described.

Single-factor experience curves The basic form of the experience curve characterized by Eq. (3.7) is, by definition, only dependent on cumulative production; thus it is also known as one-factor learning curve (OFLC). Hence, the corresponding technology learning rates can be derived from data on investment costs and cumulative installations, which are relatively easy to collect and well documented compared to other underlying cost drivers [83]. However, this simple form with its single learning rate is often not appropriate or sufficient to describe the cost-reduction effects seen for certain technologies. Therefore, Ferioli, Schoots, and van der Zwaan [84] proposed an adaptation of the OFLC by splitting it up into two parts: one that is apparently affected by technological learning and one whose costs remain roughly constant (see Eq. (3.7), where α represents the initial cost share of the *learning part*). With this two-component approach, it becomes possible to differentiate between cost components that underlie certain reductions by gained experience (e.g., production processes) and those that do not (e.g., labor or certain material costs). Its practicability for energy technologies was demonstrated by Ferioli, Schoots, and van der Zwaan [84] and van der Zwaan *et al.* [85].

$$C_t = \alpha C_0 \left(\frac{X_t}{X_0} \right)^{-r} + (1 - \alpha) C_0 \tag{3.9}$$

Naturally, this concept results in different learning rates based on historic data, or conversely, in different forecast costs compared to the basic OFLC. However, Ferioli, Schoots, and van der Zwaan [84] showed that multi-component experience curves achieve a better match to the historic correlations between costs and production volumes. However, they also outlined that simple extrapolations of cost development over several orders of magnitude of cumulative production can lead to significant errors in the forecast. In addition to the presumption of unchanging technology and thus a constant learning rate, the simplification to two components, learning and non-learning, may lead to under- or overestimation of individual learning effects. Additionally, because established technologies and components still underlie technological learning, their implementation in high numbers as part of an entrant technology can still lead to significant cost reductions, which may be unconsidered as part of the non-learning component. Therefore, a more detailed division of relevant components would lead to more precise results. Furthermore, it would allow better transferability and reusability of individual experience curves between technologies, even though data on particular cost structures and manufacturing processes are probably non-existent or challenging to derive [79, 85].

Two- and multifactor experience curves While the limitation of the OFLC to cumulative production as its sole dependency allows a simple aggregation of cost-reduction drivers in a single factor, this concept only corresponds to observation. Therefore, other implied drivers, such as the impact of investments in R&D, cannot be differentiated directly. However, an evaluation of these factors can be invaluable from a policymaker's perspective for identifying and assessing the effectiveness of policies supporting R&D investments [78, 83]. Consequently, Kouvaritakis, Soria, and Isoard [86] proposed an extension of the OFLC by separating *learning-by-doing* from *learning-by-researching* in a two-factor learning curve (TFLC):

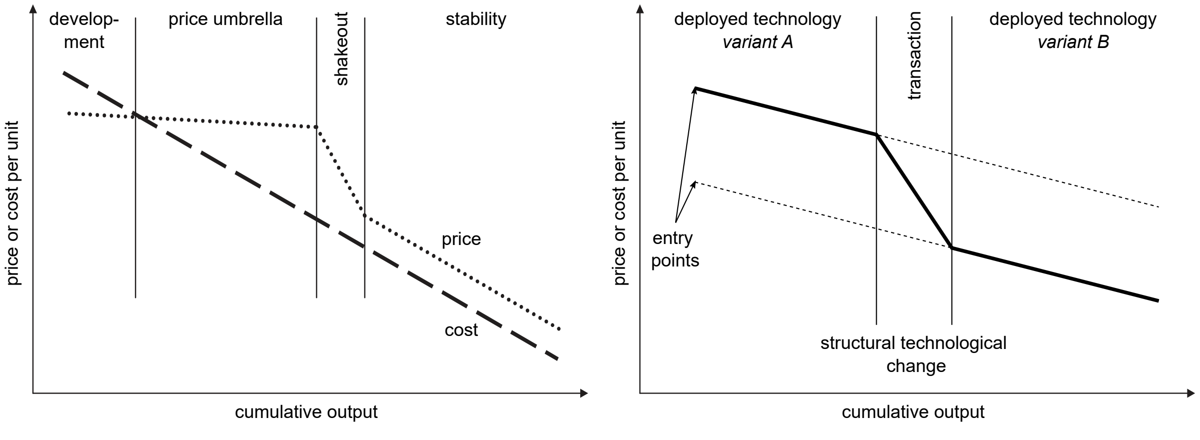
$$C_t = C_0 \left(\frac{X_t}{X_0}\right)^{-r} K^{-s} \quad \text{with} \quad LR_{R\&D} = 1 - 2^{-s} \tag{3.10}$$

In Eq. (3.10), the former part is identical to the OFLC and represents the *learning-by-doing* effect. The latter part describes the cost reductions driven by R&D efforts, which are measured by the *knowledge stock* K and the corresponding learning parameter s. As for the OFLC, unit costs decrease by the *learning-by-researching* rate $LR_{R\&D}$ with each doubling of the R&D effort. However, the knowledge stock parameter K must consider that the knowledge gained from R&D expenditures does not directly affect cost reductions, but with a certain time lag, and the knowledge stock depreciates when no further research is conducted [78].

The application of the TFLC in various studies supports the assumption of a correlation between R&D expenditures and technology cost reductions, and to some extent suggests a more significant effect contribution than learning-by-doing (see also [86–92]). However, analyses by Miketa and Schrattenholzer [93] show that a general improvement in the accuracy of describing cost reductions by technological learning is not expected. Furthermore, it has to be considered that the availability of appropriate public data on R&D expenditures, or number of patents as a proxy value [89], is often limited or inadequately disaggregated – or even incorrect, if companies intentionally avoid patent applications to not forfeit competitive advantage.

Beyond the models described by OFLCs and TFLCs, additional approaches are found in the literature (see [78, 94]) to decouple and accommodate the impact of additional technological learning effects, such as unit-related scaling, commodity prices, labor costs, and process efficiency changes.

Methodological issues Although technological learning is an important resource for evaluating the long-term competitiveness of novel technologies, the method itself also has some substantial issues. One major issue is the differentiation between cost- and price-related data. The learning and experience curve models discussed here represent the correlation between cumulative production and manufacturing costs. However, for most technologies, manufacturing *costs* are not directly available; hence, most studies on experience curves use unit *prices* or investment costs. However, market prices and manufacturing costs are often not directly related, but, besides some fixed margins, are affected by many parameters, such as marketing strategy, supply and demand, market competitiveness, and subsidies [78]. Because the knowledge of the actual margin between production and purchase costs is usually confined to the manufacturer, influences, such as price umbrellas or shakeout effects (see Figure 3.7a), can hardly be considered [79, 95, 96]. In addition to this uncertainty in the available cost data, there may also be uncertainties in the correlation between cumulative production and costs, which result from spillover effects. These spillover effects describe the impact of technological learning observed for one technology, which is also reflected in other technologies with related applications or use. Hence, appropriate learning effects cannot be related only to the cumulative production of a single technology. Consequently, a comprehensive structural technological change can also lead to discontinuity in the experience curve (see Figure 3.7b). Thus, such radical changes allow related technologies to benefit from each other's experience by enabling a change in the entry point of the experience curve and possibly the learning rate itself [79, 97, 98].



(a) Price-cost relation for a new product **(b)** Structural technological change

Figure 3.7: Methodological issues of technological learning. Source: Böhm, Goers, and Zauner [79]

Above all, the evaluation of the corresponding learning rates is usually performed by analyzing the historic cost and market volume development. Because an appropriate analysis requires a certain magnitude of produced units, this is not reasonable for applications on a low TRL. Nevertheless, TEAs of energy technologies are often done before their entry in the market, and thus, before an appropriate technology maturity is reached, to support initial decisions on investments or funding. Simple reuse of the learning rates found for established technologies with comparable functionality or usage is usually not feasible, as minor changes in technologies can have a significant influence on the underlying learning mechanisms. Therefore, alternative approaches must be used to identify the learning rates at an early development stage [79]. A recent review by Thomassen, Van Passel, and Dewulf [99] provides a comprehensive overview of the application and use of technological learning methods in prospective technology assessments.

3.2.2.2 Economies of unit scale – Technological scale-up

In addition to the cost reduction induced by technological learning, scaling effects related to the unit scale of the installation are relevant to the cost development of emerging technologies. In a strict sense, economies of unit scale do not represent a reduction in absolute technology costs but describe a non-linear dependency between implementation scale (size, capacity, and nominal power) and costs. Therefore, they only represent a decline in specific costs. A logarithmic relationship is commonly used to describe this type of scaling effect.

$$C_b = C_a \left(\frac{S_b}{S_a} \right)^f \tag{3.11}$$

In Eq. (3.11), C_a represents the absolute costs for a known reference implementation at scale S_a and C_b is the absolute cost at scale S_b. f denotes the applied scaling exponent. Because $f = 0.6$ is a reasonable guideline for an initial estimation in various applications, especially in the field of chemical engineering, the given approach is also known as the *six-tenth factor rule* [100]. The mathematical description is similar to that of the OFLC, although the scaling exponent is always positive, and thus, always $C_b \geq C_a$. Furthermore, unit upscaling is not necessarily attended by reduction of specific costs; hence, values of $f > 1$ are also observed for certain equipment. The actual scaling exponent is individual for each application or component; however, appropriate values for common equipment are often well known and can be found in chemical engineering handbooks (e.g., [100, 101]). However, in practice, scaling exponents are not universal and are, thus, only valid for limited scale ranges. Generally, the above method should not be used for a ratio of scales greater than 10. Furthermore, differences in the type and material of construction, operating conditions concerning pressure and temperature, and other pertinent variables can influence scaling parameters between two similar types of equipment and must thus be considered when applying the method [100].

3.2.2.3 Scaling effects relating to power-to-gas applications

According to the issues discussed above, the estimations of learning rates for PtG technologies are limited owing to the novelty of the technology, and thus, the limited availability of data on cumulative production volumes and costs. The corresponding findings are outlined in Böhm *et al.* [47] and Böhm, Goers, and Zauner [79] and summarized and updated below.

Analyses by Schoots *et al.* [102] on hydrogen production by electrolysis indicated a learning rate of 18 % but covered with significant uncertainty ($R^2 = 0.28$) because of the low number of considered data points. Schmidt *et al.* [80] published similar results with a learning rate of 18 ± 6 %, though apparently basing on the data used by [102]. Even though neither study mentioned a specific technology, a clear dominance of alkaline cells in the observation period (1920–2017) can be presumed because of the relative novelty of PEM and solid oxide cells. A recent compilation dedicated to AEC by Krishnan *et al.* [103] led to a slightly lower rate of 16 ± 6 % between 1956 and 2016. In addition, studies with a clear indication of the underlying electrolysis technologies based on historic cost data are currently not available. However, an expert elicitation study on water electrolysis technology development by Schmidt *et al.* [20] resulted in the estimation of comparable learning rates for PEMEC and AEC of 18 %. However, the learning rates for SOEC were identified to be substantially higher, with a mean value of 28 %, although covered with high uncertainty of ±16 percentage points.

Given the potential technological similarities of electrolysis cells to their corresponding fuel cell (FC) pendants, appropriate similarities may also be represented in terms of technological learning parameters. In this context, fuel cells have been investigated in more detail and discussed in relevant publications. Investigations of various FC types performed by Schoots, Kramer, and van der Zwaan [104] revealed a learning rate of 18 ± 9 % for alkaline fuel cell (AFC), which is comparable to the corresponding electrolysis technology. However, for PEM, the found differences between electrolysis and fuel cells are more significant with learning rates of 21 ± 4 % for PEM fuel cell (PEMFC). Similar rates have been reported in other studies, either dedicated to PEMFC (16–21.4 %, [105–107]) or without specification of the technology (18 %, [108, 109]). An application of the TFLC method by Mayer *et al.* [92] suggested a learning-by-doing rate of 13 % and a learning-by-research rate of 21 % for PEMFC. Concerning solid oxide fuel cell (SOFC), a comprehensive analysis was performed by Rivera-Tinoco, Schoots, and van der Zwaan [110], where they not only differentiated between R&D, pilot, and early commercial states, but also considered different drivers of cost reduction (learning, economies of scale, and automation). Thus, considering all cost reduction phenomena, discovered learning rates cover a wide range of 12–44 %, whereas the average value over all development stages resulted in a rate of 35 %. The highest learning rates were found in the pilot stage and the lowest in the early commercial state. Interestingly, the range was identical to the values elicited by Schmidt *et al.* [20] for SOEC. By contrast, appropriate analyses of SOFC in commercial stationary combined heat and power (CHP) applications did not reveal any significant cost reductions [107, 108]. However, considering that these market analyses usually relate to the system rather than the cell level, a direct comparison between electrolysis and fuel cell applications is probably limited due to significant differences in peripheral components, even though cell designs may be similar.

Regarding methanation, there is no known published analysis of technological learning, apart from those published in the course of this thesis, either for chemical or biological processes. Schoots *et al.* [102] identified a learning rate of 11 ± 6 % for steam methane reforming (SMR), which also represents a nickel-based catalytic process, but apart from that is hardly comparable to chemical methanation from a process-related perspective. By contrast, biological methanation is potentially comparable to biogas production, for which Junginger *et al.* [111] identified a learning rate of 12 %. Furthermore, de Wit *et al.* [112] suggested learning rates of 1–20 % for different biofuel production processes.

Concerning economies of unit scale, publications based on manufacturer data suggest that cost-reduction effects are significant for alkaline electrolysis at a single-MW scale, but flatten with further increases in system size [49, 103]. This was also found by Morgan, Manwell, and McGowan [113], who suggested that the potential for large cost reduction via upscaling is limited due to technical constraints for the cell size (e.g., problems with leakages). Therefore, modularization is expected to be preferable for increasing the system capacities. Appropriate scaling factors for electrolyzers are not found in the literature and would be highly inaccurate based on cost data at the system level (see Böhm *et al.* [47]). Hence, an individual assessment of the included equipment is preferred and reasonable. For methanation, scaling factors suggested for biofuel production, as suggested by de Wit *et al.* [112] in a range of 0.7–0.85, may be reasonable based on the comparability of chemical engineering. However, the limitations of the system-wide evaluation discussed for electrolysis also apply to methanation technology.

3.2.3 Techno-economic assessment practices for early-stage technologies

The techno-economic assessment generally represents an emerging methodology for the prospective evaluation of (chemical) processes and technologies in terms of economic profitability [114]. Consequently, common standards for unified procedures or indicators are still lacking, even though corresponding efforts have been increasing and enforced in recent years (e.g., [66, 67, 115]), albeit not exempt from criticism of different aspects [116]. In addition to precise guidelines, Van Dael *et al.* [117] suggested harmonizing the general structure of TEA in four steps:

(i) A market study to determine prices and market volumes, along with the suitable market potential for the assessed product or technology.

(ii) The elaboration of the process flow diagram for the assessed system, including the calculation of mass and energy balances.

(iii) The economic analysis evaluating the profitability of the system based on specific indicators representing investment criteria.

(iv) A risk analysis to evaluate uncertainties on the calculated indicators and their sensitivities to certain parameters.

However, while the TEA method presumes an industrial implementation, namely, the n^{th} of its kind, it is primarily applied to technologies under development [114]. Therefore, the evaluation of market and process data is usually prospective and major technical challenges must be overcome. Consequently, the lower the TRL, the more assumptions are required; thus, the methods of TEA may vary according to the level of process or technology maturity. Therefore, Thomassen *et al.* [114] proposed an overarching assessment framework (for integrated environmental techno-economic assessment) that applies to all stages of TRL. According to them, in the first TRLs (1–3), the use of qualitative or semi-quantitative screening methods (e.g., strengths, weaknesses, opportunities, threats (SWOT) analysis) can be considered an early and rough prospective assessment method, as the availability of quantitative data is usually limited at this stage. This could also include decomposition analysis, decomposing the investigated technology into its components, materials, and energy and resource streams to identify and depict potential critical elements. Starting with TRL 4, qualitative assessment can evolve into a streamlined quantitative assessment, which basically already covers the same scope as a full-scale technology assessment. Beyond the definition of the system boundary, this requires the collection of technological and economic data on the components and streams involved. Although early data can be derived from lab-scale experiments starting at TRL 3–4, and some process- and market-related primary data may be TRL-independent, secondary data must be derived from the available primary data [71]. However, to assess the n^{th} of its kind, appropriate scale-up methods and procedures must be utilized. These methods must be identified based on required data [114]. At high TRLs (7–9), which are basically only defined by the first demonstration and proven in operational environment (see Table 3.3), these scale-up methods are mainly required for economic data, while technological data are mostly available. The corresponding scaling methods have been previously discussed.

By the method itself, prospective TEA naturally requires making assumptions and estimates, which implies a certain degree of uncertainty in the derived results. Therefore, the execution of an appropriate uncertainty and sensitivity analysis is an integral part of a sound techno-economic evaluation. Even though the range of potential analysis methods in this context is broad, the applicability of advanced methods, particularly at low TRLs, is often limited (cf. van der Spek *et al.* [118]). At an early stage of technology development, it is usually sufficient to evaluate the impact of individual parameters on the investigated output parameters, thus allowing the identification of critical process parameters. Therefore, local sensitivity analyses are a common method of choice in which one or more variables are varied from a nominal or reference value. The simplest form of such a local method is the *one-at-a-time sensitivity analysis*, which is applied by varying the selected input parameters by a certain amount or percentage (e.g., ±25 %) and determining the respective impact on the assessment results (e.g., generation costs). An illustration of the individual impacts in descending order according to their corresponding impacts results in a so-called *tornado diagram* (see Figure 5.7). Even though this method allows for the simple identification of high- and low-sensitivity variables, the fixation on a certain amount of variation makes the method inappropriate for variables with a nonlinear dependency. In this context, a *one-way sensitivity analysis* is more appropriate, where variables are again varied one by one, but impacts are determined over their entire range of potential values [118]. Consequently, this allows the identification of critical value ranges for individual parameters with a nonlinear impact on the assessment results. To reduce the number of calculations for many parameters and identify interdependencies between them, this can be further extended to *n-ways sensitivity analysis* by varying multiple (n) parameters simultaneously and representing the result as a surface function [119]. Additional established and emerging methods of uncertainty analysis, along with their applicability to individual evaluations, are discussed in the relevant literature [118–120].

3.3 Techno-economic evaluations of power-to-gas applications

Renewable gases in terms of hydrogen and SNG are considered an indisputable part of energy transition and de-fossilization in various fields. Thus, numerous studies in recent years have shown the techno-economic potential of PtG in different fields of application. However, even though the cost factors of actual implementations have been and are being analyzed as part of relevant research projects, such as, for example, the H2020 project STORE&Go, which included the development and techno-economic investigation of three different PtM plants [59], applications at reasonable scales still have to rely on prospective evaluations using the aforementioned methods. Based on the expected market potential and early estimations of experience curves, most publications suggest significant cost reductions in the medium and long term. For instance, the learning rates analyzed for PtH_2 by Krishnan *et al.* [103] imply a reduction in capital expenditures (CAPEX) by 27 % for an increase in cumulative installations by 80 GW, which is identical to the capacity stated in the EU hydrogen strategy [8]. Other studies predicted a reduction of approximately 60 % for PEMEC in a similar deployment scenario (90 GW by 2030) [121]. However, concerning the broad application of hydrogen and SNG as energy carriers, in addition to the initial CAPEX, final product costs are of considerable relevance. According to Hydrogen Council [121], the current costs of renewable hydrogen from electrolysis relate to 5.3 €/kg in Europe. Recent studies project levelized costs of hydrogen (LCoH) from RES of approximately 2.3 €/kg for 2030 and down to 1.5 €/kg for 2050 on a global scale, depending on the energy source (onshore wind and PV; the cost of offshore wind is partially significantly higher).[2] Under optimistic conditions, even lower LCoH (<1 €/kg) are conceivable. However, owing to the high share of electricity costs, LCoH differ significantly between countries, and thus, are potentially afflicted with additional costs for (transnational) transport and distribution [122].

In contrast to hydrogen from electrolysis, the projections of the generation costs for SNG from PtM are more diverse. Gorre, Ortloff, and van Leeuwen [123] identified methane production costs of 50–90 €/MWh$_{\mathrm{SNG}}$ for 2030 and 25–65 €/MWh$_{\mathrm{SNG}}$ for 2050 at plant scales of 10 MW$_{\mathrm{el}}$, presuming significant cost reductions for both, CAPEX and operational expenditures (OPEX). A similar cost range of 30–80 €/MWh$_{\mathrm{SNG}}$ was found in [124]. In addition, Gorre, Ortloff, and van Leeuwen [123] showed that there are high dependencies on market conditions (long-term contracts vs. short-term markets) and flexibility (direct coupling with RES, seasonal storage). Furthermore, significant cost reduction potential by optimization of interlinking of electrolysis and methanation, and consideration of intermediate storage was found [123, 125]. This may become particularly relevant if PtM is considered as a flexibility or (seasonal) storage option, because electrolysis and methanation processes can be temporally decoupled to increase individual full-load hours (FLH) and decrease nominal methanation capacities for economic optimization. In this context, centralized production to satisfy local demand may be beneficial from a techno-economic perspective and could enable additional scaling effects. Appropriate cost reduction potential by upscaling has been identified in several relevant studies [126–129].

However, while a broad application of PtG technologies for the provision of renewable energy carriers allows for a more generalized projection of potential generation costs, this is hardly possible for more specific usage scenarios. Since electrolysis and methanation processes are considered potential options for de-fossilization in various fields of application, corresponding TEAs are numerous. The economic performance of renewable hydrogen, and thus electrolysis, beyond energy use, is of increasing relevance as a process gas for industrial applications, especially as a reducing agent in metallurgy, including, among others, steelmaking and copper production [130]. Furthermore, PtM could be part of future large-scale CCU applications to either produce a renewable energy carrier from biological or atmospheric CO_2 or to establish closed carbon cycles for sectors with hard-to-abate carbon emissions. Thus, TEAs for corresponding applications are also of particular interest (e.g., as discussed in [27, 131]), especially when it comes to appropriate consideration of potential synergies of process integration. Consequently, it should be noted that a generalized TEA of PtG processes is not reasonable and usually has to be applied on a per-application basis. All the more, the definition of harmonized and reusable assessment methods is of considerable relevance.

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[2] Exchange rate USD/EUR=1.13

4 Methodology and material

To overcome the challenges that arise from the techno-economic assessment of emerging technologies in general and PtG in particular, different evaluation methods and materials were developed and used to assess and project cost developments of PtG technologies and applications. Most importantly, this includes the distinct use of a disaggregated experience curve model for the prospective evaluation of cost reductions for early-stage technologies. These methods and materials are summarized in the following subsections, but are related to specific applications in different publications. Thus, for a comprehensive description, it may be referred to the indicated article. Within this thesis, primarily used in the context of PtG, these methods are intended to be widely generic and largely transferable to other technologies.

4.1 Consideration of component-based economies of scale

As discussed in 3.2.2, the application of economies-of-scale effects on a technology or product holistically is usually unreasonable or afflicted with significant uncertainties. This particularly applies to technologies at low maturity, that is, with a low number of cumulative productions, where a derivation of learning effects from observed developments is not feasible. Consequently, a disaggregated experience curve model was defined in the course of this thesis to allow for the evaluation of cost effects on a per-component basis. Furthermore, this also enables the consideration of external learning effects, namely spillover effects. The definition of the model and its associated impacts on the evaluation of technology cost development are discussed briefly below.

A detailed description of the experience curve model is provided in Böhm, Goers, and Zauner [79], using the example of different electrolysis cell technologies. An application to PtG in general was published in Böhm *et al.* [47], along with consideration of unit-scaling factors. Furthermore, in Böhm, Lehner, and Kienberger [27], the model was adapted to a specific integrated PtG system as part of the TEA of an industry-related application.

4.1.1 Disaggregation of experience curves

By disaggregating the investigated technology in individual subcomponents or cost shares, the experience curve model allows comparing technological learning effects and simple scaling and innovation processes, based on

- *Cost reductions from series/mass productions*: Cost savings that result from an upscaling of the manufacturing process are easier to distinguish on a per-component basis.
- *Changing material costs*: The variety of materials used in the manufacturing process becomes more manageable on a per-component basis. Thus, variations in raw material costs can be directly attributed to the cost effects.

- *Reductions in material usage*: Cost reductions from reduced material usage can be evaluated in more detail at a component level, along with an overall potential for material savings.
- *Improvements in manufacturing time*: Improvements in processing time can be determined and evaluated more precisely at the component level than for the whole appliance. This not only includes machine processing costs, but also manual working time costs.

Despite the increased complexity and number of cost effects involved, which still require a certain amount of technological experience gained through a few orders of production, many individual components are not reinvented for every single purpose, but are often reused within different applications. Hence, knowledge of the underlying cost effects allows for a low-level evaluation of component- or process-wise learning effects, which can then be reused between comparable applications. Consequently, this enables the transferability of learning rates from well-established applications.

4.1.1.1 Implementation on module level

A simple form of multi-component experience curves was already proposed by Ferioli, Schoots, and van der Zwaan [84], as shown in Eq. (3.9), who separated costs into two components – a learning and non-learning ($LR = 0$) part. More comprehensively, technologies can be considered an aggregate of several components, or cost factors, each of which follows its own experience curve. Consequently, the overall technology costs can be described as follows:

$$C_t = \sum_{i=1}^{n} C_{0i} \left(\frac{X_{ti}}{X_{0i}}\right)^{-r_i} = C_{01} \left(\frac{X_{t1}}{X_{01}}\right)^{-r_1} + C_{02} \left(\frac{X_{t2}}{X_{02}}\right)^{-r_2} + \cdots + C_{0n} \left(\frac{X_{tn}}{X_{0n}}\right)^{-r_n} \tag{4.1}$$

with:

C_{0i} ... initial costs of component i at time $t = 0$
C_t ... total costs at a given time t
X_{0i} ... initial cumulative production of component i at time $t = 0$
X_{ti} ... cumulative production of component i at a given time t
r_i ... learning/experience parameter for component i (where $LR = 1 - 2^{-r}$)

The approach in Eq. (4.1) allows for the detailed integration of the aforementioned learning effects observed for manufacturing, potentially including all spillover effects from concurrent component usage and development. However, an exact analysis of individual production volumes decoupled from the technology in question is rarely feasible. Furthermore, the relevance of spillover effects is partially disputable or not within the own area of influence, particularly for (small) purchased parts. Therefore, further simplification is used to relate each individual experience to the cumulative production of the investigated technology (Eq. (4.2)). While this approach disregards certain factors, such as spillover effects for components, it is more practical for early cost estimations, and a certain level of disaggregation usually outweighs these effects. Tsuchiya and Kobayashi [132] used a similar approach to estimate mass production costs for automotive PEM fuel cells.

$$C_t = \sum_{i=1}^{n} C_{0i} \left(\frac{X_t}{X_0}\right)^{-r_i} = C_{01} \left(\frac{X_t}{X_0}\right)^{-r_1} + C_{02} \left(\frac{X_t}{X_0}\right)^{-r_2} + \cdots + C_{0n} \left(\frac{X_t}{X_0}\right)^{-r_n} \tag{4.2}$$

This already allows for the transferability of learning rates between technologies, as long as the respective component is used in a similar or even identical manner. However, if the observed learning effects are estimated differently between technologies, such as, for example, using different reference units, the corresponding learning rates can probably not be directly reused. For instance, as stated in [79], for the membranes used in PEM fuel and electrolysis cells, comparable learning rates can be expected according to material and component usage. However, it can also be assumed that, relating to the cell power, there will be variance in the development of the current densities for these two technologies along with the material usage of the membrane itself. This could be considered directly within the appropriate learning rate per application, but this would disable the advantage of comparability and interchangeability of learning rates between the two technologies. The concept of *learning properties* is introduced, to consider such non-linear dependencies within the model. A *learning property* describes a technology property that is also affected by its increasing cumulative production. Therefore, relevant properties are also defined to follow a simple OFLC with a certain learning rate (see Eq. (4.3)). A similar approach was used by Tsuchiya and Kobayashi [132] to describe the evolution of PEMFC power densities along the experience curve.

$$P_t = P_0 \left(\frac{X_t}{X_0}\right)^{-r_p} \tag{4.3}$$

with:

P_0 ... initial value of property P at time $t = 0$
P_t ... value of property P at a given time t
X_0 ... initial cumulative production at time $t = 0$
X_t ... cumulative production at a given time t
r_p ... learning/experience parameter for property P (where $LR_p = 1 - 2^{-r_p}$)

Learning properties can then be applied to affected components as per Eq. (4.4). Therefore, an appropriate *influence exponent ex* can be used, which defines the mathematical dependency between the properties and components. For instance, quadratic dependency ($ex = 2$) can be used to match properties related to a single dimension (e.g., length-based) with components requiring a two-dimensional relationship (e.g., area-based).

$$C_{ti} = C_{0i} \left(\frac{P_0}{P_t}\right)^{ex} \left(\frac{X_{ti}}{X_{0i}}\right)^{-r_i} = C_{0i} \left(\frac{X_{ti}}{X_{0i}}\right)^{-r_i + ex \cdot r_p} \tag{4.4}$$

In Böhm, Lehner, and Kienberger [27], the concept of *learning properties* was used to couple the experience curve effects of electrolysis (related to electric input power) and methanation (related to SNG output power) technologies to a common reference (electric input power). As a potential increase in electrolysis efficiency affects the dependency between the two references, an appropriate *learning property* was applied to the methanation components to consider this impact.

A full representation of the experience curve model for a multi-component module, where a single component supports multiple *learning properties* and vice versa, is described as follows:

$$C_t = \sum_{i=1}^{m} \left\{ C_{0i} \prod_{j_i=1}^{n_i} \left[\left(\frac{P_{0j_i}}{P_{tj_i}} \right)^{ex_j} \right] \left(\frac{X_t}{X_0} \right)^{-r_i} \right\} \tag{4.5}$$

4.1.1.2 Implementation on system level

In the relevant literature, experience curves for emerging technologies often only consider the novel share itself (e.g., electrolysis cells or stacks for water electrolysis systems), while the costs of the surrounding parts and peripherals are of less interest. Hence, to consider the full system costs, the cost effects for these parts could either be neglected, which would correlate with the approach of Ferioli, Schoots, and van der Zwaan [84] in Eq. (3.9), or technological learning is integrated accordingly. However, the learning effects on a macro-level (system) must consider additional aspects. On the one hand, experience curves for common peripheral parts are probably only marginally driven by the production of the investigated novel technology [83]. Although the corresponding spillover effects are presumed to be negligible at the module level, their impact is considerably higher at the system level. This is not only due to parallel usage in other applications, but also related to cost effects already experienced in the past, thus resulting in a corresponding higher starting point of cumulative production. Disregarding the latter can result in a significant overestimation of learning effects for entrant technologies (see Section 4.1.1.4). On the other hand, learning rates for common peripheral parts are probably well-known or easier to estimate, based on their less significant impact, whereby a detailed evaluation on a component-basis is usually not reasonable.

To account for learning effects, which are not directly related to the cumulative production of the investigated technology but still represent cost reductions as a function of its market development, production volumes have to be decoupled for peripheral modules. Consequently, while individual time-series data for the production volumes of every single module and complete system are defined, the relationship between the time series of those two observation levels is determined for every single module inside the calculation model. The module developed in this thesis implements four dependencies [79].

- *Direct:* The time-series data, which was defined for the overall system, is also used for the modules' learning curve. This implies that the learning effects for certain modules are also directly coupled to the production amounts of the system ($X_{module} = X_{system}$).
- *Independent:* Only the time-series data of the module itself is considered for the calculation of its learning curve. Consequently, an increase in the production of units of the system does not have a direct influence on the cost reduction of the module ($X_{module} = X_{module,ind}$).
- *Indirect:* The time-series data of the module and the system are added. Hence, direct learning effects from the production of new units of the investigated system as well as indirect effects from other usages (in concurrent applications) of the particular module are considered ($X_{module} = X_{module,ind} + X_{system}$).

- *Constant:* The cumulative production of the particular module is supposed to be constant. This means that theoretically, no additional units are produced, and therefore no learning effects occur, which is equal to defining a learning rate of $LR = 0$ for a particular module ($X_{module} = \text{const.}$).

Note: X_{module} ... cumulative production of the module
X_{system} ... cumulative production of the investigated system
$X_{module,ind}$ ... cumulative production of the module, independent of the system (concurrent usage)

4.1.1.3 Impact on the module cost structure

The disaggregation of experience curves into a component-based model has some noteworthy effects compared with the conventional application of the theory. One is the impact on the defined cost structure of the disaggregated module. Owing to different learning rates for the individual cost shares, the learning effects that result from an increase in the cumulative production lead to distinct cost (reduction) effects (presuming a common production volume for all cost shares, as per Eq. (4.2)). Hence, cost shares with higher learning rates are more strongly affected by production changes than those with lower learning rates. Consequently, this leads to a change in the initial cost structure along the experience curve (see Figure 4.1), where components with higher (positive) learning rates become less important to the overall module costs. Under the assumption that the defined learning rates represent real cost effects and remain constant, a detailed analysis of the development of the cost structure along the experience curve allows for the identification of cost-relevant components at every stage of product deployment.

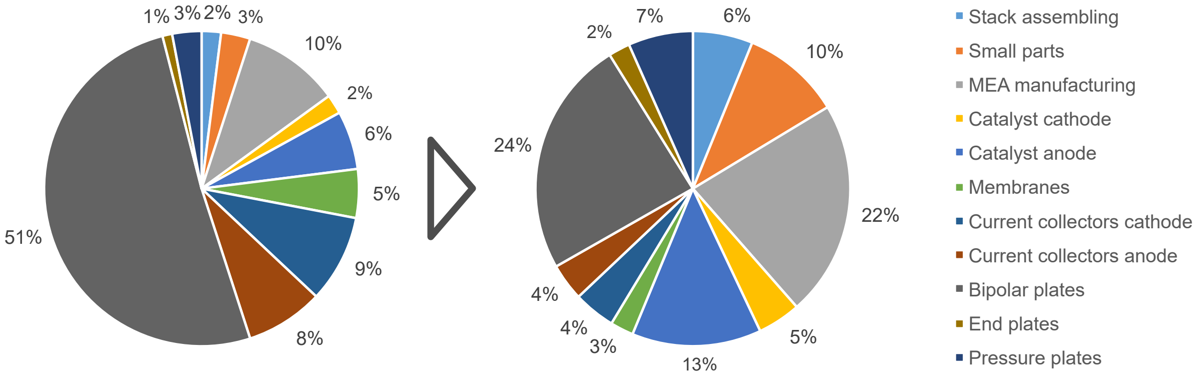

Figure 4.1: Development of the cost structure of PEM electrolysis cells for different cumulative amounts of produced units (left: initial; right: factor 1,000). Source: Böhm, Goers, and Zauner [79]

Another effect induced by the change in the module cost structure is observed in the accumulated learning rate of the module. In contrast to the constant learning rates set for the individual components, the overall learning rate decreases with increasing cumulative production because of the decreasing cost impact of components with high learning rates. This effect could potentially describe the difference in learning rates that are often observed for technologies over different stages of development and product maturity (e.g., for residential PEMFC in Japan [133]).

4.1.1.4 Impact of spillover effects

Another important effect revealed by the experience curve model is the impact of the spillover effects from the concurrent usage of peripheral system modules. As discussed in Section 4.1.1.2, a relation between all learning effects and the change in cumulative productions of the main technology can lead to a significant misinterpretation of learning effects, which results in a disregard for additional cost reductions. As shown in Figure 4.2, the relation between the learning effects of common peripheral parts and the relative increase in cumulative production of a novel technology leads to an overestimation of cost effects. Since doubling of cumulative production, and thus cost changes by the means of the learning rate, occurs faster (and more often) at low initial production volumes for the novel technology, than they do at an established stage for common technologies, a distinct relation of production increase to the individual basis is important. This represents the past *learning capital* of established technologies. Figure 4.2 shows the described potential for overestimation of learning effects for the different electrolysis technologies. It must be noted that the impact is significantly higher for early-stage technologies (SOEC) than for mature technologies (e.g., AEC).

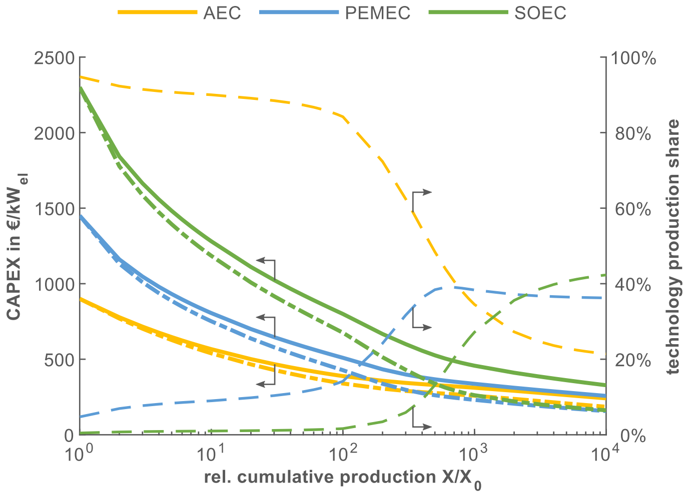

Figure 4.2: Impact of spillover effects on the calculated experience curves for electrolysis systems. Source: Böhm, Goers, and Zauner [79]

Note:
solid lines ... experience curves according to proposed module structure and dependencies;
dash-dotted lines ... experience curves without considering spillover effects (only 'direct' module dependencies);
dashed lines ... experience technology production shares.

4.1.2 Application of scaling factors

Along with the evaluation of the experience curve effects, the disaggregation of the investigated applications also allows for a more practical consideration of the unit-scaling effects. Even though, the relevant level of disaggregation may differ between the use for technological learning and size-related cost effects – the former is usually more targeted to savings for individual materials and processes, while the latter is more related to self-contained appliances and components – component structure data can still be widely reused or derived from basic process diagrams. Hence, common scaling methods as per Eq. (3.11), can be and are also applied to a disaggregated component structure rather than on complete systems.

Appropriate scaling exponents for relevant components are often found in dedicated literature, such as, for example, in chemical engineering guidelines. However, as discussed in Section 3.2.2.2, these scaling exponents are typically static and limited to a certain range of validity. Additionally, certain technologies and components may be limited to certain scales, especially during the early development stages, whereas these limits often evolve. For instance, electrolysis stacks are expected to be limited in size, or rather in their nominal power per unit, for various reasons (e.g., problems with leakage); thus, they are expected to be scaled by modularity (scaling by numbers) [47, 113]. To account for these limitations, a dynamic scale factor is implemented as follows:

$$f = 1 - (1 - f_0)\, e^{-\frac{S}{S_{max}}} \tag{4.6}$$

Here, f_0 represents the basic scaling exponent, S is the question scale, and S_{max} is the average maximum scale for the considered period. This provides a scale factor that is dependent on the system scale itself and reduces the scaling effects for large-scale applications. This approach was used to evaluate the costs of future large-scale PtG applications, such as those performed in [27, 47, 59, 134].

4.1.3 Demand potentials for power-to-gas technologies

Along with the experience curve model and the corresponding learning parameters, an evaluation of the actual market and deployment potentials of the technologies is required to assess the appropriate cost reduction potentials. As analyzed and discussed in Böhm *et al.* [47], there are numerous studies on the future potential and requirements for PtG technologies in the course of the energy transition by 2050. However, it has also been found that they differ significantly with regard to the included sectors, regional settings, and scope, by which they are hardly comparable. Thus, a deduction of the overall demand potential for PtG technologies based on these was found to be unreasonable, especially for global applicability, which would be required for a comprehensive analysis of technological learning. Therefore, the executed assessments were limited to future industrial hydrogen demands, as they were found to be less disputed in terms of predictability and renewable alternatives to today's supply.

According to a recent IEA report [7], the global hydrogen demand related to about 90 $\mathrm{Mt_{H_2}}$ by 2020, whereof the vast majority is utilized in industrial applications. Among these, the top single uses are in oil refining (43 %), ammonia (35 %) and methanol (13 %) production, and direct reduction in steelmaking (5 %). Even though future demands for oil refining (despite still increasing) could be expected to decrease following general de-fossilization, the overall demand of the other sectors is expected to increase. This has a high potential for renewable hydrogen production, especially since today's production is almost exclusively fossil-based (76 % from natural gas, 23 % from coal) [5].

A significant increase in hydrogen demand is expected from ammonia production, which is less driven by the current fertilizer production demand, expected to increase by 1.7 % per year until 2030 [5], but more by the increase in other industrial uses of ammonia, which currently accounts for only 10–20 % of the global demand [135]. For methanol, production is expected to increase by 50 % until 2030 and almost double by 2050 relative to today's amounts because of the increasing use of methanol as a fuel additive and additional demands as a base chemical for the renewable production of HVC [135]. Accordingly, the total hydrogen demand for the chemical industry is expected to result in >96.4 $\mathrm{Mt_{H_2}}$ per year in 2050, following current utilization pathways [5]. Based on an average electrolyzer efficiency of 70 $\%_{\mathrm{LHV}}$, this would result in a total capacity of 4590 $\mathrm{TWh_{el}}$ to be produced from renewable electricity.

For the iron & steel industry, the application of DRI, which is currently utilizing about 4 $\mathrm{Mt_{H_2}}$ per year, is expected to increase significantly as being widely discussed as the most promising path to significantly reduce emissions of primary steel production. By 2030, the annual demand is expected to reach 8 $\mathrm{Mt_{H_2}}$ or even 9–11 $\mathrm{Mt_{H_2}}$ for zero-emission scenarios. However, a more significant increase is expected in the long term, reaching 62 $\mathrm{Mt_{H_2}}$, or 47–67 $\mathrm{Mt_{H_2}}$ for zero-emission scenarios per year by 2050 [5]. Assuming a hydrogen supply from renewable electricity, an additional electrolyzer capacity of 2050–3000 $\mathrm{TWh_{el}}$ is required.

For the future demand for methanation technologies in terms of PtG, predictions are sparse because their role in future energy systems is disputed [9]. However, Pleßmann *et al.* [136] provide an estimate of the global PtM storage demand potential for 100 % renewable electricity supply, relating to 2360 $\mathrm{GW_{el}}$ or 1690 $\mathrm{TWh_{SNG}}$. Despite the limitations of the study according to the simplifications of the model used, these values were further used as a reference for the assessed technological learning effects for PtM.

Based on these demand potentials, three deployment scenarios were defined for the assessment of experience curves for PtG technologies during the course of this thesis (see Table 4.1). To relate the given electrolysis capacities for the industrial demands to the electric input power, a cost-optimized operation at 3500 annual FLH, as found in [123] and [5], was assumed. For the PtM power-related demand, it was differentiated between the decoupled operation of electrolysis and methanation (e.g., by using intermediate storage) at 7000 FLH for the methanation part and a direct transformation consistent with electrolysis. The corresponding capacities were assumed to be the target values for 2050 for the individual scenarios. Implementation development was calculated using logistic growth functions (see also Böhm *et al.* [47]).

4.2 Assessment of product generation costs

While various indicators for the evaluation of techno-economic performance are known, levelized product generation costs were found to be the most suitable comparative parameter for a generalized assessment of product-related energy technologies, such as PtG, in terms of providing a substitute energy carrier. In addition, it allows benchmarking with alternative forms of energy, such as electric energy costs. Therefore, the evaluations performed in Böhm *et al.* [47] and Böhm, Lehner, and Kienberger [27] used an approach comparable to the calculation of *levelized*

Table 4.1: Investigated deployment scenarios for power-to-gas applications until 2050 as defined in Böhm *et al.* [47]

Scenario	Electrolysis capacity in GW_{el}	Methanation capacity in GW_{SNG}	Notes
Low	1310	0	• PtH_2 for chemical industry only • No PtM
Moderate	3670	280	• PtH_2 for chemical industry only • PtM with intermediate H_2 storage
High	4530	1360	• PtH_2 for chemical & steel industry • PtM without intermediate H_2 storage

costs of energy (LCoE) [137, 138]. The assessed total annual plant costs (incl. operation) are related to the energy content of the product. To calculate the total annual plant costs, the annuity method following VDI 2067 [139] was used. The annuity of the total annual payments A is defined as the difference between the annuity of proceeds A_P and the sum of the annuities of capital-related A_C, demand-related A_D, operation-related A_O, and other (miscellaneous) costs A_M.

$$A = A_P - (A_C + A_D + A_O + A_M) \tag{4.7}$$

Variable costs (and proceeds) C_{var}, which are dependent on the input and output streams related to the time of operation (e.g., electricity and CO_2 input costs, proceeds from by-products such as oxygen or heat), are added according to their occurrence. The levelized costs of product (LCoP), representing, with regard to PtG, the generation costs for hydrogen and SNG related energy content, are then calculated as follows:

$$LCoP = \frac{-A + \sum_i C_{var,i}}{P_{out}} \tag{4.8}$$

4.2.1 Capital and (fixed) operational costs

The capital-related costs mainly include investment costs for system components and expected replacements during the observation period, as per Eq. (4.9):

$$A_C = (I_0 + I_1 + \cdots + I_n - R)\, a \tag{4.9}$$

where I_0 represents the initial investment amount, $I_1, \ldots, I_n$ are the cash values of the first to n-th procured replacement (4.10), R is the residual value of the investment (4.11), or last replacement, respectively, and a is the annuity factor (4.12). Replacements and the residual value are hence dependent on the interest rate factor q and deprecation period T_N.

$$I_1 = \frac{I_0}{q^{1 \cdot T_N}} \quad \ldots \quad I_n = \frac{I_0}{q^{n \cdot T_N}} \tag{4.10}$$

$$R = I_0 \frac{(n+1)T_N - T}{T_N \cdot q^T} \tag{4.11}$$

$$a = \frac{q^T(q-1)}{q^T-1} \tag{4.12}$$

In addition to the direct capital costs of the dedicated main equipment (e.g., electrolyzers, compressors, storage tanks, and methanation reactors), the annuity of additional costs A_M is usually considered by the use of overhead factors, which are known as *Lang* or *Chilton factors* and found in the literature [100, 101, 140].

Operation-related costs include maintenance and inspection costs O_M, as well as insurance and administration costs O_I. The corresponding annuity is calculated as:

$$A_O = (O_M + O_I)a \tag{4.13}$$

Demand-related costs, described by their annuity A_D, are related to their occurrence and included in $\sum_i C_{var,i}$ and thus not considered as per Eq. (4.7).

4.2.2 Variable costs and proceeds

As described above, $\sum_i C_{var,i}$ subsumes all variable costs for consumable supplies, as well as the potential proceeds from byproduct sales. The most relevant streams related to PtG processes are discussed below.

4.2.2.1 Electricity supply costs

Electricity supply is the most relevant input for the electrolysis process. However, the corresponding electricity supply profiles and costs largely depend on the underlying source of electricity. With regard to the integration of PtG in future renewable energy systems, this technology is often discussed as a storage option, especially in terms of seasonal storage to transfer excess energy (e.g., from PV) from summer to winter [141, 142]. However, to directly operate electrolysis or PtG facilities under fluctuating electricity generation scenarios, such as wind or PV power plants, appropriate flexibility of the applied technologies is required. In this context, some technologies are better suited than others (see Section 3.1). This circumstance must be considered in the TEA, i.a. for a reasonable consideration of attainable full-load hours of operation. A possible method for integrating the dependence of the presumed minimal consecutive times of operation and non-operation applied to the synthetic generation profile of a wind power plant was shown and discussed in [27]. In terms of the electricity supply costs involved, LCoE is a common method to assess the generation costs of electric energy. Considering the globally increasing number of renewable power plants being installed in tandem with the energy transition process, the corresponding LCoE for these technologies are expected to decrease as well because of the economies-of-scale effects. For instance, the IEA regularly publishes appropriate forecasts for LCoE development of different electricity generation technologies for selected regions in their annual World Energy Outlook [143] (see Table 4.2), which can be used as a guideline for cost assessment. However, especially in terms of the exploitation of excess energy, it must be considered that an operation with peak production probably has to be evaluated differently.

Concerning the application of PtH_2 and PtM for industrial demands or a broad supply of renewable gases, an operation with a direct electricity supply from fluctuating and/or

Table 4.2: Projected LCoE development in the European Union. Excerpt for two scenarios as published in the IEA World Energy Outlook 2021 [143]

	LCoE in USD/MWh					
	Sustainable Development Scenario			Net Zero Emissions by 2050 Scenario		
	2020	2030	2050	2020	2030	2050
Nuclear	150	120	115	150	120	115
Coal	200	n.a.	n.a.	250	n.a.	n.a.
Gas CCGT	95	120	n.a.	100	150	n.a.
Solar PV	55	35	30	55	35	25
Wind onshore	50	45	45	55	45	40
Wind offshore	75	45	30	75	40	25

Notes: CCGT = combined-cycle gas turbine; n.a. = not applicable

intermittent RES, and thus low annual full-load hours, becomes less viable. Therefore, operation at high annual loads and, consequently, a constant supply from (public) electricity grids is also reasonable. However, despite a more homogeneous supply profile, the use of market-related electricity comes with additional difficulties in terms of cost assessment. On the one hand, the considered electricity markets and products (e.g., spot market vs. long-term contracts) have a (potentially significant) impact on the corresponding supply costs (cf. [123]). Further, it may also govern the supply profile, at least for theoretical assessments, for instance, if the operation is presumed to prefer the lowest hourly spot market prices (based on historic market data; cf. [27, 47]). On the other hand, cost projections, particularly for long-term forecasts, are not feasible. This is attributed to the variety of external impacts, such as interacting markets, dependency on resource and fuel prices and availability, changing regulations, shares and availability of RES. These uncertainties can also significantly impact short-term projections, which was just recently confirmed by rapidly increasing electricity (and natural gas) prices at the end of 2021 and beginning of 2022 (see Figure 4.3). Therefore, for the assessments performed as part of this thesis, calculations of dynamic operations related to grid electricity supply utilize historic market data, such as EXAA spot market data [144].

4.2.2.2 CO_2 supply costs

In terms of PtM (or PtX), the required carbon source is probably the second most relevant input stream for the PtG process. Even though CO_2 is widely understood as an (unwanted) waste stream, especially as content of combustion off-gases, its consideration as a freely available resource is questionable. As shown in Figure 4.4, which was elaborated as part of a comprehensive study on CO_2 potentials in Europe [145], common industrial processes (albeit generalized by sectors) may be considered reasonable sources for CO_2 for centralized PtX applications because of high point emissions. However, even after excluding the combustion of fossil fuels, the related carbon is mainly of fossil or geogenic origin. Therefore, sustainable applicability in terms of CCU requires consideration of product end-use (e.g., in closed carbon cycles, as discussed in [27]) and involved fixation periods for the utilized carbon. Furthermore, with regard to EU Emissions Trading System (ETS), the appropriate costs for emission certificates must be considered when assessing CO_2 resource costs for PtG processes. In this context, CO_2 from

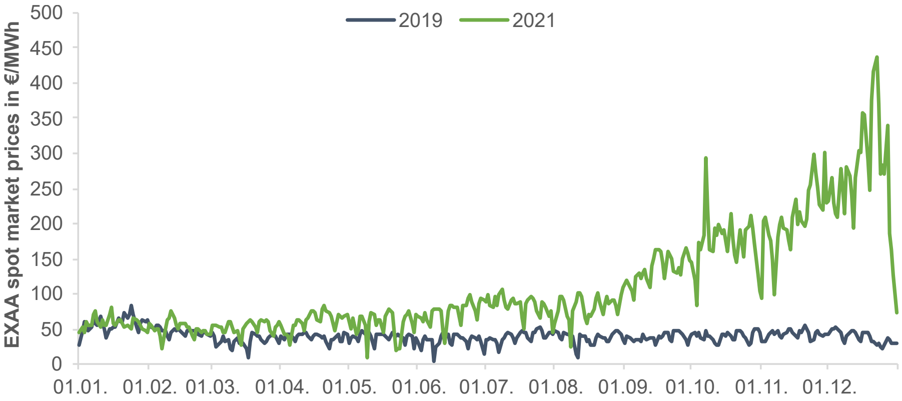

Figure 4.3: Comparison of spot market electricity prices (daily average) for the years 2019 and 2021. Source: EXAA Market Data

Note: The annual average spot market price for 2021 (109.02 €/MWh) exceeds the highest hourly price in 2019 (108.99 €/MWh).

biogenic sources, such as fermentation processes for bioethanol or biomethane, or combustion of biomass, is preferred for climate neutrality.

An additional factor for the utilization of CO_2 in the TEA of PtG processes is the effort required to capture and separate the CO_2 content from the source stream (e.g., combustion off-gas). Therefore, a literature review on the assumed carbon capture costs from different sources was performed as part of [145]. This analysis was primarily categorized by industrial sector (as referenced in EU ETS) and carbon-neutral (biogenic and atmospheric) sources. The results presented in Table 4.3 in a condensed form already show that corresponding costs to capture CO_2 from these individual sources cover relatively wide ranges. In particular, novel approaches, such as direct air capture (DAC), are naturally afflicted with high uncertainties. However, it is already apparent from the gathered data that efforts, and thus costs, for CO_2 capture are higher for processes with lower concentrations of CO_2 in the respective source stream, such as combustion off-gases in the energy sector (<20 vol%) or atmospheric air (≈400 ppm), than for highly pure streams, such as bioethanol or ammonia production (up to 100 vol%) [145, 146]. Furthermore, the lowest capture costs were found for biogenic sources. However, non-pure streams, such as biogas fermentation (≈40 vol%), also represent a wide cost range, going down as low as zero. This is also driven by the fact that it is not specified whether the costs for the separation of CO_2 are accounted for in biomethane production – due to this step being required to be allowed to feed into natural gas grids – or are being passed on to the subsequent utilizer of the CO_2 [32]. Additionally, with the direct utilization of raw biogas in the methanation process, separation costs can be omitted as well (see also Section 3.1.2). Considering these issues, the gathered data were used in the TEAs performed in the course of this thesis.

4.2.2.3 Proceeds from by-products sales

To optimize the economic performance of production facilities, it is reasonable to exploit the potential byproducts of the process. Oxygen and waste heat are potential byproducts of the

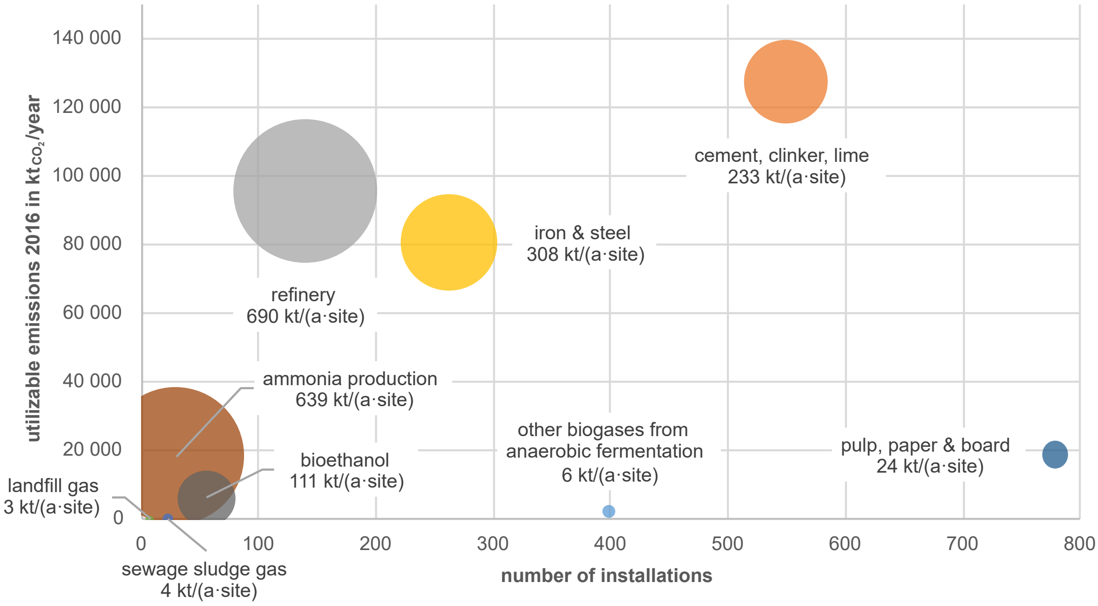

Figure 4.4: Technically utilizable CO_2 potentials from different industrial and biogenic processes. Source: Rodin *et al.* [145]

Note: The bubble area illustrates the average emissions per year and site for each process category.

PtG process. Even though they are only sporadically considered in the relevant literature and assessments (e.g., [130]), it has been found in industry expert interviews that a more comprehensive system integration by considering these value products is desirable [18]. In particular, oxygen, which arises as a byproduct of the electrolysis process in a significant amount (approximately $8\,\mathrm{kg_{O_2}/kg_{H_2}}$), can be considered a valuable resource because conventional production from cryogenic air separation (CAS) is conducted with high energy demands of approximately $250\text{–}300\,\mathrm{kWh/t_{O_2}}$ [147]. Therefore, previous studies have shown that prices range between $50\,€/\mathrm{t_{O_2}}$ [131, 148] and $150\,€/\mathrm{t_{O_2}}$ [149]. This can significantly impact the effective production cost of PtG plants, as discussed in [27].

Regarding the utilization of waste heat, an appropriate exploitation potential is difficult to assess. Depending on the technology, the corresponding temperature levels vary significantly (see Section 3.1). As discussed in Böhm *et al.* [18], the broad utilization of waste heat from commercially available low-temperature electrolysis systems (AEC and PEMEC systems) requires appropriate modern heating grids (4th & 5th generation district heating (DH)) to allow for the integration of temperatures below 100 °C. In contrast, the operating temperatures of high-temperature electrolysis are only of limited suitability to modern DH networks from an exergetic perspective. Consequently, high-temperature waste heat could preferably be utilized directly in industrial applications. Additionally, the actual availability and amount of waste heat for SOEC are highly dependent on its operating point. High potential for waste heat utilization is also involved in the methanation process, particularly chemical methanation with reasonable temperature levels. However, for an integrated PtM process, internal recuperation may be beneficial over coupling-out of the heat to increase the process efficiency when used in

Table 4.3: Average capture costs for CO_2 from different sources. Condensed representation from Rodin *et al.* [145]

CO_2 source		capture costs in €/t_{CO_2}
energy industry; power & heat	coal	19–63
	natural gas	35–101
	biomass	54–101
chemical industry	refinery	29–97
	ammonia production	12–54
	other chemicals	12–52
iron & steel production		19–83
cement, clinker & lime production		17–82
pulp, paper & board production		18–87
biogenic sources	biogas upgrading	0–90
	bioethanol fermentation	0–25
	bioethanol fermentation (incl. co-generation)	42–111
direct air capture (DAC)		18–475

combination with an endothermal electrolysis process [150]. Hence, even though the utilization of waste heat in PtG applications is desirable for overall efficiencies and potentially also comes with positive economic effects, its actual consideration in the TEA has to be addressed individually in the specific use case. Therefore, in the general assessments performed within the scope of this thesis, no external utilization of process waste heat was considered. However, the system analysis elaborated in [18] discussed the general waste heat potentials from low-temperature electrolysis arising from corresponding capacities in Austria and the EU projected in accordance with the national energy and climate plans (NECPs).

5 Results and discussion

In the following sections, the results of the analyses elaborated in this thesis and published in the related articles, are summarized and discussed. First, it includes the results of the evaluation of future costs of PtG technologies based on the application of the disaggregated economies-of-scale model. Furthermore, the induced effects on product generation are discussed with the systemic impacts and benefits of PtG technologies.

5.1 Learning rates for electrolysis and methanation

The learning curve model was calibrated for alkaline electrolysis using historic cost and production data published by Schmidt *et al.* [80] (resp. dataset [151]). The resulting learning rate for the main components of the AEC stack was found at a value of 18.04 %. A comparison of the resulting experience curve to a conventional OFLC applied to the historic data revealed no significant differences between the two approaches (see Figure 5.1). The coefficient of determination (R^2) shows that both approaches are fitting, hypothesizing that the proposed approach is at least as good as the common theory for the given data points (see Böhm, Goers, and Zauner [79]).

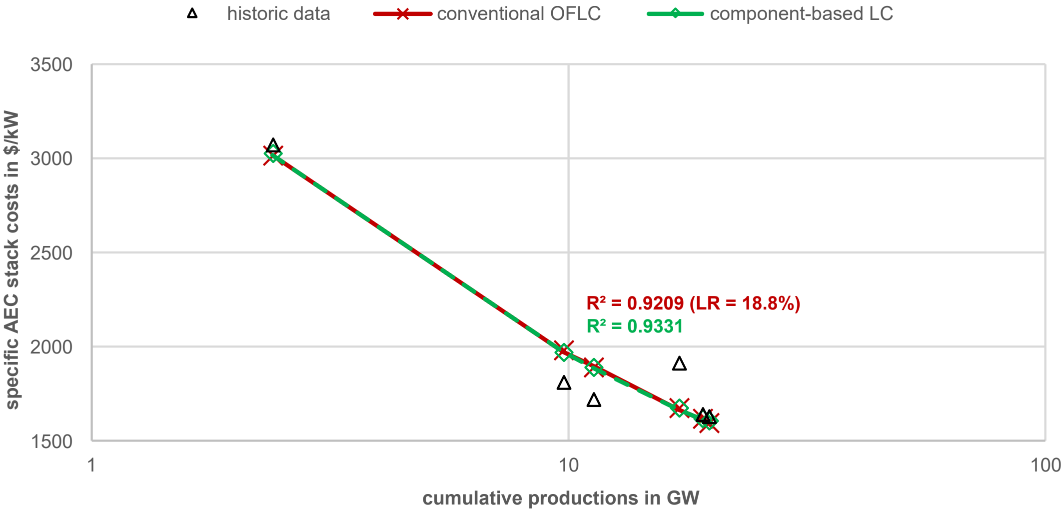

Figure 5.1: Comparison of the component-based and the conventional experience curve by fitting to available historical cost data. Source: Böhm, Goers, and Zauner [79]

Utilizing the methodological benefits of the disaggregated approach, learning rates were transferred per component to the PEM and solid oxide electrolysis stack models. Therefore, the initial learning rates are related to 19.3 % (AEC), 17.4 % (PEMEC), and 20.5 % (SOEC) for the individual electrolysis technologies, including the additional effects induced by individual *learning properties*. Compared to the evaluations in the literature (cf. Section 3.2.2.3), these learning rates are comparable, though at the lower bound of the given ranges, especially for SOEC. In addition, because of the effects of changing cost structures (see Section 4.1.1), these learning rates show a decrease with increasing amounts of cumulative production, and this decrease is steeper for technologies with higher initial rates. From a system-level perspective, the resulting learning rates are highly impacted by spillover effects (see Figure 4.2), and thus vary significantly according to the presumed technology production shares and overall production [79].

For the investigated methanation technologies, the learning rates evaluated in Böhm *et al.* [47] were less diverse owing to the lower number of considered subcomponents. Therefore, the rates also only decrease slightly for the presumed range of future production volumes – from 12.1 % to 11.7 % (chemical) and from 12.3 % to 11.8 % (biological), respectively. Despite the lack of specific comparative literature data for methanation learning rates, these values are at least comparable to the corresponding values found for roughly comparable technologies, as stated in Section 3.2.2.3.

5.2 Future technology costs of power-to-gas

Based on the discussed disaggregated experience curve model and the fulfillment of the proposed deployment potentials, the cost development for the relevant PtG technologies (electrolysis: AEC, PEMEC, SOEC; methanation: chemical, biological) was calculated for a reference scale of 5 MW (related to electric input for electrolysis and SNG output for methanation)[1]. The resulting experience curves for electrolysis technologies, as shown in Figure 5.2, suggest a significant cost reduction for all technologies under given conditions. The highest cost reduction potential was found for high-temperature electrolysis, which is partially related to the higher learning rate compared to the other technologies (cf. Böhm, Goers, and Zauner [79]). It is, however, mainly driven by the initially low amount of cumulative production (0.1 $\mathrm{GW_{el}}$ presumed for 2018) and thus high learning potential. Therefore, this technology is expected to reach input-related cost levels comparable to those of alkaline electrolysis. In contrast, PEM electrolysis is about to outperform AEC cost-wise in the coming years, owing to a comparably steep experience curve, despite a relatively lower learning rate. Hence, the cost reduction for this technology is also driven by the high relative increase in cumulative production, as well as by the higher cost share (60 %) of the stack module on the overall PEM system costs.

Furthermore, Figure 5.2 shows that scenario-related uncertainty is mostly defined by the difference between the *low* (upper bound) and *moderate* scenarios, representing an increase in capacity by a factor of 2.8. Further increase in the *high* scenario (lower bound), representing a factor of ≈1.2, is negligible. In addition, this effects that the impact of the presumed production shares is insignificant for a certain overall production volume, as shown in detail in [79]. The curves to reach the targets set in the EU hydrogen strategy [8] added in Figure 5.2 suggest that the respective cost reductions could be reached even earlier if these targets are achieved. Moreover, these values only consider EU targets compared to otherwise global volumes.

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[1] The 5 MW reference scale is used as a representative for both technologies individually. Considering the overall efficiency chain of the PtG process, 5 $\mathrm{MW_{el}}$ is approximately 2.5 $\mathrm{MW_{SNG}}$ for currently available technologies.

However, even though the logistic functions defined for the investigated scenarios already result in relatively high compound average annual growth rates (CAAGRs) (see Böhm *et al.* [47]), the fulfillment of the EU targets supposes significantly higher expansion rates, especially in the short term and thus the following years.

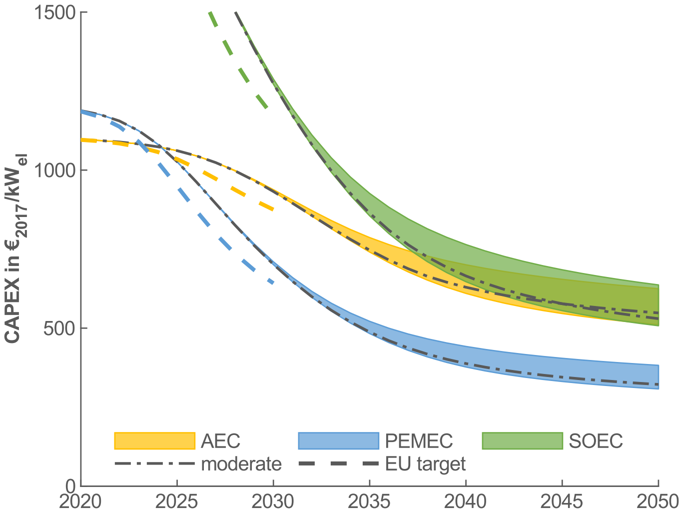

Figure 5.2: Estimated ranges for technological learning of electrolysis for the defined deployment scenarios related to electric input power. Adapted from Böhm *et al.* [47] (supplemented by EU target curves).

Note: The EU target curves use the capacity values stated in the EU hydrogen strategy [8] as fitting points for 2024 (6 GW$_{el}$) and 2030 (80 GW$_{el}$) in the growth function.

While the electric-input-related representation in Figure 5.2 shows a clear preference for PEMEC and AEC in the short term and PEMEC in the long term, this becomes less apparent if additional techno-economic aspects are considered. Figure 5.3 shows the same experience curves but related to H_2 output under consideration of electric efficiencies and their development for the individual technologies. In this context, SOEC is expected to outperform AEC from the long-term cost perspective. Although long-term CAPEX are still expected to be higher than for PEM electrolysis, additional advantages can be gained in combination with downstream processes, such as methanation. In particular, the application of co-electrolysis has the potential to achieve an optimal combination of synthesis gas composition and thermal integration of subsequent methanation [25–27].

The experience curves evaluated for methanation show similar trends, even though they are flatter than those of electrolysis. This is primarily caused by moderate average learning rates of approximately 12 %. Additionally, the presumed cost share of the core modules, namely the reactor, being higher for biological methanation and the relatively low learning rate for the catalyst in the chemical process, leads to a long-term cost advantage (CAPEX-related) for the biological process. However, both technologies are expected to be competitive from a cost perspective over the entire observation period (cf. Böhm *et al.* [47]).

In addition to the experience curves for the 5 MW reference scale, the cost effects to be expected for large-scale implementations were analyzed by considering appropriate unit-scaling

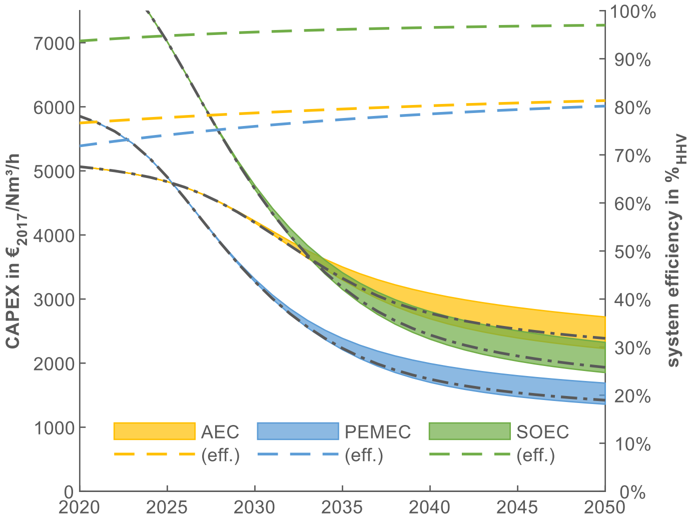

Figure 5.3: Estimated ranges for technological learning of electrolysis for the defined deployment scenarios related to hydrogen output with developing efficiencies. Source: Böhm *et al.* [47]

effects. Figure 5.4 shows the corresponding results for the investigated electrolysis technologies. It can be seen that the calculated effects vary between technologies, which is primarily the result of the disaggregated application of scaling factors and individual cost characteristics. While all technologies show strong scaling effects around the reference scale, these effects decline relatively rapidly for AEC and PEMEC for higher scales. In comparison, the dependency of CAPEX on the system scale is more significant for SOEC. This effect is caused by the higher cost share of the peripheral modules (power electronics, gas conditioning, balance of plant (BoP)), which are expected to show better scalability than the stack module scaled by modularization (scaling by numbers) [113]. Furthermore, scaling effects are found to increase with later implementation time because modules with lower unit-scaling potentials, namely stacks, are more affected by technological learning. Thus, their impact on unit scaling increases over time or, respectively, with increasing cumulative production. Compared to the available literature, it was found that for AEC and PEMEC, unit-scaling effects seem to be overestimated in relevant studies for larger scales (>50 MW). In contrast, comparable values found for SOEC confirm the evaluated effects of unit scaling quite well, while they seem to overestimate technological learning. However, this was somewhat expected given the high uncertainty of the learning rates for SOEC found in the literature (28 ± 16 %) as discussed in [79] (see also Section 3.3). Hence, the applied component-based learning model was more conservative in this context.

Unit-scaling effects evaluated for methanation are generally more significant than for electrolysis technologies, mainly based on the lower scaling factors for the individual components, as found in the literature (see Figure 5.5). However, it was also found that the scale factors of <0.6, as stated in certain literature [35, 152], were quite low for the overall methanation process. Therefore, it must be assumed that these scaling effects are accompanied by technological learning without differentiation. Comparing the calculated cost curves in 5.5 with relevant literature estimates, the corresponding values were found to fit quite well for long-term (2050) costs, especially for chemical methanation routes.

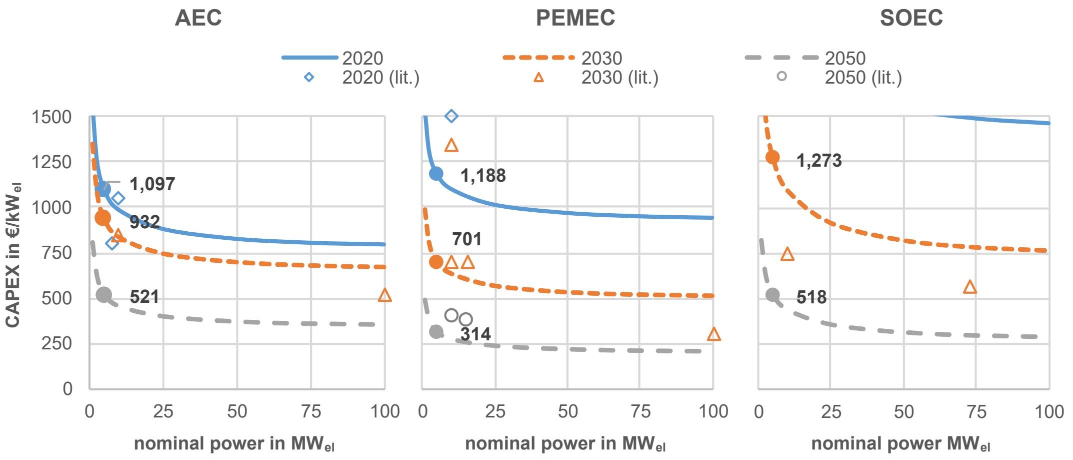

Figure 5.4: CAPEX development for electrolysis systems compared to projections from the literature study (lit.) for installations ≥10 MW$_{\mathrm{el}}$ (the marked and labelled values refer to the reference size of 5 MW$_{\mathrm{el}}$). Source: Böhm *et al.* [47]

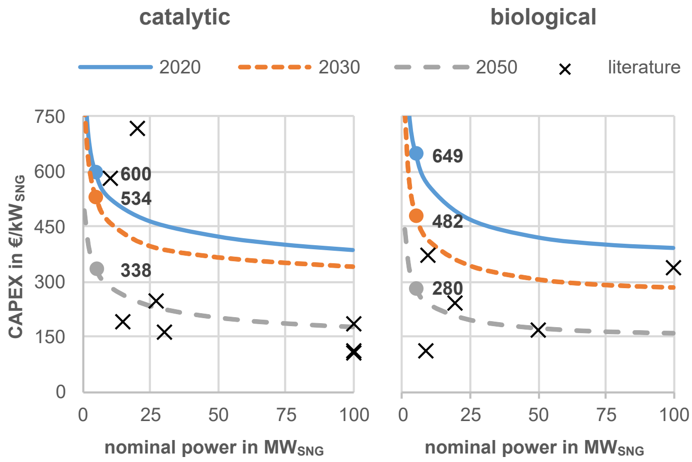

Figure 5.5: CAPEX development for methanation systems compared to projections from the literature review for installations ≥10 MW$_{\mathrm{SNG}}$ (no target time frames available; marked and labelled values refer to reference size of 5 MW$_{\mathrm{SNG}}$). Source: Böhm *et al.* [47]

Even though PtG plant scales of >50 MW appear to be a distant prospect from the current technology state, an effective application of these technologies for energy transition will require significantly higher scales. To put these scales in context: to adapt steel production from today's blast furnace (BF) to a DRI route, an electrolysis capacity of 3.5 $\mathrm{GW_{el}}$[2] would be required, solely for the largest Austrian steelwork. Regarding the methanation of (biogenic) industrial CO_2 sources, a conversion of the average annual per-site emissions of European bioethanol production (see Figure 4.4) would require a methanation capacity of approximately 90–180 $\mathrm{MW_{SNG}}$ and a respective electrolysis capacity of approximately 300 $\mathrm{MW_{el}}$ (presuming 3500–7000 h/a for methanation and 3500 h/a for electrolysis). Referring to Figure 4.4, for the methanation of emissions from fossil-based industries, the corresponding capacity demands are generally even higher.

5.3 Future costs of electrolysis-based hydrogen and SNG

Naturally, changes in CAPEX also affect the resulting value of LCoP. Consequently, the development of product generation costs was evaluated as part of [47], mainly for large-scale PtG applications up to 100 MW in general and in [27] for more specific PtM implementation scenarios. The results of both studies suggest that generation costs can be expected to be reduced by 75–80 % for PtG products in general in large-scale deployments, and even more for optimized and highly integrated applications. Hence, the long-term generation costs for hydrogen are expected to reach levels <5.5 c€/kWh, or <1.8 €/kg, respectively, which are consistent with recent projections from other studies (cf. Brändle, Schönfisch, and Schulte [122]). Similar results were found for PtM with cost levels down to <75 €/$\mathrm{MWh_{SNG}}$ [47] and <50 €/$\mathrm{MWh_{SNG}}$ for highly efficient applications[27], even though comparable studies are more optimistic in this context (cf. [123, 124]). However, as can be seen in Figure 5.6, which serves as an exemplary result, there are other cost factors that significantly impact LCoP – and their share, and thus impact, partially increases with decreasing CAPEX. The main cost factors, besides CAPEX and CAPEX-related fixed OPEX, are the costs of the electricity supply and potential proceeds that can be generated from byproduct sales.

5.3.1 Impact of electricity supply

As shown in Figure 5.6, electricity supply costs, and energy-related shares in particular, represent a relevant cost share on the overall LCoP of the PtG process, becoming even more significant when equipment costs decrease by economies of scale. However, as discussed in Section 4.2.2.1, the costs of electricity supply are widely dependent on the considered source of electricity and operating strategy. While the LCoE for renewables in 2020 were approximately 45–65 €/MWh[3] for Europe (see Table 4.2), the average spot market prices in Austria were approximately 40 €/MWh (≈38 €/MWh in DE; data for 2019), whereas hourly costs varied between −25 €/MWh and 109 €/MWh. However, an operation only at the lowest electricity cost is usually not viable, because the resulting FLH also affect LCoP in terms of levelized

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[2] Presuming a hydrogen demand of 51 $\mathrm{kg/t_{CS}}$ [153], an annual crude steel (CS) production of 5.05 $\mathrm{Mt_{CS}}$ [154] and an operation at 3500 h/a as per Section 4.1.3.

[3] Exchange rate USD/EUR=1.13

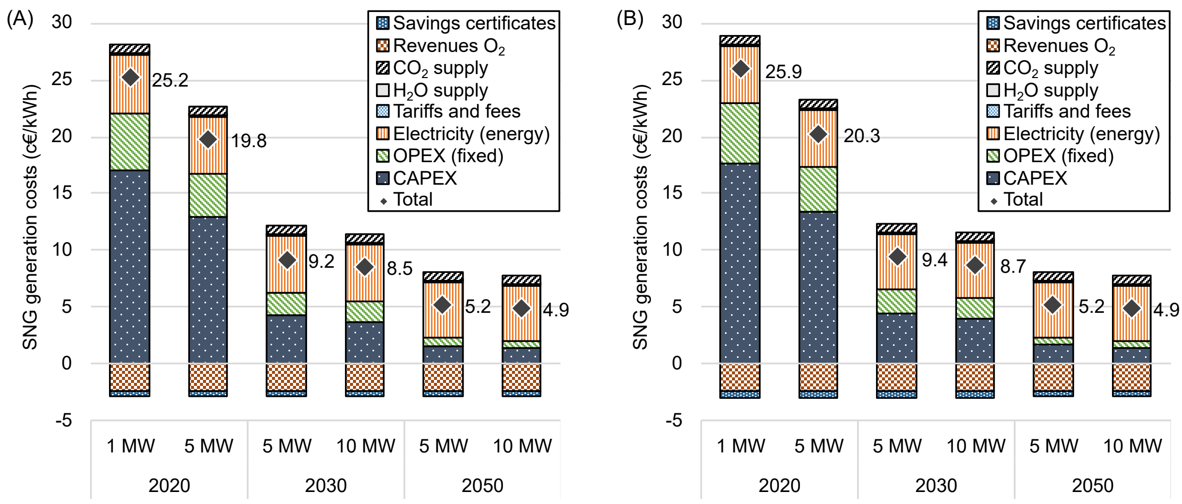

Figure 5.6: SNG generation costs for different plant capacities and years of implementation. (A) steel industry scenario, (B) cement industry scenario. Source: Böhm, Lehner, and Kienberger [27]

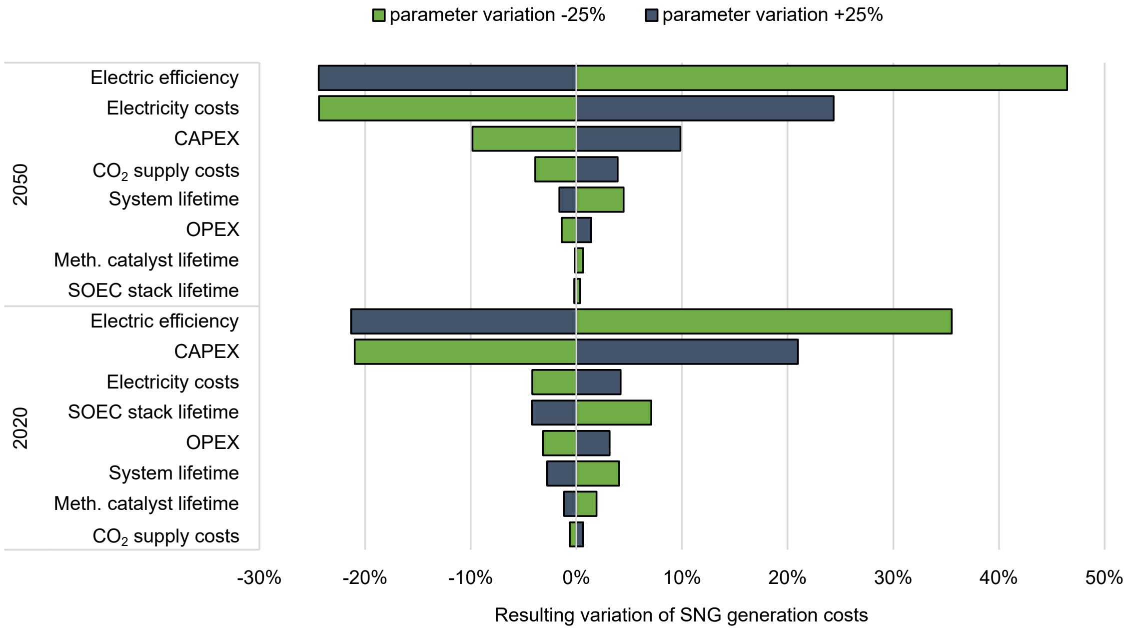

Figure 5.7: Impact of calculation parameter variation on the resulting SNG generation costs. Source: Böhm, Lehner, and Kienberger [27]

CAPEX. Therefore, even though LCoE for wind and PV are comparable or even slightly lower for PV, the overall generation costs for H_2 and SNG are generally higher for plants that rely solely on PV than on wind supply, as shown in Böhm *et al.* [47]. This is attributed to the lower amount of FLH attainable from PV power plants over wind farms. The results presented in [47] and [27] also show that an operation preferring only lowest market prices is not necessarily favored because high CAPEX in the short and medium terms result in a cost optimum for PtG plants at the highest possible annual FLH. However, both studies also showed that the FLH-related LCoP curves flatten out with decreasing CAPEX and thus potentially allow for cost-optimal operation at lower loads (4000–7000 FLH).

In addition to the general assessment of LCoP concerning specific electricity sources, the applicability of the PtG process in combination with the intermittent supply of renewables was investigated. The results generated for an SOEC-driven process, which is often stated as inflexible owing to long start-up times (see Table 3.1), indicate that an operation targeted at the peak production of wind power plants is generally feasible. However, as the energy-related costs presented in Figure 5.8 show, a cost optimum solely related to electricity supply is only achieved in combination with grid supply. Especially for high-temperature electrolysis, an additional energy supply is often required to maintain temperatures in hot-standby operation bridging (short) timespans without supply. However, because an economically viable operation is not reached for low loads supplied by PV or wind (<3000 FLH per year), this type of operation is mainly considered to be beneficial in terms of grid-supportive integration of PtG plants.

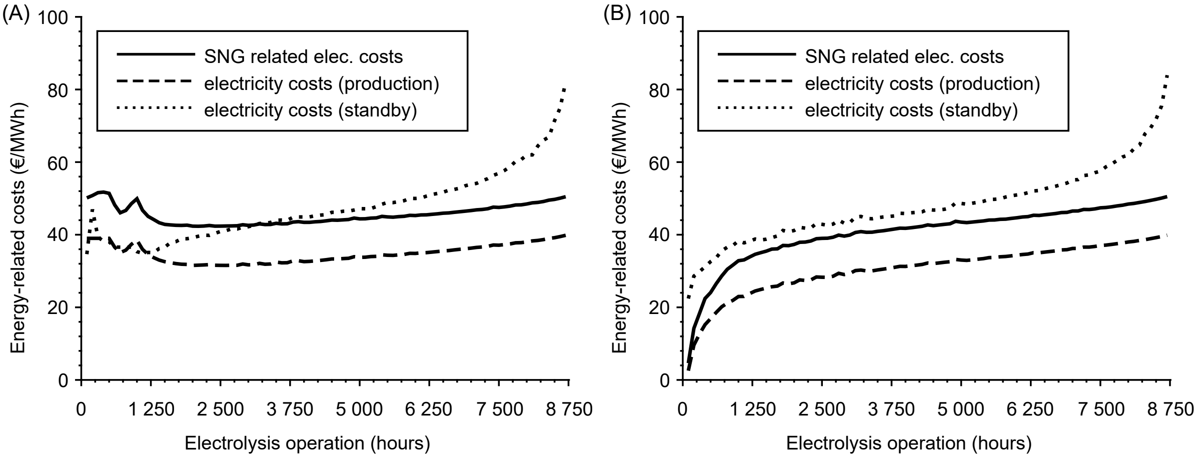

Figure 5.8: Electricity supply costs and electricity-related production costs for (A) hybrid electricity supply and (B) grid-only electricity supply. Source: Böhm, Lehner, and Kienberger [27]

Concerning the high impact of electricity supply costs on LCoP, especially in the long term, a major challenge and research topic for PtG applications is regarding increasing the electric efficiency of the process. This is also found in the corresponding sensitivity analyses performed in the individual studies, revealing that electric efficiency is the most significant impact factor over all scenarios (cf. Figure 5.7). As outlined in [27], the consequent exploitation of synergies between electrolysis, downstream synthesis, and surrounding processes is considered an important aspect of future commercial implementations of PtG applications.

5.3.2 Impact of byproduct utilization

Beyond the absolute costs of hydrogen and SNG production, the effective generation costs can be lowered by selling potential byproducts. The most relevant byproduct of the PtG process is oxygen generated during the electrolysis of water. As discussed in 4.2.2.3, the actual estimates of attainable prices for selling oxygen vary significantly. Even with the more conservative price used in [27], the impact on the effective generation costs is found to be relevant, especially in terms of overall decreasing generation costs, as depicted in Figure 5.6. Furthermore, the direct utilization of oxygen can potentially be beneficial for various processes that favor the implementation of PtH_2 or PtM processes (e.g., steel production or oxyfuel combustion as discussed in [27]). In this context, apart from the oxygen supply costs, utilizing appropriate synergies could significantly reduce the overall energy demand by omitting separate energy-intensive oxygen generation (e.g., by CAS).

Another relevant byproduct of the PtG process is the waste heat produced during electrolysis and methanation. While this waste heat can (partially) be reutilized within the PtG process for certain applications to optimize the overall system efficiencies (see [27]), other utilization pathways should be considered if internal use is not applicable. However, the economic impacts of heat utilization were not directly addressed in the TEAs within the scope of this thesis.

5.4 Further systemic considerations

In addition to economic considerations, some additional systemic perspectives of PtG in future energy systems are discussed as part of the assessments along with this thesis. In this regard, the potential synergies of PtH_2 with the heat supply of DH systems were discussed in the systemic analysis performed in Böhm *et al.* [18]. The results of this study suggest that even the waste heat from low-temperature electrolysis is sufficient to feed modern DH networks. The revealed potential for waste heat utilization from PtH_2 processes in Austria, which is expected from the implementation of electrolysis capacities in accordance with the NECP [155], relates to about 56–84 TWh_{th}. In relation to annual DH sales [156], 2–12 % of today's Austrian DH demand could potentially be supplied from renewable hydrogen production by 2030. Applying these calculations to an EU scope, 6–10 % of the DH demands could be covered by electrolysis waste heat utilization. In addition, the industrial demands for space heat, hot water supply, and process heat at temperatures <100 °C represent an additional potential heat sink with capacities comparable to DH [157, 158]. Considering the primary industrial use of electrolysis-based hydrogen production, proper heat integration could supply a significant amount of this heat demand. Hence, the quantification of these potential synergies is essential for future research [18].

Furthermore, with the integration of CO_2, the PtM process also represents a form of CCU and thus a potential sink for CO_2 emissions. In this context, the reutilization of CO_2 from fossil sources practically only delays carbon emissions; it still results in a net flow of fossil carbon from geological reservoirs to the atmosphere [159]. Additionally, because of the primary use of methane, or SNG, as fuel, the effective fixation period for emitted CO_2 before transmission to the atmosphere is relatively short (weeks to months at best) [160]. Therefore, net emissions from carbon-based fuels, such as SNG from PtM, can only be avoided if the utilized CO_2 is either extracted from the atmosphere (via biomass or DAC) or kept constantly within the CCU process.

The latter is also termed *closed carbon cycle* and still presumes a constant carbon budget, that is, no additional use of fossil fuels, to achieve climate neutrality. However, establishing such *closed carbon cycles* may still be a transitional option for industrial applications in hard-to-abate sectors, as discussed in [27]. However, the analyses performed in [27] have also shown that the current treatment of carbon costs – basically CO_2 pricing via EU ETS and other regulatory measures – does not favor the implementation of such net-zero emission CCU processes (see Figure 5.6 for the impact of current CO_2 certificate costs on the effective SNG generation costs). As outlined in [27], carbon prices must reach $150\,€/t_{CO_2}$ for long-term technology costs, and up to $330\,€/t_{CO_2}$ in the medium term, for carbon-neutral PtM processes to be competitive with natural gas – related to an average gas price of about 25 €/MWh as per the first half of 2021[4]. However, this also shows that early implementations could benefit from long-term savings if carbon costs reach appropriate levels. Furthermore, recent increases in natural gas prices in Europe by a factor of four in by the end of 2021, up to a factor of ten due to the war in Ukraine in early 2022 [161], have shown that the competitiveness of PtG is also dependent on external impact factors with corresponding volatility. In this context, in addition to the need for renewable hydrogen production, which is to a large extent undisputed, the utilization of CO_2 from biogenic sources or DAC and the prevention of transition costs for infrastructure already in existence may still lead to PtM becoming a sustainable cornerstone of the energy transition.

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[4] The EU average price for non-household consumers (excluding taxes) was derived according to [162].

6 Summary and conclusions

The need for new technologies to provide renewable energy carriers and base materials as a part of the overall energy transition is evident. Furthermore, although the role of gaseous energy carriers as a part of this transition is still disputed, the demand for renewable hydrogen other than for energy use is expected to increase by 75 % or more until the year 2050. In this regard, power-to-gas (PtG), especially power-to-hydrogen (PtH_2), will contribute significantly to making these demands available in the short and medium term, as relevant technologies have already reached commercial maturity, are already available on an advanced scale, and are an integral part of today's national and international energy and climate plans all over the world. In addition, PtG can serve additional functions in future energy systems based on renewable energy sources (RES), such as providing storage capacities for extended storage periods (e.g., seasonal storage), peak shaving for fluctuating electric power generation, or simply replacing fossil energy carriers for energy (heat and electric power) supply, with a significant advantage regarding transport and storage due to high energy densities.

However, relevant decision-makers – authorities providing corresponding legal and regulatory frameworks, as well as implementors actually making the relevant investments – have to rely on available technology data for their decisions. To enable the optimal potential of the technologies and allow for early and targeted implementations, comprehensive and prospective assessments are required to estimate relevant developments that may have a significant impact on these decisions. Although, individual PtG technologies are classified as mature regarding their technology readiness level (TRL), substantial technical and economic developments are expected in the course of broad roll-outs and large-scale implementations. In this context, the techno-economic assessment (TEA) methods elaborated and applied within the scope of this thesis are aimed at identifying fundamental cost-decisive factors and potentially critical properties of currently relevant PtG technologies.

The potential for future cost reduction is commonly described by experience curves that represent the impacts of technological learning. However, the evaluation of corresponding learning rates requires the observation of cost developments over a few magnitudes of cumulative production for a technology, which is not applicable for relatively novel electrolysis and methanation technologies, apart from alkaline electrolysis, where the broad application of chlor-alkali electrolysis in the 20^{th} century can serve as a reference. Therefore, a disaggregated approach was used to allow for the interchangeability of component-wise learning rates between technologies, and thus the applicability of the experience curve theory for technologies with still limited amounts of cumulative production. It has been shown that the results are at least comparable to conservative one-factor learning curve (OFLC) models when applied to known historical cost-reduction effects. Despite the higher level of detail, the disaggregated model enables the consideration of learning effects at a much lower, and thus more comprehensible, level. Hence, learning rates can be assessed based on apparent cost effects, such as material savings, reduction in processing time, or certain adaptations in the processing of single components.

Furthermore, the importance of considering spillover effects for the concurrent usage of components shared between (related) technologies was shown. Since evaluations of experience curves for novel technologies often only consider learning effects for determining core parts, learning effects driven by the use of peripheral parts in other applications are widely disregarded. This is particularly relevant if technological learning is examined as a whole along with the production of core components, without considering additional production volumes (past and concurrent) for peripheral components. This may lead to a significant overestimation of the learning effects, which is higher when the initial amount of cumulative production of an impacted technology is lower. Consequently, the learning effects (costs) for lower-TRL technologies, such as high-temperature electrolysis, are considerably impacted by the experience gained with more mature counterparts, such as alkaline electrolysis systems. Additionally, these spillover effects lead to a reduction in the uncertainty in the experience curve regarding unknown future production shares between technologies. As shown, owing to the high expected demands, and thus, the production of PtG technologies in the individual categories (electrolysis and methanation), the impact on long-term cost reductions is low, even if the dominance of individual technologies is different from today's projections.

According to the estimated future demand of 1310–4530 $\mathrm{GW_{el}}$ for electrolysis and up to 1360 $\mathrm{GW_{SNG}}$ for methanation capacities by 2050, the elaborated experience curves show an induced cost reduction potential of 30–75 % (electrolysis) and 30–60 % (methanation), respectively, depending on the respective technology. However, in addition to these cost-reduction effects from *scaling-by-numbers*, additional effects can be expected from the upscaling of individual implementation capacities. Therefore, the disaggregated cost model enables a more precise application of unit-scaling techniques. Consequently, the overall cost reduction potentials derived from the scaling of the technologies are calculated to be >75 % for all investigated technologies, indicating an individual target capacity of 50 MW related to a current reference scale of 5 MW. The reduction in capital expenditures (CAPEX) naturally implies a reduction in hydrogen and synthetic natural gas (SNG) production costs generated from PtG applications. Based on the given production volumes, the production costs for hydrogen in large-scale applications are expected to reach values of 100 €/MWh (3.3 €/kg) and below. The additional conversion of the methanation process leads to long-term costs of approximately 150 €/MWh, considering the production driven by intermittent wind or photovoltaics (PV) supply. Even under the lowest cost considerations, the production cost for SNG is expected to remain above 80 €/MWh, which is significantly higher than the current cost for natural gas, without further optimization. However, investigations on specific implementation scenarios have shown that the utilization of synergy effects between PtG technologies themselves, as well as the facilities they are meant to be integrated into, contain substantial cost-saving potentials. Consequently, thermal integration of the exothermal methanation process and the endothermal operation of solid oxide electrolysis cell (SOEC) could undercut the aforementioned lower cost limit for SNG production in the medium term, and reach effective costs of <50 €/MWh in the long term. Additionally, decreasing CAPEX implies a shift in cost optimums from maximum annual system loads (>7000 h/a) toward lower and wider ranges of full-load hours (FLH) (3000–6500 h/a).

However, it was also shown that there are additional factors besides CAPEX and electric efficiency that significantly impact the effective levelized costs of product (LCoP) of PtG technologies. The most significant, especially in the case of decreasing CAPEX, are those for electric energy supply. Actual market prices for electricity are difficult to predict because of the interdependency of generally extended electrification and increasing RES (e.g., grid expansion, transborder markets, storage demands, etc.), but also based on recent short-term changes. However, because levelized costs of energy (LCoE) for RES are expected to generally decrease in the long term (cf. Table 4.2), and the executed investigations have shown the basic applicability of electrolysis technologies for direct integration with RES, the cost share of electric energy supply on the LCoP for PtG potentially decreases. Even more if grid-supportive operations, such as peak shaving or seasonal storage capabilities, are considered. At the same time, the cost of the fossil energy supply is expected to increase, thus increasing the competitiveness of PtG products. In addition to electricity costs, the costs and availability of CO_2 as a main input for methanation are often discussed in the context of PtG technologies. As the executed analyses show, the impact of CO_2 on SNG generation costs is minor. Furthermore, it was found that related costs for capturing tended to be lower for renewable sources, and the corresponding biogenic potentials were still far from being exhausted. In contrast, the potentials from individual point sources are higher for industrial sites and, thus, from fossil origin. From an ecological perspective, biogenic sources should be preferred for carbon capture and utilization (CCU) applications, especially for short fixation periods expected for fuel use. However, the assessments also proposed potential application scenarios for the utilization of CO_2 in closed carbon cycles, which can be economically viable while avoiding effluent emissions.

In addition, the economic viability of PtG applications depends on the utilization of synergy potentials. In addition to the exploitation of internal synergies, such as thermal integration of the processes, to increase system efficiencies, the sale of byproducts, especially oxygen, can have a significant impact on effective product generation costs. Furthermore, the utilization of waste heat should not be left unconsidered, not only from an individual economic perspective but also regarding primary energy efficiency, as electrolysis waste heat could provide substantial amounts of future heating demands below 100 °C in modern district heating (DH) networks. Consequently, future implementations of PtG capacities should be considered from a systemic rather than a per-application perspective to adequately address the sector-coupling potential of the technology. Therefore, PtG is expected to represent an important cornerstone of future energy systems, which is different from the exclusive purpose of substituting fossil energy carriers by providing storage and peak-shaving capabilities for renewable energy supply, and also reducing the existing energy demands for oxygen and heat that are supplied separately at present.

Finally, the elaborated studies and discussions show, that prospective TEA represents a valuable measure for evaluating the potential applicability and viability of early-stage technologies in future industrial implementation scenarios. Therefore, it is also important to be able to access appropriate and comprehensible methods to allow early and effective evaluation. In this context, the elaboration and application of assessment methods, such as component-based scaling effects and LCoE\LCoP, which are transferable to other technologies, was a major aspect of this thesis. Such similar methods will also be necessary for an early and smooth, rather than a late and steep, energy transition within the scope of the set climate targets.

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Publications

This section includes the following peer-reviewed articles, which constitute the major part of this doctoral thesis, in full text:

1. H. Böhm, S. Goers, and A. Zauner, “Estimating future costs of power-to-gas – a component-based approach for technological learning”, *International Journal of Hydrogen Energy*, vol. 44, no. 59, pp. 30 789–30 805, 2019. DOI: `10.1016/j.ijhydene.2019.09.230`
2. H. Böhm, A. Zauner, D. C. Rosenfeld, and R. Tichler, “Projecting cost development for future large-scale power-to-gas implementations by scaling effects”, *Applied Energy*, vol. 264, p. 114 780, 2020. DOI: `10.1016/j.apenergy.2020.114780`
3. V. Rodin, J. Lindorfer, H. Böhm, and L. Vieira, “Assessing the potential of carbon dioxide valorisation in Europe with focus on biogenic CO_2”, *Journal of CO2 Utilization*, vol. 41, p. 101 219, 2020. DOI: `10.1016/j.jcou.2020.101219`
4. H. Böhm, S. Moser, S. Puschnigg, and A. Zauner, “Power-to-hydrogen & district heating: Technology-based and infrastructure-oriented analysis of (future) sector coupling potentials”, *International Journal of Hydrogen Energy*, vol. 46, no. 63, pp. 31 938–31 951, 2021. DOI: `10.1016/j.ijhydene.2021.06.233`
5. H. Böhm, M. Lehner, and T. Kienberger, “Techno-economic assessment of thermally integrated co-electrolysis and methanation for industrial closed carbon cycles”, *Frontiers in Sustainability*, vol. 2, p. 77, 2021. DOI: `10.3389/frsus.2021.726332`

INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 44 (2019) 30789–30805

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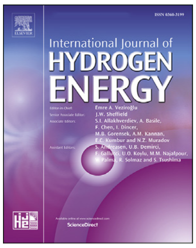

Estimating future costs of power-to-gas – a component-based approach for technological learning

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HIGHLIGHTS

- Approach to evaluate learning curves for technologies at low TRL.
- Analysis of learning effects on component level.
- Consideration of spill-over learning effects from concurrent technologies.
- Estimation on future costs of water electrolysis based on production volumes.
- Enabling interchangeability of learning rates.

ARTICLE INFO

Article history:
Received 7 June 2019
Received in revised form
24 September 2019
Accepted 28 September 2019
Available online 5 November 2019

Keywords:
Technological learning
Experience curve
Water electrolysis

ABSTRACT

Technological learning is a major aspect in the assessment of potential cost reductions for emerging energy technologies. Since the evaluation of experience curves requires the observation of production costs over several magnitudes of produced units, an early estimation of potential future technology implementation costs often presumes a certain degree of maturity. In this paper, we propose a calculation model for learning curves on the component or production process level, which allows to incorporate experience and knowledge on cost reduction potentials on a low level. This allows interchangeability between similar technologies, which is less feasible on a macro level. Additionally, the model is able to consider spill-over effects from concurrent technology usages for the inclusion of peripheral standard components for the assessment in an overall system view. The application of the model to the power-to-gas technology, especially water electrolysis, has shown, that the results are comparable to conventional approaches at the stack level, while providing transferability between different cell designs. In addition, the investigations made at the system level illustrate that the consideration of spill-over effects can be a relevant factor in the evaluation of cost reduction potentials, especially for technologies in an early commercial state with low numbers of cumulative productions.

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https://doi.org/10.1016/j.ijhydene.2019.09.230
0360-3199/

Introduction

The transformation of the energy system to facilitate the intensive use of renewable energies is based on the assumption that the technology costs concerning the generation, applications, and storage of renewable energy will decrease in the future. Especially in terms of the transition of the global energy system to renewable energy sources, the learning curve theory can be an important measure to estimate the learning investment, which is necessary for renewables to be competitive with incumbent technologies. Furthermore, this allows recommendations on regulatory frameworks as a driving factor for the enforcement of novel technologies [1,2]. In current models of the energy system analysis, technological progress is no longer considered exogenously but needs to be integrated into the model in the form of learning curves and thus endogenized.

Since common approaches for the estimation of learning effects, which define a single learning rate for a certain technology, require an observation of the production cost development over several magnitudes of cumulative production volumes, they are obviously unsuitable for emerging technologies on low TRL, which have not yet reached high market penetration. The approach in this study to investigate the effects of technological learning at a component or production process level allows circumventing these difficulties. It provides possibilities to incorporate experiences on direct production processes, while the component-level view enables interchangeability between different products and technologies with minimal adaptions. Furthermore, the stiffness of the learning curve following the conventional theory of constant learning rates at a macro level suspends the possibility for an easier adaptation of the learning curve to the various stages of technology readiness.

The first approach of exogenous technological progress is based on Schumpeter's invention-innovation-diffusion paradigm [3,4]. Based on this approach, invention implies the generation of new knowledge and ideas. Inventions are further developed and converted into new products, whereas diffusion covers the extensive implementation of new products. Further, Solow [5] qualified the inexplicable element of augmented productivity growth of the economy as technological progress. This technological change was mainly considered in the new macroeconomic endogenous growth literature and in the development of the learning curve concept in microeconomic analyses also including evaluations in the energy sector [6,7]. Later, the literature on economics, and energy and climate economics literature, focused on the topic of technical change. Particularly, the Stern Review of the economics of climate change [8] integrated assumptions regarding learning rates of technologies into its long-term cost projections. Learning rates are as important for technology analyses as discount rates are for cost-benefit analyses. Adapting the concept of learning curves of industrial production activities to innovation and technological development is a substantial step that involves consideration of the nature of innovation as a process [7]. Consequently, it is essential to evaluate the potential and restrictions of learning curves as an analytical tool in energy, technology, and policy analysis [6].

The production of hydrogen and synthetic natural gas from renewable electricity, also known as power-to-gas (PtG), is widely promoted as a promising solution in the transition of the energy system to renewable sources [9,10]. However, since maturity of technologies in that sector is low, specific costs for appropriate applications are still too high to be competitive in commercial applications [9,11]. Therefore, the evaluation of learning effects is relevant to estimate learning investment and predict future technology costs (cf. Fig. 1). Even though, learning curves for water electrolysis have been investigated in the past [12–16], these studies had to rely on limited data available for commercial technologies, like alkaline electrolysis cells, or the elicitation of expert opinions for newer technologies, like polymer electrolyte membrane (PEM) or solid oxide electrolysis cells (SOEC).

To be able to compare and transfer learning effects between different applications – within a shared field of application, like electrolysis, or beyond – these effects have to be analysed on a more detailed level. Hence, based on the deliberations of Ferioli et al. [17] and Tsuchiya et al. [18], the investigated technology is divided into its underlying components, allowing to evaluate cost reductions by material savings or improved processes directly. As a result of the determination of learning rates at a component level, components with known learning effects can be defined faster and independent from the rest. Thus, the necessity of estimating single learning rates, and the potential for errors, is reduced to a minimum number of individual components. This also allows for specifying relevant scenarios more precisely.

Additionally, the approach presented in this study enables opportunities to incorporate and evaluate spill-over learning effects from concurrent usage of individual parts of the investigated systems. Due to decoupling of peripheral components, learning effects not directly related to the entrant technology are separated, allowing to take past learning investments on these components into account.

The following sections provide an overview of the concept of technological learning in general and existing evaluations of learning rates for electrolysis in detail. Subsequently, in section Methods the methodology behind the proposed approach is explained and applied to most common

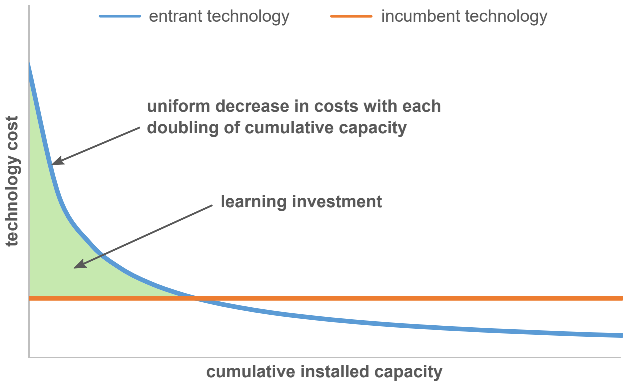

Fig. 1 – Development of entrant and incumbent technologies' costs; based on [21].

electrolysis technologies. In section Results and discussion the results are discussed, followed by a summary of the conclusions and remaining issues.

Technological learning

There are several causes behind the phenomenon of learning and learning curves, whose effects have not been fully investigated. The essential feature, however, is the acquisition of experience in the entire manufacturing process: the greater the cumulative production of a good, the greater this experience and the lower the product costs will be over time, since the manufacturing process can be optimized, resources can be saved, and economies of scale can be used. Similar observations have already been made by Wright in the 1920s [19], revealing a constant decrease of required airplane production hours through repetition of production. The essence of technological learning can be formulated as follows – a competitive environment enables individuals, companies, and industries to enhance their performance. The cost reduction is connected to the activity in the market – the actual production of the good as opposed to pure research and development.

In the 1960s, the phenomenon of learning curves was scientifically examined by the Boston Consulting Group [20], and the term experience curve was characterized. Contrary to the concept of the learning curve, the experience curve approach does not relate to individual input costs, such as labour costs, but to the total cost of a production process. This means that all costs incurred until the product reaches the end user are included (e.g. R&D, distribution and marketing costs). Though, in practice, the concepts of learning and experience curves cannot be clearly separated because the experience curve concept has its origin in the learning curve. The main difference between learning effects and experience curves are the following: While a learning curve refers exclusively to the ratio of the cumulative application rate to the production time, an experience curve refers to the ratio of the cumulative application rate to the cost of production. Since, these terms are often used interchangeably in the literature without annotation, they are not further separated herein as well, if not stated differently.

In applying the simple form of the learning curve (cf. Eq. (1)), the rate of progress remains constant over the entire learning curve. This means that entrant technologies learn faster from market experience than incumbent technologies at the same learning rate owing to the significant effect of the same absolute increase in cumulative production at the beginning of a product's lifecycle. Therefore, the concept of technological learning demonstrates the benefit of early investment in, and policy interventions, concerning emerging technologies. Learning curves are applied to deduce past cost reductions to ascertain future cumulative production levels and offer an indication of the “learning investments”, which are necessary for the deployment of an entrant technology, while learning effects cover the gap between the costs of the entrant and the incumbent technologies (cf. Fig. 1).

In addition to technological learning in the narrower sense (improvements in technology), learning curves also include the learning of employees (faster execution of recurring as well as non-recurring work), economies of scale, and other effects. Economies of scale – in terms of manufacturing scale – denote the cost advantages of mass production and provide a basis for the competitive strategy to attain cost leadership, defined as striving to reduce the cost to the lowest level among all competitors. The economies of scale explain why many companies and corporations are striving to increase their size, conquer new markets, or purchase other companies. To distinguish pure scale effects from learning curve effects, it is important to clarify the different explanatory variables.

Economies of scale refer to cost reductions per input with an increase in output. The costs serve as a function of the output produced at a given time. Conversely, learning curve effects are based on the cumulative output. Therefore, learning curve effects can also occur without an increase in the production capacity [22]. While the scope of this paper implies economies of manufacturing scale, scaling effects from changed product capacities (economies of unit scale) are not considered as technological learning in this context. However, a general increase in typical nominal capacity of a technology can also influence its learning rate [23], which is not distinguished from direct learning effects.

Formal description

The concept of learning curves describes this empirical finding of decreasing costs at each doubling of cumulative productions by a constant percentage, which is commonly denoted by the learning rate [19,24,25]. Although often referred to as the “learning-by-doing” (LBD) rate, this learning rate parameter serves as a proxy for all aspects that contribute to observed changes in the cost [15]. Thus, the learning curves represent the relationship between the following two quantities: the cost of a product and the experience expressed in cumulative production of that product.

Product costs can be represented as a function of the cumulative production:

$$C(X) = C_0 \cdot \left(\frac{X}{X_0}\right)^{-r} \tag{1}$$

where C denotes the costs at a given time, X is the cumulative production at that time, C_0 are the initial costs at a cumulative production of X_0 , and r is the (positive) learning parameter.

A twice-logarithmic plot of the costs related to the cumulative production results in a linear function with the gradient $-r$. If the cumulative production is doubled, then the costs would decrease to 2^{-r} of the original costs. This number, the so-called progress ratio (PR), is often used when comparing different learning curves and calculated according to

$$PR = 2^{-r} \tag{2}$$

In addition, the aforementioned learning rate LR is used, which describes the cost reduction when the cumulative production or capacity is doubled:

$$LR = 1 - PR = 1 - 2^{-r} \tag{3}$$

The one factor learning curve (OFLC)

The concept of the one factor learning curve (OFLC), following the basic function in Eq. (1), benefits from relatively easy accessibility of data. Investment costs and production (or installation) volumes are often well-documented when compared to other underlying cost drivers [21]. Hence, learning curves that are more consistent can be derived for economic modelling.

As discussed in Wiesenthal et al. [21], for a number of technologies, the learning effect is less apparent or even non-existing, such as gas pipelines [12,26]. In other cases, the OFLC can be derived, but a low statistical significance may imply high annual fluctuations in costs. Moreover, a rise in the net cost can occur, e.g. if the market tightness and commodity price increases counterweigh the cost-reducing technology learning effects [21]. Hence, a proposed development to the OFLC is to divide the accumulated cost into more of its core factors and analyse the parts independently. This would not only put the focus on investment costs but also on the conversion efficiency, maintenance costs, safety features, and reliability of factors of the demand side [21]. This approach can, according to Eq. (1), be formally explained by

$$C(X) = \alpha \cdot C(X_0)\left(\frac{X}{X_0}\right)^{-r} + (1-\alpha)\cdot C(X_0) \tag{4}$$

where α represents the initial cost share of the learning component [17,21].

By this multi-component learning analysis, Ferioli et al. [17] and van der Zwaan et al. [26] reveal that some cost components involve learning (e.g., the production process), while others do not (e.g., labour costs and material costs) involve learning. Additionally, the concept of multi-component learning analysis may produce diverse results regarding historical data and technology forecasts or energy scenarios compared to the OFLC. Ferioli et al. [17] show, for the case of gas turbines, that the concept of multicomponent learning could lead to different learning rates compared to the one-factor approach and realizes a better match with statistical data. Further, significant differences can arise when different learning curve concepts are applied for energy forecasts and future scenarios. As outlined in Ferioli et al. [17], the simple extrapolation of cost data over several orders of magnitude of cumulative production can imply substantial errors with regard to the breakeven capacity and the learning investment when a not appropriate learning model is applied. It must be noted that the overall costs that involve learning represent an accumulation of the costs of the specific components of the technology. Every individual fraction can have a diverse learning index. Hence, it is possible to study the impact of learning on the components independently. Nevertheless, data on particular production processes and costs may be non-existing or challenging to derive [21].

The Two Factor Learning Curve (TFLC)

While the concept of the OFLC has its strengths in the aggregation of numerous essential factors of cost reduction in one factor, this finding corresponds to observations. Therefore, individual drivers of cost reductions like research and learning-by-doing cannot be differentiated, and hence the identification of the impacts of policies addressing R&D investments is particularly limited [21]. In this regard, the division of the OFLC into a *Two Factor Learning Curve* (TFLC) was realized by Kouvaritakis et al. [27], which can, analogous to Eq. (1), be depicted formally as

$$C = C_0 \cdot \left(\frac{X}{X_0}\right)^{-r} \cdot K^{-s} \tag{5}$$

where the former part again represents the learning by doing. K denotes the knowledge stock which is approximated through the sum of R&D investments, while s is the elasticity of learning by researching.

Empirical evaluations of the concept of TFLC by Rubin et al. [15] suggest that R&D investments support cost reductions at all the stages of technological progress, and, in several cases, R&D's contribution is more significant than learning by doing (see also [27–33]). Further analyses indicate the existence of correlations between R&D expenditures and subsequent cost reductions (see [29–32]). In contrast, although the investigations of Miketa and Schrattengolzer [34] support the general feasibility of this concept, they also show that the accuracy of the representation of the cost reductions is not improved. Finally, it must be noted that research on the TFLC often limits consideration to public R&D expenditures because data on private R&D spending are usually not accessible or adequately disaggregated [21] and the role of R&D on technological learning is judged diversely [32,35–37].

Besides the described *One-* and *Two-Factor Learning Curves* different decomposition techniques are used in related studies to separate and describe additional learning effects, like economies of scale, material input prices, labor costs, efficiency improvements, etc. Hence, according *Three-* or *Multi-Factor Learning Curve* models can be found in literature as well [38].

Learning curves for power-to-gas

To assess future implementation costs for Power-to-Gas (PtG) systems using the theory of technological learning, appropriate knowledge of expectable learning effects is mandatory. Due to the novelty of the technology and therefore low numbers of real implementations, availability of data on cumulative productions and production costs is limited. Schoots et al. [12] provided an assessment of cost reductions observed for different hydrogen production technologies including water electrolysis, which has shown a moderate progress ratio of 82% ($lr = 18\%$), but covered with high uncertainties (coefficient of correlation $R^2 = 0.28$) due to low number of available data points. A more recent study by Schmidt et al. [39], analysing experience rates for different storage technologies for electrical energy, extends these investigations up to the year 2017, providing a comparable learning rate of $18 \pm 6\%$ for electrolysis cell stacks. While both studies do not distinguish between different stack technologies, according to the investigated timeframe (1920–2017), a dominance of alkaline cell technology is assumed. A differentiation between alkaline, PEM and solid oxide cell technology based on past productions is not available in relevant literature, since PEMEC and SOEC technologies still have low market adoption. Estimations based on expected future costs from expert elicitation studies [14] revealed similar experience rates of 18% for alkaline and PEM electrolysis cells. The experience rate for SOEC is expected to be substantially higher with a mean value of 28%, though this comes with a high uncertainty of ±16% and is therefore terminally significant.

Since fuel cells and their characteristics show technological similarities to electrolysis cells, relevant literature on technological learning for fuel cell technologies has been reviewed as well. Investigations on cost reductions by learning curve effects for established configurations, like alkaline (AFC), phosphoric acid (PAFC) and PEM fuel cells (PEMFC), established by Schoots et al. [40] have shown similar rates for AFC compared to its electrolysis counterpart (18 ± 9%) for an investigation period of 7 years. For PEMFC technology, differences to electrolysis cells are higher, showing a learning rate of 21 ± 4% for the fuel cell in a global observation. An observation of micro-CHP applications in Japan and Korea confirms an average learning rate of 18% [41,42] without differentiating between fuel cell technologies used, and rather 16–21.4% for PEM technology in detail [43–46]. Mayer et al. [33] used a two-factor approach (TFLC) to estimate 2020 target costs for PEMFC, resulting in a learning rate of 20% for R&D-based effects and 13% for general learning. Solid oxide fuel cells (SOFC) have been investigated in an individual study [47], which is also distinguishing between R&D, pilot and early commercial state and explicitly taking different phenomena of cost reduction (learning, economies of scale and automation) into account. Considering all stages and learning effects, a learning rate of 35% was found for SOFC, which is relatively high compared to the other designs. Though, highest rates are found for the pilot stage, whereas for the commercial state a significantly lower potential of technological learning at a rate of 12% is expected. On the other hand, investigations on SOFC for commercial stationary CHP applications on the US market did not show any observable cost reductions for the technology [41,44].

According to the gathered data, the technological similarities between electrolysis and fuel cells, in the case of alkaline and PEM cells, seem to be also representative when it comes to the effect of technological learning. However, this dependency is not observable in a meaningful extent for solid oxide cell. In reference to the even higher expected comparability for this technology, since solid oxide cells also promote reversibility of the conversion process [48], and the high uncertainties given for the observed learning rates, this clearly shows that the applicability of the conventional learning curve theory requires a minimum amount of cumulative productions to have taken and therefore presumes an adequate level of maturity of the technology. Additionally, an unclear definition of system boundaries in the investigations does not allow to distinguish between learning effects of main parts (cell stacks) and peripheral components that differ significantly between electrolysis and fuel cell applications. Therefore, a more detailed analysis would potentially show a higher compliance of learning effects on technologically similar components.

Methodological issues

Though the aim of using the learning curve theory is to allow prospects of future technology costs, this is hardly possible for novel applications at a low technology readiness level (TRL). Since significant effects, which are described through technological learning, can only be evaluated after a few magnitudes of produced units, the technology under investigation must reach a certain degree of maturity to allow an assessment on the further development of production costs. Nevertheless, it is often mandatory to consider technological learning, along with an analysis of future market potentials of the product, to perform techno-economic assessments before these technologies enter the market, supporting initial decisions on investments. Therefore, alternative approaches for an early estimate of the used experience rate must be used.

An obvious approach would be the reuse of experience rates already found for applications with comparable functionality or usage (e.g. assume similar effects for offshore and on-shore wind power plants). Though this seems theoretically feasible, it is usually not, since minor changes in technology can have significant influence on its observable technological learning process. Beyond that, differences in observed learning rates can be caused by many factors. On the one hand, they can be predicated on the available price basis, which often only considers installation or investment costs instead of pure production costs. Since the knowledge of the margin between those cost levels (production vs. purchase) is often confined to the manufacturers, it is hardly possible to consider influences like price umbrellas or shakeout effects [49,50] (cf. Fig. 2). On the other hand, those differences can result from learning spillover effects (cf [51]). In this context, it can be expected that technological experiences made on one technology are also reflected in the experience curve of a related technology, and can therefore not be directly assigned to the increased cumulative productions of one of them. This can also reflect technology comprehensive structural technological change, as stated by Wene [52], leading to a discontinuity in the learning curve, as shown in Fig. 3. Such radical changes allow to change the entry point of the learning curve and possibly the learning rate, thus similar technologies can benefit from the experience gained by each other.

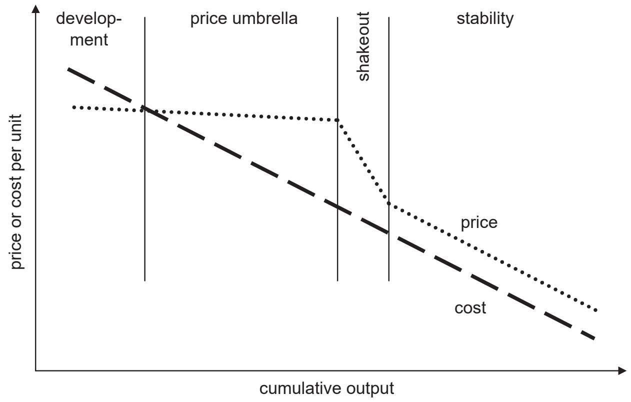

Fig. 2 – Price-cost relation for a new product; based on [50].

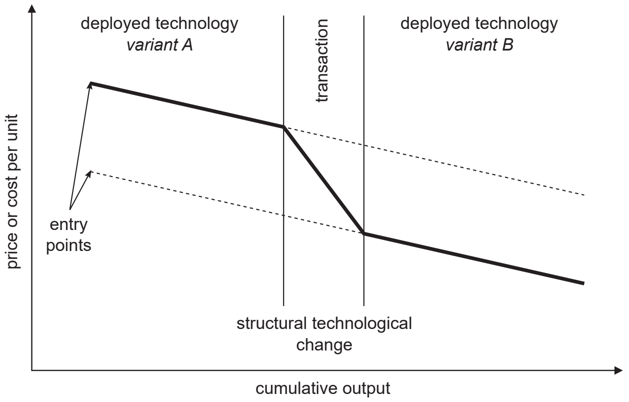

Fig. 3 – Structural technological change; based on [52].

Methods

Apart from reusing learning rates between different technologies, this paper proposes an approach to get a rough and reasonable estimate on experience rates for low maturity technologies by dividing an appliance into its subcomponents. Hence, the theory of technological learning can be applied to every single component and subsequently summed up to an overall experience curve for the appliance. Therefore, it is mandatory to know the initial underlying component and corresponding cost structure in detail. While the acquisition of this data should be feasible in many cases, additional learning rates for every used component should be known or estimated. Though the complexity and the number of learning technologies (components) to investigate increases respectively, it provides additional and, in some cases, easier methods to evaluate certain cost reduction effects. This means that, on a component basis, factors that influence the production costs can be partly determined and described by simple scaling and innovation processes like the following:

1. Cost reductions from mass productions: By investigating learning rates on a component level, a decline in production costs that occurs by upscaling of the manufacturing processes can be distinguished easier.
2. Changing material costs: By a breakdown to several contained components, the variety of materials used per component becomes more manageable, allowing a more accurate estimation of future production costs development based on raw material costs.
3. Reductions in material usage: An analysis at component level also allows for separating and substantiating expected savings in the usage of expensive raw material for cost-intensive parts.
4. Improvements in manufacturing time: Improvements in processing time can be determined and evaluated more precisely at component level than for the whole appliance. This does not only include machine processing costs but also manual working time costs.

While this approach still requires a certain amount of experience made through a few orders of productions, many individual components are not reinvented for every single purpose, but they are often reused within different applications. As a conclusion, it is reasonable to combine both approaches to assess learning effects for novel technologies — evaluate cost reduction potentials at a component level and use existing experiences from comparable component usage in well-established applications.

Module level implementation

Components and modules

As already stated by Ferioli et al. [17], a certain product, process, or technology can be considered as an aggregate of several components or costs factors, wherein the costs for each component follow the learning curve theory. The resulting cost curves can be subsequently summed up to present the total costs of the investigated application.

$$C(X_t) = \sum_{i=1}^{n} C_{0i}\left(\frac{X_{ti}}{X_{0i}}\right)^{-r_i} = C_{01}\left(\frac{X_{t1}}{X_{01}}\right)^{-r_1} + C_{02}\left(\frac{X_{t2}}{X_{02}}\right)^{-r_2} + \cdots + C_{0n}\left(\frac{X_{tn}}{X_{0n}}\right)^{-r_n} \tag{6}$$

where the variables are defined as follows:

X_{0i}…cumulative number of component *i* produced at time $t = 0$
X_{ti}…cumulative number of component *i* produced at time *t*
C_{0i}…costs of component *i* at time $t = 0$
$C(X_t)$…total costs at time *t*
r_i…learning parameter for component *i* (where $lr = 1 - 2^{-r}$)

While, in Ferioli et al. [17], the approach is simplified by only considering learning and non-learning parts (cf. Eq. (4)), this can be used to break down the xinvestigated technology to an appropriate level of detail. This approach is simplified by relating all individual learning curves to the cumulative productions of the overall system only (Eq. (7)). Though this excludes some factors, like spillover effects between components, it is still adequate and more practical for early learning rate estimations at a component level. A similar approach was used by Tsuchiya et al. [1] for the evaluation of mass production cost for PEM fuel cells.

$$C(X_t) = \sum_{i=1}^{n} C_{0i}\left(\frac{X_t}{X_0}\right)^{-r_i} = C_{01}\left(\frac{X_t}{X_0}\right)^{-r_1} + C_{02}\left(\frac{X_t}{X_0}\right)^{-r_2} + \cdots + C_{0n}\left(\frac{X_t}{X_0}\right)^{-r_n} \tag{7}$$

In this context, experience curves for single components, or rather cost elements, can be estimated by means of the aforementioned factors from individual forecasts, such as the variation in material costs. If no such data is available, comparable use in other applications have to be consulted to evaluate certain learning curve effects. This can either be done by screening relevant complementary technologies, or by considering values of experiences from manufacturers, especially for estimations regarding reductions in processing times and material usage. The main difficulty in the implementation of the given concepts arises as a result of generating an appropriate amount of experience, as well as estimating the comparability within and between different fields of application. Thus, the involvement of particular (part) manufacturers and their expertise can represent an essential advantage in the evaluation. The proposed calculation model allows to combine both approaches, consideration of expectable cost reductions (e.g. material savings or price decline) and reuse of observations of technological learning from comparable technologies, materials or manufacturing processes.

Learning Properties

Although similar or identical components or cost factors, respectively, with comparable learning rates, are consulted for the calculations, additional criteria that have at least an indirect influence on the learning rate need to be considered, especially, if the learning rates are applied to the particular applications in different extents. Practically, this means that if a component is used in a similar or even identical manner within two considered applications, individual properties of the single applications can nevertheless result in different experience rates for that component.

As an example, for the membranes used in PEM fuel and electrolysis cells, comparable experience rates can be expected according to the material and component usage. However, concurrently, it can be assumed that, relating to the cell power, there will be a variance in the development of the current densities of these two technologies along with the material usage of the membrane itself. This could be considered directly within the appropriate learning rate per application, though this would impede comparability and interchangeability of learning rates between the two technologies.

In the presented model individual properties, which indirectly influence the learning rate of one or more components are applied to every affected component as a so-called "Learning Property." In this context, each of these "Learning Properties" is defined by its own learning rate and, likewise, follows the basic equations of the OLFC theory as a function of the cumulative production of the overall module. A similar application of this concept for the power density of PEMFC can also be found in Tsuchiya et al. [1].

$$P_t = P_0 \left(\frac{X_t}{X_0}\right)^{-r_p} \tag{8}$$

with:

X_0…cumulative number of productions at time $t = 0$
X_t…cumulative number of productions at time t
P_0…initial value of property P at time $t = 0$
P_t…value of property P at time t
r_p…learning parameter for property P (where $lr = 1 - 2^{-r}$)

When applying such a "Learning Property" to an appropriate component i , its value relates to its initial value:

$$C_i(X_t) = C_{0i}\left(\frac{P_0}{P_t}\right)^{ex}\left(\frac{X_t}{X_0}\right)^{-r_i} = C_{0i}\left(\frac{X_t}{X_0}\right)^{(-r_i + ex*r_p)} \tag{9}$$

where the exponent *ex* represents an "influence exponent" that defines the mathematical dependency between the property and the component. Hence, an "influence exponent" of $ex = 1$ connotes linear dependency. A quadratic dependency ($ex = 2$) is required, if the variation of the property is related to a single dimension (e. g., length-based), whereas a two-dimensional relationship (e.g., area-based) is used for the considered component it is applied to.

Since the model supports the application of several "Learning Properties" to a single component and vice versa, the mathematical determination of the total learning curve for the overall module is defined as follows:

$$C(X_t) = \sum_{i=1}^{m}\left\{C_{0i}\cdot\prod_{j_i=1}^{n_i}\left[\left(\frac{P_0}{P_t}\right)^{ex}\right]\left(\frac{X_t}{X_0}\right)^{-r_i}\right\} \tag{10}$$

Additionally, "Learning Properties" allow to prepare individual, clearly distinguishable learning effects like cost reduction by material savings and those by savings in working or processing time, without the need of additional cost factors within the component structure.

System level implementation

Need for a full system view

The examination of learning curve effects for overall systems is accompanied by several additional challenges. Basically, when studying relevant literature, it can be observed that the learning curve theory is mainly applied for the most novel technology, by means of plant components (=modules in the context of this paper) with the lowest technology readiness level (TRL) and therefore highest expected potential for cost reduction are analysed primarily (e. g. cell stacks within an electrolysis plant). In this respect, the simplest method would be splitting the system into a "learning" part, including the novel technologies with certain learning curve effects, and a "non-learning" part, describing the miscellaneous plant components, matching the two component approach described by Ferioli et al. [17] in Eq. (4).

To consider the learning curve effects from peripheral plant modules, a similar approach as used in the module level is reasonable. The experience curves for the single modules are analysed individually, and then summed up to the overall system learning curve, consisting of a compound of different entities of the introduced "Learning Modules". With the introduction of an additional "System Level" the analysis of learning curves and cost structures for modules (and components) can be executed independently from the overall system, and hence a differentiation and grouping of the individual parts is possible.

Consideration of spillover and indirect learning effects

When considering peripheral plant components (=modules) that only partially indicate the novelty and therefore the potential for technological learning, some supplemental aspects must be respected in the evaluation of overall learning effects. Cost reductions that are observable for certain plant modules, which are also used inside other technology in an identical or comparable manner (e. g., gas conditioning/compression), cannot thoroughly be assigned to the cumulative production of the observed system [21]. While these effects were not considered on the module level of the model, at the system level, the individual plant parts mostly comprise components that are independent from the main technology itself but widely used elsewhere. In addition, independent of the chosen system boundaries, the major part of technological learning can often be confined to a limited amount of individual modules.

To qualify learning curve effects, which are not directly assignable to the increasing production of the complete system, but rather justified by secondary usages within other systems, certain dependencies between the time series data of production amounts of the overall system and those of the single modules were defined inside the calculation module. This means that while individual time series data for the production volumes of every single module and complete system is defined, in an additional step, the relationship between the time series of those two observation levels is determined for every single module inside the calculation model. The model includes four different dependencies:

X_{module}…cum. production of the module,
X_{system}…cum. production of the investigated system,
$X_{module,ind}$…cum. production of the module independent from system (concurrent usages)

- *Direct:* The time series data, which was defined for the overall system, is also used for the modules' learning curve. This means that the learning effects for certain modules are also directly coupled to the production amounts of the system ($X_{module} = X_{system}$).
- *Independent:* Only the time series data of the module itself is considered for the calculation of its learning curve. As a result, an increase in the production of units of the system does not have a direct influence on the cost reductions of the module ($X_{module} = X_{module,ind}$).
- *Indirect:* The time series data of the module and the system are added. Hence, direct learning effects from the production of new units of the investigated system as well as indirect ones from other usages (in concurrent applications) of the particular module are considered ($X_{module} = X_{module,ind} + X_{system}$).
- *Constant:* The cumulative production of the particular module is supposed to be constant. This means that theoretically no additional units are produced and therefore no learning effects occur – being equal to defining a learning rate of $lr = 0$ for the particular module ($X_{module} = const$).

Methodological advantages

Through the composition from individual components, each with their own learning rates, the resulting overall learning rate does not remain constant, but depends on the underlying time-related cost structure. Since costs for components decrease faster with a higher learning rate, the division of the module costs over all used components will vary depending on the cumulative productions. Therefore, cost shares for components with higher experience rates will decrease, whereas those components with low rates will become higher rated. This allows a detailed analysis of the development of the cost structures on each level providing an insight which components or modules are cost determining at which level of technological learning – as long as the initial costs and learning rates are chosen reliably.

On a full system view, the model allows a consideration of spillover and indirect learning effects, providing a significant advantage over common models and therefore allow estimations on the influence of learning curve effects of well-established plant components, which are not the main drivers in cost reduction by technological learning on a novel technology.

Application on electrolysis technologies

Within this study the proposed model is applied to the three main technologies of water electrolysis (PEM, alkaline, solid oxide) used in the Power-to-Gas (PtG) process. Though they are at different stages of maturity each, from mature (alkaline) over commercial (PEM) down to demonstration (solid oxide) stage [14], none of them has reached mass production yet. Hence, water electrolysis still provides significant potentials for technological learning, while still missing production data in relevant amounts, at least for PEM and solid oxide technology, to evaluate this cost reduction potentials using conventional methods of the learning curve theory. Additionally, similarities in certain components between the three subtypes match the advantages of the discussed calculation model.

Electrolysis cell stack module

To evaluate experience curves for electrolysis cells by using the presented component-based approach, a definition of the cell composition is mandatory. While all investigated water electrolysis technologies basically use the same input (water and electric current) and produce equal output (hydrogen and oxygen) flows, their technological composition is rather different. Despite that, some individual components share similar functions and are built analogically at a component level when compared between technologies, allowing a feasible comparison of learning rates according to the described methods.

In this regard, the classifications for PEMEC and AEC are primarily based on data by E4tech [53] and Smolinka et al. [11,54], since they also provide cost structure data. This component structure is, on the whole, comparable to data and descriptions available in other literature about PEM cell stack technology, be it electrolysis [10,55–57] or fuel cell [18,58,59]. Compared to AEC and PEMEC, solid oxide electrolysis cells are an emergent technology that is expected to reach high current densities at elevated operating temperatures and appropriate integrated heat management [48,60,61], making an investigation within this study reasonable. Since the SOC technology is expected to allow operation in both electrolysis and fuel cell modes, with the same cell configuration [48,60], also technology and cost data available for solid oxide fuel cells (SOFC) have been used for the investigations. Therefore, relying on component structure according to Smolinka et al. [11], while appropriate cost data was derived from Scataglini et al. [62], providing a direct manufacturing cost model for SOFC stacks. Component structures are summarised in Table 1 together with particular shares on total cell stack costs.

Table 1 – Cost shares and learning rates chosen for analysed electrolysis cell modules (grey: technological main components).

Technology	Component	Initial cost share	lr	Notes
PEMEC	Stack assembling	2%	8%	initial cost shares based on [11,53]; learning rates based on [18] and iterative calculation
	Small parts	3%	5%	
	MEA manufacturing	10%	8%	
	Catalyst cathode	2%	8%	
	Catalyst anode	6%	8%	
	Membranes	5%	18%	
	Current collectors cathode	9%	18%	
	Current collectors anode	8%	18%	
	Bipolar plates	51%	18%	
	End plates	1%	8%	
	Pressure plates	3%	8%	
AEC	Structural Rings	15%	5%	initial cost shares based on [11,53]; learning rates based on [18] and PEMEC findings
	PTFE sealing	4%	8%	
	Bipolar plates	7%	18%	
	Pre electrode	8%	18%	
	Anode	26%	18%	
	Cathode	25%	18%	
	Membrane	7%	18%	
	Flanges	4%	5%	
	Tie Rods	3%	5%	
SOEC	Stack Assembling	9%	8%	initial cost shares based on [11,62]; learning rates based on [18] and PEMEC findings
	Electrolyte	12%	18%	
	Catalyst Anode	15%	18%	
	Catalyst Cathode	23%	18%	
	Current Collector (PTL)	8%	18%	
	Interconnector (Flowfield)	12%	18%	
	Sealings	15%	5%	
	End Plates	2%	8%	
	Pressure Plates	4%	8%	

For the definition of component-wise experience rates, following the approach of interchangeability of component characteristics between different use cases, as one of the fundamental ideas behind the introduced module, values based on the learning effects identified and described by Tsuchiya et al. [18] for PEM fuel cell applications were used (cf. Table 1). Generalized, for technology-independent standard parts, a rather low learning rate of $lr = 0.05$ is chosen, while peripheral parts that are specific to the technology are defined with $lr = 0.08$ throughout the investigated technologies. Learning rates for technology-decisive parts, like membranes, electrodes or bipolar plates, were determined iteratively to match experience curves observed from past cost development for electrolysis cells (cf. section Comparison with the conventional OFLC).

Despite all comparability between the PEMFC components used by Tsuchiya et al. [18] and those used for PEM and other electrolysis cells, technological differences are expected, which are not covered by the learning effects given in Table 1. Particularly, the power density of the cells significantly differs between the technologies both in value and evolution. At the same time, when comparing experience rates between cell designs, for single components this is only feasible in an area-related manner. Therefore, the power density acts as the transformation factor between area and power related views. As the specific production or installation costs are commonly analysed in relation to their rated power (e.g. €/kW$_{el}$), the power density will have a relevant impact on the evaluated learning curves. Thus, the power density was implemented as a “Learning Property”, influencing relevant (area-related) components to overcome those circumstances. The property characteristics and the influenced components for each cell design are shown in Table 2.

The learning rates chosen for the power density property possess negative values. Hence, the property's value will increase with an increase in the amounts of produced units representing the trends expected for future implementations of the cells. Furthermore, components like “Stack assembling” are not influenced by the “Power density” property as they are expected to be rather independent of the power obtainable per cell area.

The supposed value for the property's learning rate is based on a literature review combined with iterative calculations. The NOW study by Smolinka et al. [54] provides mid-term and long-term forecasts of PEMEC and AEC characteristics, as listed in Table 3. The initial values were derived based on literature on current densities and cell voltages [11,14,53–55]. The development of the power densities of SOEC is more difficult to define since this parameter is highly dependent on the electrode materials, operation mode (fuel cell or electrolysis in reversible cells) and temperature, and not least operating point (e.g. thermo-neutral operation) [61,63]. In reference to recent developments [63] power densities are expected to significantly increase with elevated current densities. As increasing current densities are an expected trend in the course of technology upscaling and small-scale performance,

Table 2 – Characteristics of the "Learning Properties" implemented for the electrolysis cell stacks (influence exponent of ex = 1 used in each case).

Property	Technology	lr	Influenced Components
Power density	PEMEC	−2,5%	Small parts, MEA manufacturing, Catalyst cathode, Catalyst anode, Membranes, Current collectors cathode, Current collectors anode, Bipolar plates, End plates, Pressure plates
	AEC	−5,5%	Structural Rings, PTFE sealing, Bipolar plates, Pre electrode, Anode, Cathode, Membrane, Flanges, Tie Rods
	SOEC	−8,0%	Structural Rings, PTFE sealing, Bipolar plates, Pre electrode, Anode, Cathode, Membrane, Flanges, Tie Rods

Table 3 – Present and future characteristics of alkaline and PEM electrolysis technology; based on [54].

Technology		Present (2011)	Mid-term (~2015–2020)	Long-term (~2020–2030)
AEC	Power density	<1,0 W/cm^2	<1,3 W/cm^2	<1,8 W/cm^2
	Efficiency	62–82%	67–82%	67–87%
PEMEC	Power density	<4,4 W/cm^2	<5,0 W/cm^2	<5,4 W/cm^2
	Efficiency	67–82%	74–87%	82–93%

together with the low maturity of the technology, rather high potentials for increase are also expected for power densities. Based on the results gained from learning curve analysis of PEM and alkaline stacks, the learning rate for the power density was therefore set to 8%, whereby resulting overall learning rate for the SOEC stack complies with the values found in literature [47].

The starting point of the experience curve is defined by the initial overall costs for the cell stack. To comply with the chosen component and cost structure based on [11,53], these costs are presumed with average system costs of 1,450 €/kW$_{el}$ for PEM and 900 € kW$_{el}$ for alkaline electrolysis, accordingly. This cost estimation is also confirmed by data found in a review study [56] for the observed timeframe. In relevance to the appropriate breakdowns, these costs are split up in four main modules as shown in Fig. 4: (i) stack, (ii) power electronics, (iii) gas conditioning, (iv) balance of plant. Comparable classifications found in other literature differ slightly in the subdivisions used, but show similar shares for the stack part [11,55]. For consistency, identical system modules were used for SOEC systems. Cost shares have been evaluated according to [11], stating values of 30% each for stack and power electronics module. Due to comparable gas conditioning efforts between all three technologies, absolute costs for this module are presumed at the same level. This cost structure is consistent with estimations found in other literature [62,64].

With the assumptions given above, the investigations for SOEC only aim to give a very rough estimation of the technology since the TRL of solid-oxide cells is rather low when compared to PEMEC and AEC, and therefore component and cost structure are difficult to validate. Due to significant differences in used materials and operating conditions (cf

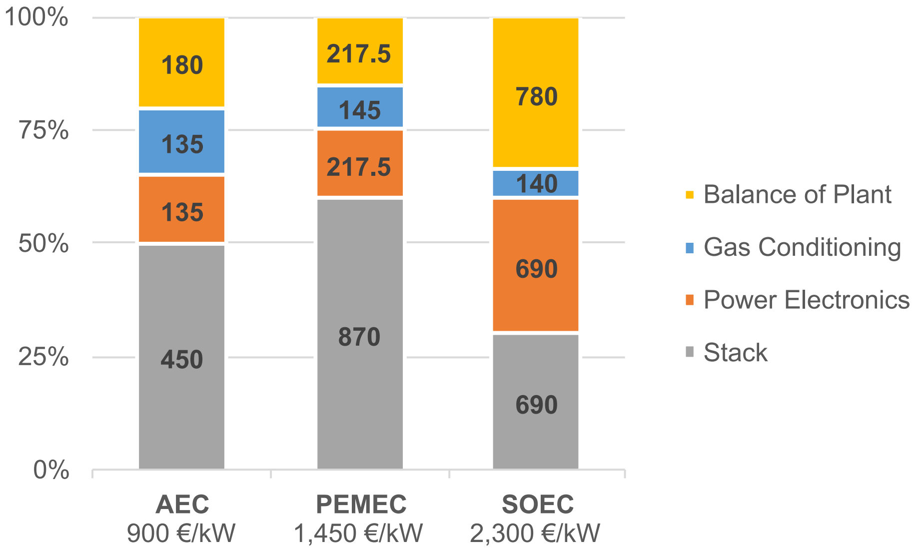

Fig. 4 – Cost breakdown for investigated electrolysis systems; based on [11,53,64].

[11,65–67]), presumed learning rates have to be observed and eventually adapted with upcoming development and market uptake of the technology.

Electrolysis system

To evaluate cost reductions for PtG technologies, an observation of the overall system costs is often desired and reasonable. This implies that learning effects for peripheral plant parts, apart from the main technology, the electrolysis cell stack, should also be considered. Since those peripheral parts are usually not cost-determining or their costs do not vary significantly, respectively, in the early stages of technology development, they are often out of the scope of long-term investigation in techno-economic studies. Hence, appropriate data about learning rates is hardly available. Additionally, the number of cumulative produced units for each module of the system, is of special interest. Since the model includes functions to preclude spillover effects from other technologies by considering the reuses of each module within other systems, a thorough analysis of the appropriate production amounts is necessary to make full use of those methodological benefits.

In the present case of electrolysis systems, some simplifications were made, compensating unknown third-party usages for the peripheral modules by either estimating lower learning rates (despite “direct” dependency, the learning effects for the module are low) or completely decoupling the module from system productions (“independent” or “constant” dependency). Additionally, peripheral modules have each been implemented with a single sub-component, resulting in a single constant learning rate for each module.

Cell stack. The electrolysis cell stack as the core module is the main driver for technological learning. For each of the three electrolysis technologies (AEC, PEMEC, and SOEC) it is implemented as defined in the previous section. As their learning effects are decoupled from each other per definitions, they are directly dependent on the cumulative production of each individual technology and therefore “direct” dependency is used on “System Level.”

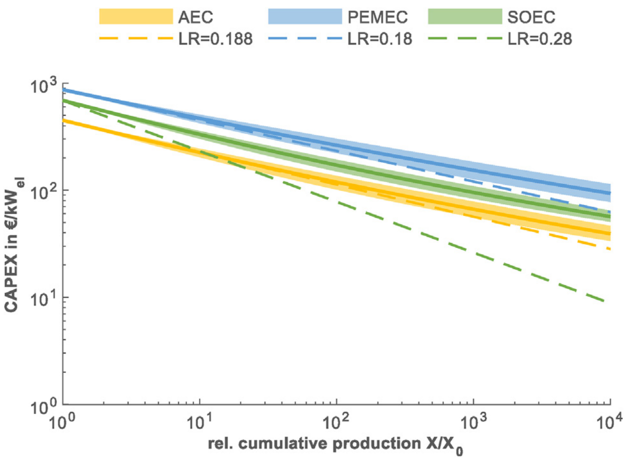

Fig. 6 – Calculated learning curves for electrolysis stack modules with learning rates for main components ranging from 15 to 22% (solid lines: learning curve with fitted learning rate of 18.04%; dashed lines: comparative one factor learning curves with mean learning rate values as found by Schmidt et al. [14]).

Power electronics. Since learning curves are usually investigated for complete systems, technologies, or products, learning rates for individual system components lack available data. Therefore, observed learning effects for different manufacturing processes have to be used as a reference. Referring to different studies [68,69], which show typical learning rates for repetitive electronics manufacturing at 5%–10% and 15%–25% for electrical wiring, an average learning rate of 12% was chosen for power electronics in the investigated electrolysis systems.

As power electronics are part of all investigated electrolysis systems in a presumably comparable manner, spillover learning effects across the different systems are expected. To include these in the calculations, it was presumed that learning effects are not directly dependent on the cumulative production of the individual technology but more or less dependent on the overall production of electrolysis systems.

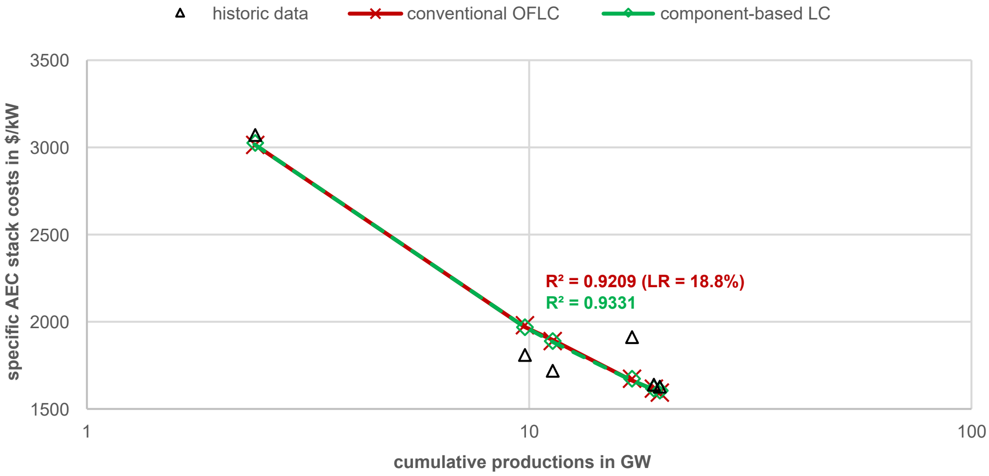

Fig. 5 – Comparison of the component-based and the conventional approach fitting to available historical cost data (data source: Schmidt et al. [39]).

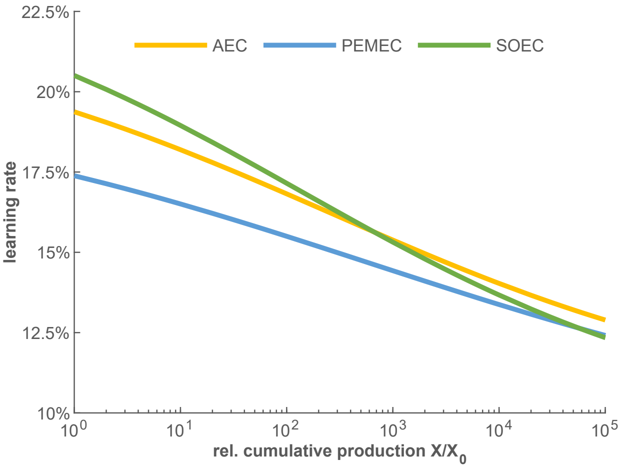

Fig. 7 – Calculated development of learning rates for electrolysis stack modules over cumulative productions.

Therefore, the dependency parameter for the power electronics module was set to “independent”, while using the overall cumulative electrolysis production.

Gas conditioning. The overall investment costs for product gas treatment in the electrolysis plant is covered in the gas conditioning module. Since this unit mainly consists of purchased parts (e.g., compressors), electronics and electrics (e.g., controls and measurement and power supply), machining and assembly and components, which are far beyond R&D, the learning rate is set relatively low at 7%, following [18,68,69].

Gas treatment for further processing is not unique to hydrogen production by electrolysis but also for conventional ways of H_2 generation like steam methane reforming (SMR). As a result, learning effects on this module will not only be influenced by the production of electrolyser units but also spillover from other forms of H_2 generation. Particularly, cumulative production and therefore learning that has happened in the past must be considered in the calculations. To take this into account, cumulative hydrogen production from 1990 to 2014 [70] is used as a base value. Hence, the gas conditioning module uses “independent” dependency parameter in the calculations by using cumulative processing of hydrogen from electrolysis on top of those conventional production values from the past.

Balance of plant. Costs of peripheral components and implementation tasks of the electrolysis system that are not covered by other modules are summed up in the Balance of Plant (BoP). Since included sub-components vary widely between different implementations of electrolysis plants (even within the same technology), a valuable allocation of a cost structure for this module would have to be done individually for specific implementations. To allow an assessment in this study, the module was reduced to a single sub-component. Referring to Strategos Inc. [69], a moderate learning rate of 13% was assumed, mostly a composition of purchased parts (12%–15%), machining (5%–10%), assembly (10%–20%), welding (10%), and comparable cost factors in a similar range [68]. Staffel et al. used a two-factor learning curve to estimate cost reductions for micro-CHP fuel cell systems, suggesting a similar learning rate of 11.7% for generic BoP components [45].

Since there is a high individuality per technology, especially in pressure levels and heat management, spillover effects were neglected for this module. Hence, a “direct” dependency between the technological learning of BoP and each individual electrolysis technology was chosen for the calculations.

Table 4 summarises the modules and presumptions used for the calculation of the technological learning potential for electrolysis systems based on AEC, PEMEC, and SOEC stacks.

Results and discussion

Stack modules

Comparison with the conventional OFLC

To prove the quality of the component-based learning curve approach when compared to the conventional theory that uses a fixed learning rate, the experience curves were calculated for available historical data on costs of electrolysis cell stacks. However, the amount of cost data available, especially in conjunction with data on the corresponding cumulative

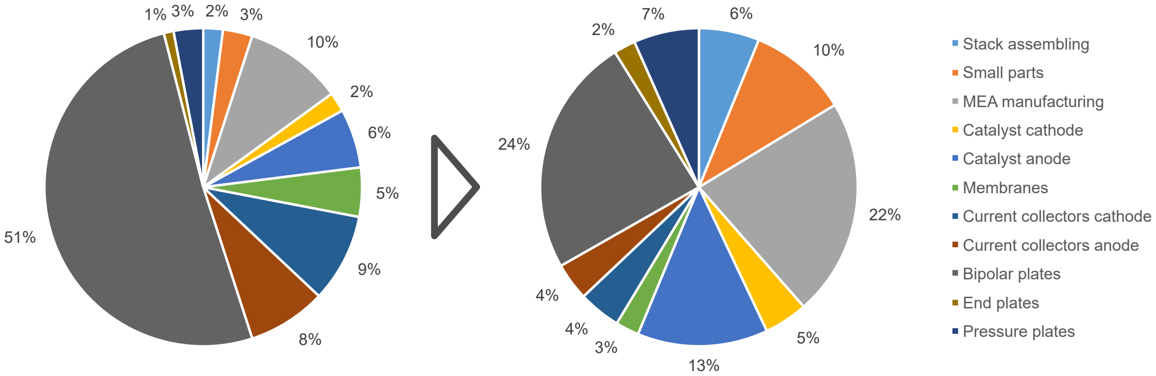

Fig. 8 – Development of the cost structure of PEM electrolysis cells for different cumulative amounts of produced units (left: initial; right: factor 1,000).

number of produced units (or nominal power), is quite limited. To provide updated information, the data disseminated by Schmidt et al. (paper: [13]; dataset [39]) was used for reference. Fig. 5 shows a comparison of the component-based and conventional approaches fitting the curves to the available historical data points.

The curve for the conventional approach was determined by fitting a common experience curve (cf. Eq. (1)) to the given set of data points, resulting in an experience rate of 18.8%. Comparing this rate to available relevant literature [12,71–73], with learning rates of 17.7–18.3%, the value seems reasonable.

Though it is not explicitly stated which kind of technology the historical cost data stands for, it is assumed, from the given time frame and the general development of the water electrolysis technologies in that time, that the major part is about alkaline cells. Hence, the parameters for the AEC presented above were used. The learning rate for the main components of the cell (cf. Table 1) where defined by fitting the model to the historic data. Thus, the resulting learning rate for these components was found at a value of 18.04%. For the subsequent results, this value was transferred to the main components of the other stack technologies, unless otherwise noted.

A comparison of both learning curves in Fig. 5 shows no significant differences. The coefficient of determination (R^2) shows that both approaches are fitting, hypothesising that the presented approach is not worse than the common theory for the given data points.

Stack results

According to definitions stated for the individual stack modules, the corresponding learning curves for these modules have been evaluated separately. As it can be seen in Fig. 6, the learning curve determined with the component-based approach differs significantly from the curve gained by the conventional approach (dashed lines), by using a single learning rate for the whole module (rates according to Schmidt et al. [14], corresponding with values from other relevant references [13,14,40]). Furthermore, the learning curve is not entirely linear in the log–log graph, as it is for a single learning rate. This development of the overall learning rate as a function of cumulative production volumes is shown in Fig. 7. It is explained by the effect that components with high individual learning rates, which often come hand in hand with high-cost shares, reduce faster in costs through technological learning when compared to components with lower rates. As a result, their shares on the overall costs decrease, together with their influence on the modules' overall learning rate. The learning curve thus gains some flexibility when compared to the common theory of technological learning. This experience can also be applied to explain some observed decreases in experience rates in different stages of technology readiness, as described by Ferioli et al. [17], which are hard to determine using common methods.

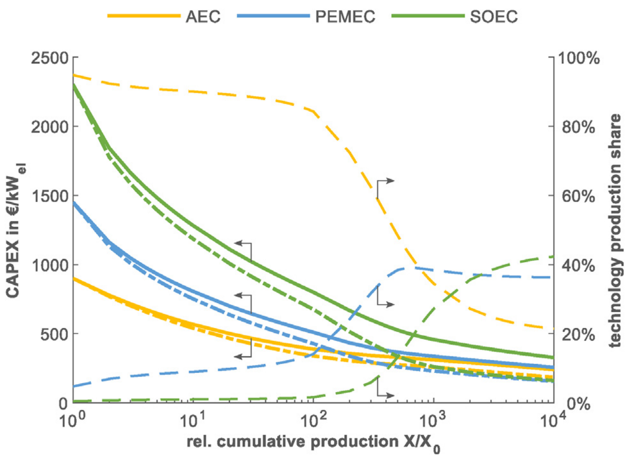

Fig. 9 – Calculated learning curves for electrolysis systems for presumed production shares (solid lines: learning curves according to proposed module structure and dependencies; dash-dot lines: learning curves without considering spill-over effects (only 'direct' module dependencies); dashed lines: presumed technology production shares).

The results reveal that the calculated learning curve for SOEC cells is significantly flatter than the static mean value learning rate. Though this literature value is covered with relatively high uncertainty (28 ± 16%), the component-based approach shows a more reasonable result when comparing resulting stack costs to AEC and PEMEC technologies with elevated maturity. Additionally, the deviation of the calculated PEMEC curve from the linear learning curve is generally a little higher compared to the values calculated for the alkaline electrolysis cell, even though the added-up cost shares of the "high-learning" main parts are on the same level for both technologies. This is explained by the fact that the development of the power density property adds another learning effect, which is significantly higher for the alkaline cell compared to PEM as per definition, since expected increase is higher for the former technology [53,54].

Defining learning effects on a component basis leads to variance in cost structures as a function of the cumulative productions for the investigated module. Fig. 8 illustrates this change for the PEMEC from the initial structure to the values for cumulative productions increased by a factor 1,000. This observation does not only influence the overall learning curve, but also shows an interesting advantage of the described method. By subdividing the modules into components and evaluating their learning curves individually, this approach allows a more detailed assessment of which components becomes price-dominant at a certain point of technology maturity. This could further allow evaluations on the time being reasonable to support investment and research activities for improving technology-critical parts instead of aiming at cost reductions on standard parts.

The calculated results on the stack modules only describe the development of the cell costs related to the electric power input. To determine the costs per generated product gas, it would be necessary to define an appropriate stack efficiency. As this conversion efficiency is expected to not be constant over time but improve by technological learning, these additional learning effects could be considered as another learning property for further investigations.

Table 4 – Summary of calculation parameters for electrolysis system.

Module	# components			initial cost share			dependency		
	AEC	PEMEC	SOEC	AEC	PEMEC	SOEC	AEC	PEMEC	SOEC
Cell Stack	9[a]	11[a]	9[a]	50%	60%	30%		direct	
Power Electronics	1 (lr = 12%)	1 (lr = 12%)	1 (lr = 12%)	15%	15%	30%		independent	
Gas Conditioning	1 (lr = 7%)	1 (lr = 7%)	1 (lr = 7%)	15%	10%	6%		independent	
Balance of Plant	1 (lr = 13%)	1 (lr = 13%)	1 (lr = 13%)	20%	15%	34%		direct	

[a] Variable learning rate calculated through the model.

Overall systems

To estimate impacts of relative increase on cumulative production of electrolysis systems in total, a presumption on development of technology share in future production was made, based on appropriate curves given in [11], but decoupled from years of implementation. Therefore, this should only provide a qualitative assessment of learning curves on system level. As a reference point for cumulative productions, following assumptions were made:

- Cumulative production of alkaline electrolysis stacks was assumed to match available data of about 20 GW_{el} (referring to [13,14,40])
- Cumulative production of PEMEC was estimated to be about 1 GW_{el} and SOEC to be about 0,1 GW_{el} (referring to [14])

The results, as shown in Fig. 9, mostly reflect the effects found on stack level, which was expected due to relatively high learning rates of included main components, while also representing major cost shares in the overall systems (cf. Table 4). Learning effects for systems using solid oxide cells are most significant, justified by the higher overall learning rate and the lower entry point for cumulative productions. The latter effect is also seen for PEM electrolysis, showing a steeper learning curve compared to the alkaline technology despite lower learning rates on module level.

Fig. 9 also illustrates the impact of spill-over effects on the learning curve. Compared to a module based setup, where cumulative productions of each module are directly related to those of the system itself (represented as dash-dot lines), there are significant differences for “Power Electronics” and “Gas Conditioning” modules observable. Concerning the former module the influence is divers. For alkaline systems module costs are increasing, due to higher starting point and missing spill-overs from emerging technologies, while PEMEC and SOEC benefit from lower individual initial productions. For the “Gas Conditioning” module, module costs are significantly lower with neglected spill-over effects for all three technologies, since external learning effects, which happened in the past and have been considered by incorporating hydrogen processing activities since the 1990s, are not taken into account. Therefore, the proposed approach for consideration of competitive usages of system modules and the resulting impact potentially provides an important factor in detailed learning curve analysis.

Conclusion

This study proposes a component-based approach for the evaluation of learning curve effects for technologies on a low TRL and therefore limited amounts of cumulative productions. Evaluating the results of the learning curve analysis, it can be concluded that the chosen approach is at least comparable to the conventional one factor learning curve for electrolysis on stack level given available data. Additionally, the assessment has shown that the consideration of potential spill-over effects from competing or parallel technology and component usage and development has a significant impact on the rating of potential cost reductions in the emergence of new technologies. This also showed that a separation between innovative system parts and peripheral components comes with different applicability, and can be an important factor in the assessment of experience curves in an overall system view. Regarding the relevant literature on technological learning on hydrogen fuel cell and electrolysis systems, this determination is often neglected, since only major technologies (e.g. electrolysis stacks) are investigated. Therefore, the observation of learning effects on commonly used peripheral components is a relevant topic for upcoming studies, even though they are already highly established and cost reductions on observed short-term are expected to be low, as emerging technologies can re-enable such mechanisms.

The evaluation of learning curves for novel and established technologies requires the analysis of an adequate amount of historical cost data. Therefore, the availability of this data is mandatory to allow reasonable predictions on the future cost development. While the component-based approach of the proposed model tries to circumvent this limitation by comparing learning effects on similar sub-components between independent technologies, the collection of base data is still unavoidable, even necessary in a more detailed view, especially in this early stage of model development. In addition, the component-wise calculation increases the number of learning curves to be evaluated according to the number of included components and thus the computational effort for aggregated system learning curves, especially for larger models. Nevertheless, the use of a component-based calculation model allows the incorporation of learning effects at a much lower level, wherein these can be determined more precisely and narrowed down to certain adaptations of the production process for single parts.

Though, the necessary level of detail must be chosen adequately and carefully. A classification that is structured perfectly comprises a corresponding high effort for the determination of the learning rates per every single component; however, it does not generate any relevant additional benefit in the calculated results. Additionally, the associated initial cost structure must be known at the forefront of the work or investigated collaterally. The calculations in this study are based on certain simplifications and assumptions due to the lack of appropriate data on learning rates for a component-wise analysis. Hence, an appropriate gathering of data and observation of manufacturing processes is mandatory to evaluate potential benefits of the presented approach to estimate future cost reductions. Although, with these requirements met, the proposed model allows the use of experience values for process improvements or the reduction of raw material costs from a unit to mass production, which is usually less obvious in a full technology view.

Though the scope of the present study is on the evaluation of learning curve effects for technologies on a low TRL, in future research the approach could also be used against more mature technologies to improve insights and explanations of past changes of learning rates, which are observed for different technologies. In addition, such ex-post analysis can help to evaluate the applicability of the proposed model on different technologies and its benefits compared to conventional approaches and support the generation of mandatory data for further analysis.

Acknowledgement

The support of this work under the Horizon 2020 research project “STOREandGO” on behalf of the European Union (contract # 691797) and by the association Energy Institute at the Johannes Kepler University is gratefully acknowledged.

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Applied Energy 264 (2020) 114780

Contents lists available at ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Projecting cost development for future large-scale power-to-gas implementations by scaling effects

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HIGHLIGHTS

- Estimated global demand potentials for PtG of up to 4530 GW.
- PtG technology costs decreasing by 12–15% for each doubling of cumulative capacity.
- Overall cost reductions of > 75% from up-scaling to multi-MW plants.
- By 2050 SNG production costs from PtG can reach values below 15 €-cent/kWh.
- H_2 production costs below 1.6 €/kg are feasible with renewable grid supply by 2050.

ARTICLE INFO

Keywords:
Power-to-gas
Electrolysis
Methanation
Scaling effects
Technological learning

ABSTRACT

Power-to-gas (PtG) is widely expected to play a valuable role in future renewable energy systems. In addition to partly allowing a further utilization of the existing gas infrastructure for energy transport and storage, hydrogen or synthetic natural gas (SNG) from electric power represents a high-density energy carrier and important feedstock material for further processing. This premise leads to a significant demand for large-scale PtG plants, which was evaluated with an amount of up to 4530 GW_{el} for electrolysis and up to 1360 GW_{SNG} for methanation capacities at a global scale. Together with the upscaling of single-MW plants available today, this will enable to achieve appropriate cost reduction effects through technological learning. Under given scenarios, reduction potentials for CAPEX of > 75% are expected for multi-MW PtG plants in the long-term, with significant advantages of PEM and solid oxide electrolysis over alkaline systems in the short- and mid-term. The resulting effects on PtG product costs were evaluated via a holistic techno-economic assessment, resulting in SNG production costs of 15 €-cent/kWh and below for large-scale appliances in 2050, depending on the renewable electricity supply.

1. Introduction

With switching from fossil fuels to renewable energy sources (RES) a transformation of existing energy systems to alternative energy carriers will be unavoidable. However, although an electrification of energy supply and its utilization to the greatest possible extent is generally reasonable in terms of energy efficiency, there will still be a significant demand for "green" gases, like hydrogen or synthetic natural gas (SNG) from renewables. Industrial processes, in particular, are expected to be reliant on these gases, be it for reduction processes (e.g. steel industry), high-density energy carriers or as feedstock material for chemicals and synthetic fuels. Besides that, power-to-gas (PtG) is often considered as a long-term storage and balancing technology for fluctuating RES or energy transport media, with the ability to use existing natural gas infrastructure, thereby potentially lowering infrastructure transition costs.

To estimate economic feasibility of different PtG concepts and evaluate impacts of regulatory measures, such as CO_2 taxes [1], the knowledge of the current state and expected development of technology and gas production costs is mandatory. While electrolysis, at least for alkaline technology, is already mature and commercially available, power-to-methane as an integrated system still represents a relatively novel technology, aside from single commercial installations, at a technology readiness level (TRL) of about 5–7 [2]. Therefore, appropriate data for commercial systems are hardly available. Thema et al. [3] analyzed cost development for electrolysis and methanation projects from 1988 onwards and provided cost projections up to 2050, assuming a continuous exponential decrease. Another approach is the

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https://doi.org/10.1016/j.apenergy.2020.114780
Received 17 December 2019; Received in revised form 26 February 2020; Accepted 28 February 2020

estimation of future cost reduction according to technological learning. Considering learning effects for alkaline electrolysis, Krishnan et al. [4] determined a potential reduction of CAPEX by 27% for a cumulative capacity addition of 80 GW by 2030. Nicodemus [5] investigated the impact of policy support on technological learning for PV powered PEM electrolysis resulting in the production costs of < 3 \$/kg H_2 by 2030. Similar results could be reached using high-temperature electrolysis [6]. Learning curves for electrolysis have already been investigated in the past using available data on alkaline electrolysis cell [4,7–9] or estimations on expert surveys [10]. Due to the low maturity of PEM and solid oxide electrolysis, appropriate learning effects often have to be estimated based on comparable technologies [11]. For the production costs of synthetic methane, Gorre et al. [12] tried to identify optimized PtG plant operation scenarios according to gas selling strategies and electricity purchase. Presuming significant reductions for CAPEX and OPEX of PtG plants, methane production costs could reach 50–90 €/MWh_{SNG} by 2030 and 25–65 €/MWh_{SNG} by 2050, under optimal conditions for plant scales of 10 MW_{el}.

In order to satisfy local demand, future demand for renewable gases will require the implementation of large-scale PtG plants for centralized production (e.g. in the steel industry [13]) and thus it will be necessary to utilize additional scaling effects. Significant cost reduction potential through up-scaling of plant production capacities for different PtG technologies were already identified by Parra et al. [14,15], Proost [16] (alkaline and PEM electrolysis and methanation) and Anghilante et al. [17] (solid-oxide electrolysis), and also considered the impacts of mass production. Gutiérrez-Martín et al. [18] evaluated power-to-SNG technology in general for large-scale storage applications, indicating levelized costs of energy (LCoE) for SNG of 30–80 €/MWh, depending on presumed electricity costs.

However, the existing studies generally only consider economies of scale either in terms of unit scale (increase in typical plant size) or technological learning (increase in number of produced units). For a holistic examination of future production costs for renewable SNG both aspects have a significant impact on overall cost reductions. Hence, our study investigates both scaling effects using component-based approach that considers the differences between common technologies and takes care of spillover effects from concurrent usage [11]. To allow this comprehensive analysis of production-related learning effects, we also include a literature review on future demand potentials for renewable gases to evaluate the according demands on cumulative productions for PtG technology components.

To provide a relation between CAPEX development and future costs for PtG products, a comprehensive techno-economic assessment for different configurations of large-scale PtG plants (≥ 50 MW_{el}) was performed. The study aims to allow the estimation of future costs of power-to-gas as a potentially valuable technology contributing to the future renewable energy systems. As such, it aims to facilitate the development of regulatory policies and measures that would be mandatory for the technology to be competitive against fossil-based technologies and processes.

2. Methodology

2.1. Consideration of dynamic learning and scaling effects

For the evaluation of power-to-gas technology costs for different combinations of installation time and capacity, an appropriate analysis of scaling effects is necessary. This study includes a detailed evaluation of two different aspects of cost reduction by economies of scale: numbering-up of produced units and sizing-up of installed system capacities.

2.1.1. Technological learning – economies of manufacturing scale

To evaluate the individual learning curves of the investigated PtG technologies, we used a component-based approach described in preceding paper dedicated to technological learning effects of electrolysis systems [11]. The model allows an investigation of learning effects on a component or manufacturing process/material level to evaluate aggregated effects for the overall system, described by Eq. (1).

$$C(X_t) = \sum_{i=1}^{n} C_{0i}\left(\frac{X_t}{X_0}\right)^{-r_i} = C_{01}\left(\frac{X_t}{X_0}\right)^{-r_1} + C_{02}\left(\frac{X_t}{X_0}\right)^{-r_2} + \cdots + C_{0n}\left(\frac{X_t}{X_0}\right)^{-r_n} \tag{1}$$

with:

X_0 … cumulative number of produced units at time $t = 0$
X_t … cumulative number of produced units at time t
C_{0i} … costs of component i at time $t = 0$
$C(X_t)$ … total costs at time t
r_i … learning parameter for component i (where $lr_i = 1 - 2^{-r_i}$)

The model also allows to take into account the learning effects for individual properties related to the specific components and hence indirectly influence their general learning curve, acc. to Eq. (2) and Eq. (3).

$$P_t = P_0\left(\frac{X_t}{X_0}\right)^{-r_p} \tag{2}$$

$$C(X_t) = \sum_{i=1}^{m}\left\{C_{0i}\cdot\prod_{j_i=1}^{n_i}\left[\left(\frac{P_{0j_i}}{P_{tj_i}}\right)^{ex_j}\right]\left(\frac{X_t}{X_0}\right)^{-r_i}\right\} \tag{3}$$

with:

P_{0j_i} … initial value of property P_j of component i at time $t = 0$
P_{tj_i} … value of property P_j of component i at time t
r_p … learning parameter for property P
ex_j … influence exponent for property P

In addition, the component-based approach supports the incorporation and evaluation of spill-over effects of technological learning from complementary technology usage on a system level. Hence, the potentials of cost reductions for peripheral components is shared between investigated technologies. This approach also considers technological learning that already occurred in the past.

For the investigated electrolysis technologies, the calculation parameters defined in [11] were used for the calculations executed in this paper. The methanation systems were also set up including 4 modules each in the learning curve model: (i) Reactor, (ii) Electric Installation & Control, (iii) Gas Conditioning and (iv) Balance of Plant. The Reactor module, which represented the core part of the methanation system, was further separated to additional Learning Components (cf. [11]) to treat different learning rates, with cost shares based on data from demonstration plants built in the STORE&Go project [19] (cf. Table 1):

- Reactor: representing the reactor vessel(s), the catalyst carrier and coupling of multi-reactor configurations
- Heat management
- Catalyst: representing the catalytic coating material and application

Table 1
Learning Components in the Reactor modules of the catalytic and biological methanation learning curve models.

Reactor module	Initial cost share	Learning rate
Catalytic		
Reactor	57%	15%
Catalyst	26%	8%
Heat management	17%	15%
Biological		
Reactor	77%	15%
Heat management	23%	15%

Table 2
Presumed maximum electrolysis stack and methanation reactor sizes related to the year of installation, based on [40,33].

Year of installation	Electrolysis			Methanation	
	Avg. max. stack size S_0 in MW_{el}			Avg. max. reactor size S_0 in MW_{SNG}	
	AEC	PEMEC	SOEC	Catalytic	Biological
2020	3	1.2	0.5	10	2
2030	4	2	1	25	5
2040	5	3.5	2	100	5
2050	5	5	3	500	5

Even though scaling limits are to be expected for almost any component in the systems, this dynamic scaling effect was neglected for the other modules except for the electrolysis stacks and methanation reactors, due to the number of included components. Component and costs structures and corresponding scale factors are summarized in Appendix A.

process

There are different methanation concepts for catalytic methanation according to Rönsch et al. [20], which can be roughly classified into fixed-bed, fluidized-bed, structured reactors, and slurry reactors. Due to this high range of technologies, the reactor is not classified further in the present investigations. Due to the lack of studies on methanation specific technological learning, only very rough estimates of learning rates were possible. Steam methane reforming, as another Ni-based catalytic process, shows learning rates of 11% ± 6% [7], apart from that the technology is hardly comparable to methanation according to the underlying processes and reactions. Anandarajah et al. [21] suggest using learning rates from 15% to 20% for novel technologies in general. Even though the methanation process is used in industry over many years, increasing demands lead to further optimization efforts in relation to state-of-the-art methanation technologies. Hence, we assumed a moderate learning rate of 15% as a starting value for the methanation reactor for both catalytic and biological technology.

While the methanation reactor component contains the carrier material for the catalytic material, the catalyst itself is treated independently within the model. In terms of learning rates, this component can be roughly compared to the catalyst used in the definitions for the electrolysis cells. While the catalyst material itself is different (methanation primarily uses Ni-catalysts), cost reduction effects will be similar assuming that material costs stay constant, while only coating thickness is reduced. Hence, the same learning rate of 8%, which is a realistic assumption, as shown for electrolysis cells in [11], was used for the methanation catalyst as well.

Another essential component in the methanation reactor is heat management. Since operating temperature and heat management are highly dependent on the technology and the operation mode used in the individual reactors [20] and therefore tightly integrated with the reactor concept itself, a learning rate of 15% was assumed in this case, referring to the values used for the reactor component and provided for developing technologies by Anandarajah et al. [21].

The peripheral modules of the methanation system models (Electric Installation & Control System, Gas Conditioning and Balance of Plant) are treated equally to their counterparts in the electrolysis systems (cf. [11]). Since Electric Installation & Control System are expected to be similar for both technologies, i.e. catalytic and biological methanation, spillover learning effects between these are taken into account by not directly coupling the technological learning of this module to the cumulative productions of each individual technology, but rather to the cumulative production of methanation systems as a whole. Hence, the modules are set to an “independent” learning characteristic. Same accounts for the Gas Conditioning module, which does additionally consider past learning effects. Since processing of the methanation product gases (e.g. gas cleaning and drying, compression) is assumed to be similar to the processing of natural gas in general, the cumulative treatment of natural gas in the overall timeframe from 1900 to 2050 was used as a basis to evaluate learning effects of the Gas Conditioning module (cf. [22,23]). The resulting Learning Modules, initial cost shares and cumulative production dependency are summarized in Table 3 (electrolysis) and Table 4 (methanation).

2.1.2. *Scaling effects – economies of unit scale*

In addition to cost reductions by technological learning, economies of scale are considered as well in the assessment of CAPEX development for PtG technologies, by sizing-up of nominal plant power. A common method to describe these scaling effects is the use of a logarithmic relationship (cf. Eq. (4)), where C_b stands for the questioned equipment costs at the scale S_b (size, capacity, nominal power) of the component, and C_a and S_a represent the costs and scale of the known reference component, respectively. f is the scale factor applied to the technology in question. Since $f = 0.6$ can be used as a guideline scale factor for an initial approximate cost estimation of chemical appliances, this approach is also called the “six-tenth-factor rule” [24].

$$C_b = C_a * \left(\frac{S_b}{S_a}\right)^f \tag{4}$$

Applied to the technology costs found in literature (see Section 2.2), the resulting scale factor for electrolysis varies a lot, reaching values of 0.51–0.96 for AEC or 0.53–0.97 for PEMEC technology, respectively. Categorized according to the appropriate system size, it can also be seen that scale factors are generally increasing with increased scale. While for capacities < 5 MWel mean scale factors are around 0.69 (AEC) and 0.72 (PEMEC), for larger scale applications values of 0.9 and above are reached [25–31]. This can partially be explained by the fact that the electrolysis stack does not show potential for large cost reduction via EoS because of its modular design [32]. An increase in stack power due to an upscaling of the electrolysis cell is unlikely for many reasons (e.g. problems with leakage); therefore, the cell is limited in size. Similar effects are observed for scaling of methanation systems, while resulting scale factors are generally lower compared to electrolysis – ranging from 0.58 to 0.71 for catalytic and 0.39 to 0.73 for biological methanation, respectively – and the variation with different sizes is not as significant [33–37].

Table 3
Summary of learning curve and scaling effects calculation parameters for electrolysis.

Module	# components			initial cost share			learning curve dependency			scale factor		
	AEC	PEMEC	SOEC	AEC	PEMEC	SOEC	AEC	PEMEC	SOEC	AEC	PEMEC	SOEC
Cell Stack	9[1]	11[1]	9[1]	50%	60%	30%	direct			0.88[2]	0.89[2]	0.87[2]
Power Electronics	1 (lr = 12%)	1 (lr = 12%)	1 (lr = 12%)	15%	15%	30%	independent			0.75	0.75	0.75
Gas Conditioning	1 (lr = 7%)	1 (lr = 7%)	1 (lr = 7%)	15%	10%	6%	independent			0.60	0.60	0.60
Balance of Plant	1 (lr = 13%)	1 (lr = 13%)	1 (lr = 13%)	20%	15%	34%	direct			0.68	0.73	0.73

[1] Variable learning rate calculated through the model.

[2] Value for reference size and year.

Table 4
Summary of learning curve and scaling effects calculation parameters for methanation.

Module	# components		initial cost share		learning curve dependency		scale factor	
	catalytic	biological	catalytic	biological	catalytic	biological	catalytic	biological
Methanation Reactor	3[1]	2[1]	21%	17%	direct		0.67[2]	0.51[2]
Electric Installation & Control System	1 (lr = 12%)	1 (lr = 12%)	20%	21%	independent		0.75	0.75
Gas Conditioning	2 (lr = 7%)	2 (lr = 7%)	12%	13%	independent		0.69	0.71
Balance of Plant	6 (lr = 13%)	6 (lr = 13%)	47%	49%	direct		0.84	0.74

[1] Variable learning rate calculated through the model.
[2] Value for reference size and year.

Since the approach used to analyze learning curve effects does already provide a modular disaggregation of the investigated systems, a similar approach was used to evaluate scaling effects on a more detailed level, rather than for the system as a whole. Scale factors were evaluated per component based on relevant literature values [24,32,38,39]. The weighted mean values per module are summarized in Table 3 for electrolysis and in Table 4 for methanation.

While this approach allows to evaluate unit scaling effects on a component basis with a dynamic dependency on changing cost structures over time, it is still static according to the actual scale of the unit. Specifically, electrolysis stacks are expected to be limited in size, or rather their nominal power per unit, for various reasons (e.g., problems with leakage); thus, they are expected to be scaled by modularity (scaling by numbers) [32]. However, the maximum cell stack size is expected to increase as TRL and technological advances increase. To take those effects into account, a dynamic scale factor was implemented for the electrolysis cell stack based on an exponential function:

$$f = 1 - (1 - f_0)\cdot e^{-\frac{S}{S_{max}}} \tag{5}$$

where f_0 represents the before mentioned basic scale factor, S the questioned scale, and S_{max} the average maximum stack size for the period under study. This provides a scale factor that is dependent on the system scale itself and minimizes scaling effects for large-scale applications. The same effect was also applied to the reactor modules of the investigated methanation systems. The presumed maximum stack and reactor sizes related to the year of installation are presented in Table 2.

2.2. Current technology costs

A review of relevant literature on the investment costs for power-to-gas appliances showed that cost estimations vary a lot depending on the technology, system size and the year of installation (see Fig. 1 for electrolysis and Fig. 2 for methanation). It also showed that a comparison of available data is difficult, since it often misses significant information concerning system size, included peripherals (e.g. gas conditioning) or capacity reference (electric input, the lower heating value (LHV) output, the higher heating value (HHV) output). A recent review study on the investment costs of electrolysis by Saba et al. [41] which considered 30 years of cost studies, confirms the above-mentioned issues. However, the study also reveals a continuous reduction of projected nominal investment costs for electrolysis since the 1990 s.

To get an estimation on current technology costs, data for years of installation from 2015 to 2020 were analyzed in more detail. For alkaline electrolysis (AEC) this resulted in a cost range of 1090–2000 €/kW$_{el}$ for capacities < 1 MW$_{el}$ [26,41,42] and 800–1400 €/kWel for 1–10 MW$_{el}$ [10,26,43–45], while other references for the given period, ranging at 630–1000 €/kW$_{el}$, do not specify a dedicated system size [40,43,46,47]. For the proton exchange membrane (PEMEC) technology, the available data proposes costs of about 1500 €/kW$_{el}$ for systems below 1 MW$_{el}$ of installed capacity [41,42]. At average size of 1–10 MW$_{el}$, the current data has a significantly wide range from 960 €/kW$_{el}$ to 2100 €/kW$_{el}$ [10,26,44,45], which is the same range as for the rest of the analyzed data with unspecified capacity [40,42,43,46,47]. For high-temperature electrolysis (solid oxide electrolysis cell or SOEC) there is hardly any data available on the actual investment costs, especially at sizes of 1 MW$_{el}$ and above, due to the limited amount of installations up to now. The reviewed literature estimates actual costs of 2250–2500 €/kW$_{el}$ for system sizes of 5–10 MW$_{el}$ [10,26,40], excluding outliers of < 600 €/kW$_{el}$ [43,48,49]. For the methanation part the evaluation period was extended to the years 2012–2020, due to the lack of recent relevant data. Hence, the analysis

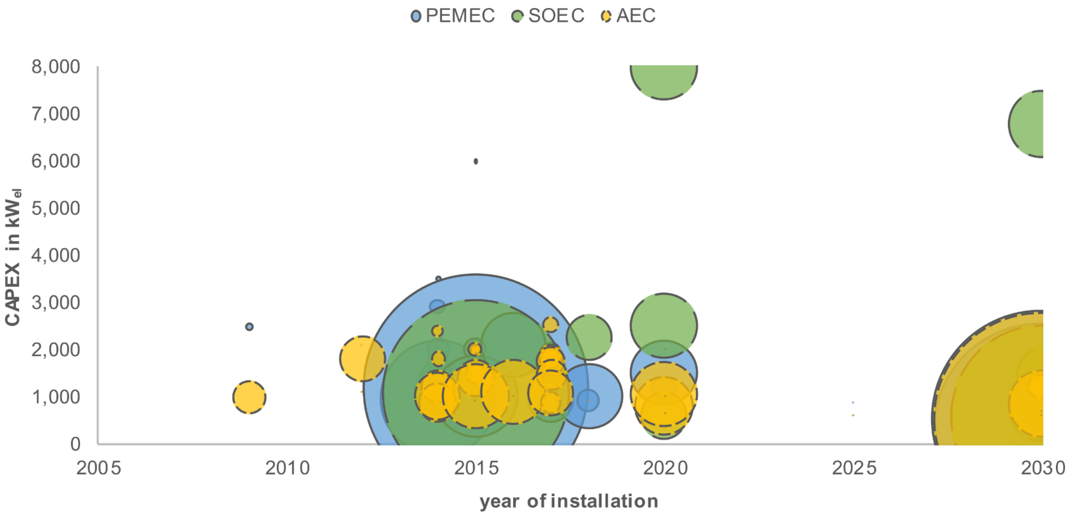

Fig. 1. Collected data on specific investment costs for different electrolysis installations related to the year of installation (bubble area indicates the rated power from 0.1 to 100 MW$_{el}$); based on [10,25–28,31,40–46,48,49,56,57].

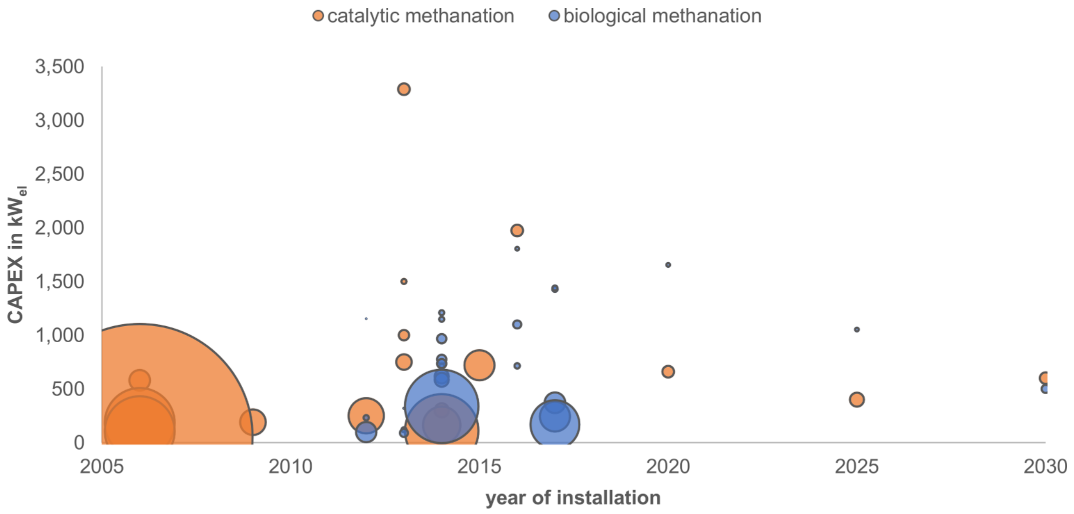

Fig. 2. Collected data on specific investment costs for different methanation installations related to the year of installation (bubble area indicates the rated power from 0.1 to 1000 MW_{SNG}); based on [25,28,33–36,50–54,58–61].

resulted in a cost range of 160–1970 €/kW_{SNG} (related to the LHV of SNG output) for chemical methanation [33,36,50,51], including nominal capacities of 1–30 MW_{SNG}. For biological methanation in the same category of size, the revealed cost are even lower at approximately 100–1430 €/kW_{el} [33,35,36,52–54]. On the contrary, recent state-of-the-art analysis in Germany presumes costs for biological methanation being higher in general [55].

For the subsequent calculations on learning and scaling effects, initial costs were defined for a reference size of 5 MW for both, electrolysis (related to electric input) and methanation (related to the lower heating value of SNG output), based on the data given above and average scaling factors. The resulting specific CAPEX for each technology, using 2017 as reference year, are summarized in Table 5.

2.3. Demand potentials for PtG products

Since the cumulative production of related technology equipment is the main driving factor for technological learning and hence for the reduction of future investment costs on the technology, the evaluation of future demand potentials for PtG products is of major relevance for the assessment of the costs of generating PtG products. The review of relevant literature has shown that estimation on future demand potentials for power-to-gas differs, depending on focused targets and sectors.

2.3.1. Assessment of literature data

Different studies conducted for the German power sector reveal a demand potential for electrolysis plants of 26–36 GW_{el}, depending on the amount of parallelly installed short-term energy storage capacities (e.g. battery energy storages) until year 2050 [62,63]. Another assessment estimate a significantly higher demand on PtG as a flexibility option in the German energy system of 89 and 134 GW_{el}, with and without short-term storage options, respectively, in the same timeframe [64]. Targeting economic aspects in a 100% RES based German electricity system, Breyer et al. [65] discovered a need for 43–45 GW_{el} of installed electrolysis capacity until 2040. The incorporation of mobility, industry and (residential) heat sectors reveals electrolysis capacity demands of about 130 up to 280 GW_{el} for the production of hydrogen and subsequent renewable energy carriers and feedstock materials through Power-to-Methane (PtM) and Power-to-X processes [65–67]. While comparable studies estimate a significantly lower demand, they also show that the potentials for PtG and PtX in the mobility and industrial sectors are expected to outperform the needs for long-term storage capabilities [68]. This is also confirmed in studies focusing on the mobility sector only [69,70].

When studying the transition of the energy system to RES in Spain by 2050, Bailera et al. [71] discovered a demand for PtG storage capabilities for excess RES of 7–19.5 GW_{el}. For a complete decarbonization of the energy system this demand could be even more than four times higher, according to Lisbona et al. [72]. Expected demands for the Italian energy system are in a comparable range of 30 GW_{el} by 2050 [73].

At the European level Blanco et al. simulated the demands for PtM [74] as well as for hydrogen and Power-to-Liquid [75] in a low-carbon EU energy system, using cost optimizations. Depending on the boundary conditions, like underground carbon storage options and positive drivers for PtM, the resulting potentials are almost equal in both cases, PtM and hydrogen, covering a wide range from 70 to 1000 GW_{el} of installed electrolysis capacity. For the PtM path the upper boundary indicates a methane production of 546 GW, which implies a coverage of 75% of the expected gas demand in the EU by 2050 [74].

At a global scale, the availability of relevant studies is rather limited. Pleßmann et al. [76] evaluated the demand for power plant and storage capacities for a global, decentralized and 100% RES-based electricity supply scenario, including long-term storage through renewable power methane (RPM) using PtG processes. The results showed a global demand for RPM storage of about 2360 GW_{el} electric input. Though the produced SNG can be injected to existing gas grids and used to provide renewable gas to other sectors besides power generation, the calculated demands only consider the needs for transformation of excess energy from volatile RES. Beyond that the study has significant limitations as it does not consider additional flexibility options, like geographically extended grid infrastructure or demand response [76].

However, reviews on the role of hydrogen in future renewable

Table 5
Initial specific CAPEX for electrolysis and methanation defined for calculations (reference year 2017).

Technology	Initial CAPEX	Reference power
Electrolysis		
AEC	1100 $€_{2017}/kW_{el}$	5 MW_{el}
PEMEC	1200 $€_{2017}/kW_{el}$	(electric input)
SOEC	2250 $€_{2017}/kW_{el}$	
Methanation		
Catalytic	600 $€_{2017}/kW_{SNG}$	5 MW_{SNG}
Biological	650 $€_{2017}/kW_{SNG}$	(LHV gas output)

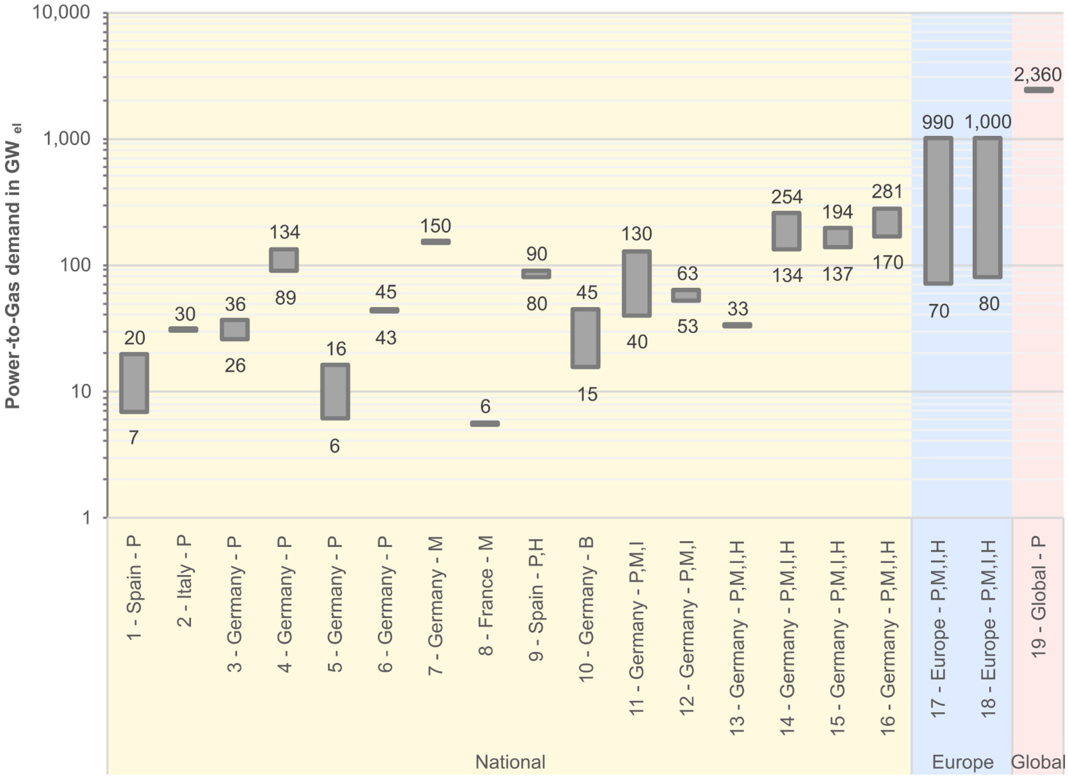

Fig. 3. National and international PtG demand potentials for year 2050 according to relevant literature; based on [62–76,80–83] (P…power sector, M…mobility sector, H…heat sector, I…industry sector, B…based on biogas).

energy scenarios show, that the results for the global demands are covering a wide range and are highly dependent on the used energy models, considered sectors of supply and application as well as the potential utilization pathways [77,78]. Additionally, a large-scale roll-out of PtG plants also requires a significant increase of RES for the production of renewable gases, including the associated impacts on future energy systems (cf. Zappa et al. [79]). Fig. 3 summarizes the findings stated above. As the Figure shows, the comparability of these studies is restricted due to the variations in their regional and sectoral scopes, as well as the underlying constraints and boundary conditions. Due to the localization to regional energy systems and individual sectors an extrapolation of national or European results to estimate global demands is limited.

2.3.2. Deduction of global PtG demands

To evaluate potential cost reductions for large-scale PtG plants according to learning and scaling effects within widely renewable future energy systems, an estimation of global market potentials is necessary to put them in context with the level of technology distribution. Due to the beforementioned limitations of regionally or sectoral constrained energy models, the present study focusses on industrial demands for hydrogen and SNG, which are considered as the main purpose for large-scale implementations.

According to the recent IEA report on the future of hydrogen and its potential within future energy systems [84], the current (2018) demand for pure hydrogen is about 74 Mt per year, almost all of this amount produced from fossil fuels (76% from natural gas, 23% from coal). The majority of today's hydrogen is used as feedstock material in the refinery industry and the production of ammonia, followed by the production of methanol. Up to 2030, the annual hydrogen demand for ammonia is expected to grow by about 7 Mt and reaching a plus of 12 Mt until 2050. Similar growths in hydrogen demand are expected for the synthesis of methanol (+6.5 Mt until 2030, +10.5 Mt until 2050), resulting in a total demand of > 96.4 Mt per year in 2050 following current utilization pathways [84]. Presuming a coverage of these demands by production from renewable electricity, this would result in an electrolysis capacity of about 4590 TWh_{el} (with an efficiency of 70% related to LHV for the electrolysis). In addition, the iron & steel industry is considered as major consumer of renewable hydrogen in future energy scenarios following the DRI (direct reduced iron) route for a decarbonization of today's steelmaking processes [85]. The current demand for hydrogen for DRI processes in the steel industry accounts for about 4 Mt per year. This amount is expected to increase significantly to 8 Mt/yr, or even 9–11 Mt/yr based on zero-emission scenarios, in 2030, and 62 Mt/yr (47–67 Mt/yr for zero-emission) in 2050, respectively [84]. Together, with the beforementioned demands in the chemical industries, this would result in an electrolysis production capacity of about 6640–7590 TWhel, or an installed electric power of 1900–2170 GW_{el} assuming a cost optimized operation at 3500 full load hours (FLH) as found in [12] and [84] (or 1310 GW_{el} for chemical industry demand only).

Demand potentials for PtM and thus implementation potentials for methanation capacities are more difficult to estimate. As stated in Section 2.3.1, calculated demands for PtM in future energy systems are highly dependent on the energy scenario, regional boundaries and technical limitations, such as interlinking of grid infrastructures. Therefore, PtM is only considered in terms of energy storage capabilities. Despite its limitations according to the simplifications of the used model, Pleßmann et al. [76] provide an estimate on PtM storage potentials for renewable electricity supply on a global scale. The results show a PtM demand of 2360 GW_{el} or 1960 TWh_{SNG}. Presuming appropriate intermediate storage capacities for the hydrogen and thus an operation of the methanation at 7000 FLH, the energy storage demand would relate to about 280 GW_{SNG} production capacity. On the other hand, a direct transformation of the electric capacity going into electrolysis to SNG without intermediate storage, methanation demand would rise up to 1360 GW_{SNG} (with an efficiency of 70% for the electrolysis and 82.5% for methanation related to LHV). These PtM

Table 6
Investigated deployment scenarios for large-scale PtG capacities until 2050.

Scenario	Electrolysis capacity in GW_{el}	Methanation capacity in GW_{SNG}	Notes
Low	1310	0	● PtH_2 for chemical industry (ammonia, refinery, methanol synthesis) only ● No PtM
Moderate	3670	280	● PtH_2 for chemical industry ● PtM with intermediate H_2 storage
High	4530	1360	● PtH_2 for chemical & steel industry ● PtM without intermediate H_2 storage

demands in return also increase the needs for electrolysis capacities accordingly.

The resulting overall demands are covered by three scenarios in the learning curve analysis (cf. Table 6). To put the resulting figures in context, the PtM potential covered is about 5% of the global gas demand in 2040 for the Sunstainable Development Scenario (SDS) stated in the most recent IEA World Energy Outlook [86]. In contrast, the estimated electrolyzer capacities relate to 5–15% of today's (2018) global electricity generation, or 20–60% counting only renewables [86].

For the annual distribution of electrolysis capacities this would imply an compounded average annual growth (CAAGR) of 17–21% globally, based on an overall global capacity of around 8 GW_{el}, as of 2014 [87]. To put that in context, power generation from wind experienced an average growth of 19% per year in the period 2007–2017, whereas PV has seen an even higher growth rate of nearly 48% from 2012 to 2017, especially driven by Asian regions [88,89]. Compared to these, the proposed growth rates for PtG seem reasonable, even though a constant high growth throughout, up to 2050, is still optimistic. According to methanation, currently PtG plants with a total capacity of 14.5 MW_{el} globally produce methane [3]. This would result in a CAAGR of 36–43% for moderate and high scenarios throughout to 2050. Considering saturation effects, early growth rates for PtG will need to be even higher and therefore not reasonable for a period of 30 years. However, looking at methanation reactors besides from PtG applications, including CO methanation from coal gasification, overall in operation capacities reach up to 33 GW (reference year 2016) [20]. Hence, the CAAGR decreases to more reasonable values of 7–13% for methanation.

2.4. Techno-economic assessment

To evaluate the effects of technological learning and plant scaling on production costs of renewable gases from power-to-gas plants, a techno-economic assessment for different plant configurations and capacities was performed. These costs were calculated by assessing the annual total plant costs related to the energy content (LHV) of the annually produced gas. This approach is comparable to the calculation of LCoE [90,91]. The methodology used to calculate the total annual costs is based on the calculation of economic efficiency using the annuity method [92]. The annuity of total annual payments A is stated as the difference between the annuity of proceeds A_p and the sum of the annuities of capital-related A_C, demand-related A_D, operation-related A_O and other A_M costs (see Eq. (6)).

$$A = A_P - (A_C + A_D + A_O + A_M) \tag{6}$$

The executed calculations presume a general interest rate of 4% and an observation period of 20 years. By contrast to the guideline [92], price change factors and price-dynamic cash values were omitted. For detailed information on individual factors and values used in the techno-economic evaluation see Appendix A.

Variable costs (and proceeds) C_{var} that are dependent on input and output streams related to the time of operation (electricity and CO_2 input costs, proceeds from by-products like oxygen or heat) are added according to their occurrence. The levelized costs of PtG (LCoP), representing the generation costs for SNG (or hydrogen) related energy content, are then calculated as follows:

$$LCoP = \frac{-A + \sum_i C_{var,i}}{E_{out}} \tag{7}$$

2.4.1. Capital and operational costs

The capital-related costs mainly include investment costs for system components and expected replacements during the observation period T:

$$A_C = (I_0 + I_1 + \cdots + I_n - R)\cdot a \tag{8}$$

where I_0 represents the initial investment amount, $I_1 \cdots I_n$ the cash values of first to n-th procured replacement, R the residual value of the investment, or last replacement, respectively, and a the annuity factor. Replacements and the residual value are hence dependent on the interest-rate factor q and the deprecation period T_N.

$$I_n = \frac{I_0}{q^{n\cdot T_N}} \tag{9}$$

$$R = I_0\cdot\frac{(n+1)\cdot T_N - T}{T_N\cdot q^T} \tag{10}$$

The annuity factor a can be derived as follows:

$$a = \frac{q^T\cdot(q-1)}{q^T - 1} \tag{11}$$

Besides direct capital costs of the main equipment (electrolyzer, methanation, storage tank), annuity of additional costs A_M for the construction and commissioning of the PtG plants was considered by using overhead factors, also known as *Lang* or *Chilton* factors [24,38,93] (see Appendix A).

Operation-related costs include costs for maintenance and inspection O_M as well as insurance and administration O_I. The according annuity is calculated as:

$$A_O = (O_M + O_I)\cdot a \tag{12}$$

Demand-related costs cover expenses for operational materials, like water and CO_2. Together with proceeds that are expected to arise from the sale of by-products, such as heat and oxygen, these were considered as variable costs C_{var} in the techno-economic evaluation.

2.4.2. Electric power supply

For the evaluation of electricity supply costs three different power sources were considered:

1. supply from wind power plant (PtG-wind),
2. supply from PV power plant (PtG-PV) and
3. supply from public grid (PtG-grid)

According to Kost et al. [94], current (2018) LCoE from onshore wind farms in Germany are ranging from 40 to 82 €/MWh depending on the location. Despite providing a higher amount of FLH, electricity

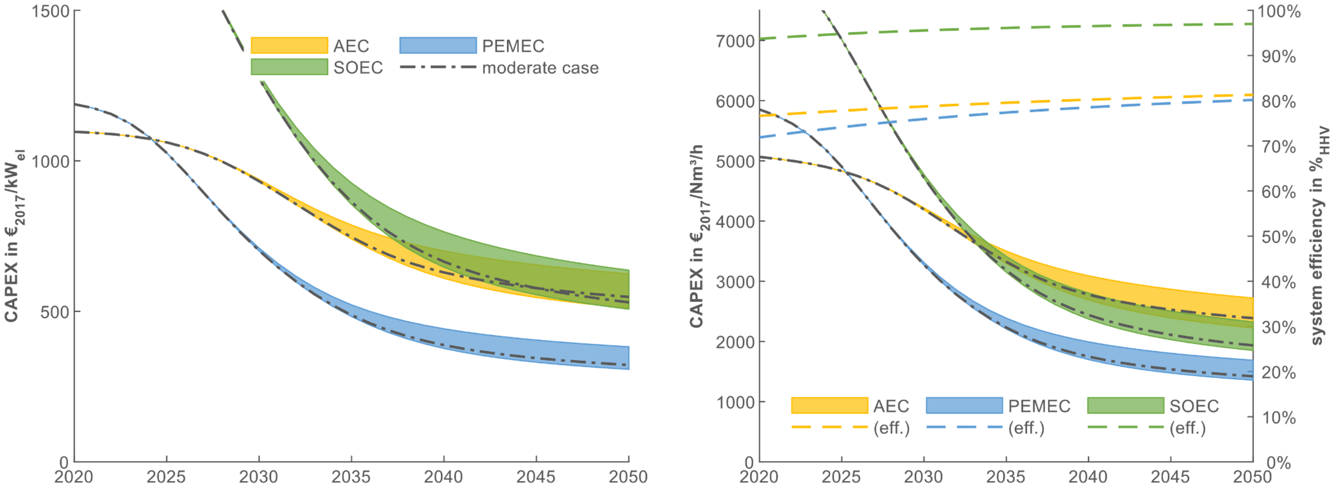

Fig. 4. Estimated ranges for technological learning of electrolysis related to the defined deployment scenarios (left. based on electric power; right. based on hydrogen output with developing efficiencies).

costs from offshore generation are stated significantly higher, at about 75–138 €/MWh. These costs are expected to decline in the medium term, reaching 35–71 €/MWh (onshore) and 57–101 €/MWh (offshore), respectively, until year 2035. Another study executed by Agora Energiewende [95] expects even lower costs for electricity from onshore wind farms estimating 30–60 €/MWh by 2030 to 25–50 €/MWh by 2050, for plants located in Germany. Assuming a more optimistic scenario with a higher potential for cost reduction, these costs could also decline to 25–45 €/MWh (2030) and 20–35 €/MWh (2050). The LCoE from open space PV power plants in Germany currently (2018) reach costs of about 37–68 €/MWh, depending on the location. These costs are expected to decline to 21–39 €/MWh by 2035 [94]. An extended study by Fraunhofer ISE [96] estimated potential long–term LCoE of about 25–44 €/MWh for southern Germany, and 18–31 €/MWh in southern Spain by 2050, based on a proposed learning curve and scaling effects. For international regions the IEA states current LCoE from 55 €/MWh (United States) to 95 €/MWh (EU) for onshore wind and 50 €/MWh (India) to 105 €/MWh (EU) for PV in their Stated Policies Scenario. Perspectives for 2040 are at 55–90 €/MWh for onshore wind and 50–90 €/MWh for PV, respectively [86]. However, these values are expected to be lower in a Sustainable Development Scenario. Hence, PtG product costs will, independent of CAPEX development, be highly dependent on to location of implementation. The electricity costs presumed for wind and PV in the techno-economic assessment in different periods of observation are presented in Appendix A.

In addition to the direct supply from renewable energy sources, we assessed production costs for PtG products based on the participation in the electricity green spot market, which would be reasonable in terms of constant production for industrial utilization of PtG products. However, to ensure an economic advantage of PtG over fossil production of hydrogen, the global warming potential (GWP) of the utilized electricity must not exceed 190 g CO_2 per kWh. For SNG production by methanation, the GWP limit further decreases to about 70–120 g CO_2 per kWh, depending CO_2 separation effort is included, due to efficiency impacts [97,98]. This has to be considered in the assessment for different electricity markets. The calculations on spot market electricity presume that the PtG plant is operated at times with the lowest electricity prices, and thus mean costs for electric power supply are dependent on the annual full load hours (FLH) of operation. The price curves used for the evaluation of electricity supply costs are based on the Austrian spot market data for the year 2017, at a time periods of 60 min [99].

3. Results and discussion

3.1. Resulting investment costs

Based on the evaluated demand potentials for PtG production capacities, we calculated learning curves related to the year of installation, presuming that cumulative production matched the demand potential by year 2050. For annual interim values a logistic growth function was assumed for the overall production capacities. Following boundary conditions were presumed:

- Current cumulative capacities are at 21.1 GW_{el} for electrolysis, according to [10,11], and at about 33 GW_{SNG} for methanation, according to [20].
- Recent surveys indicate that electrolyzer industry could ramp up production to 2 GW/year by 2020 (from 100 MW/year in 2018) [4]. This value was used as a reference for production scale-up to be reached by 2025 from current perspective.
- The total annual productions for electrolysis and methanation capacities were then subdivided into the individual technologies. For electrolysis these production shares were presumed based on the estimations given in [47], targeting about 40% each for PEMEC and SOEC and around 20% for AEC technology by 2050. For methanation an initial division of 90% vs. 10% (catalytic vs. biological methanation) was presumed, changing to 60% vs. 40% by 2050 (see Appendix A).

The results show that under presumed conditions, CAPEX are expected to decline significantly for all investigated technologies due to technological learning (see Figs. 4 and 5). Solid-oxide electrolysis is about to compete with alkaline cells in terms of capital costs in the long-term, while PEMEC are about to undercut AEC in near future and establish the lowest cost solution. This is in line with the findings in [11] at an increase of relative cumulative production by a factor of $5 \cdot 10^2$, considering the difference in initial costs based on the literature review. Under consideration of developing efficiencies based on Smolinka et al. [47] (cf. Fig. 4, right), SOEC are expected to become economically competitive to alkaline cells even sooner based on hydrogen output. Especially in PtM processes synergies could be used, considering optimized thermal integration of the exothermal methanation process and therefore maximizing overall efficiency of the PtG system [100–102].

For methanation, bio-based technologies are expected to reach lower cost levels compared to its chemical counterpart at the referenced size of installation. Despite continuously higher production shares for

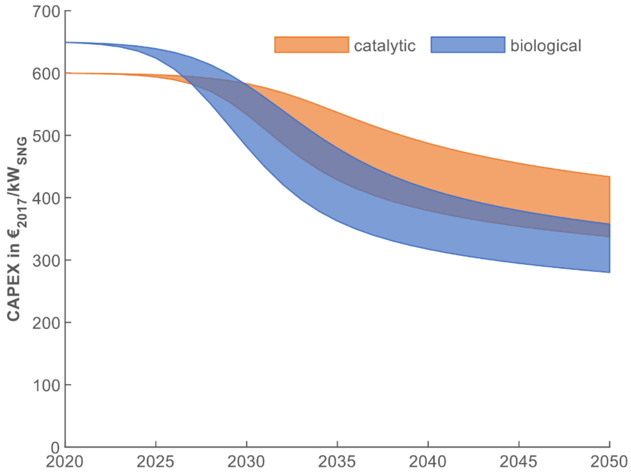

Fig. 5. Estimated ranges for technological learning of methanation related to the defined deployment scenarios (moderate and high).

the catalytic technology over the investigated time horizon the potentials for technological learning are higher for biological methanation due to the lower starting value of cumulative productions and therefore a higher relative increase of productions. The learning rates are similar and decrease only slightly over time, from 12.1% to 11.7% (catalytic) and 12.3% to 11.8% (biological), respectively. Concerning methanation technologies, there is a lack of relevant literature on technological learning. Looking at steam methane reforming (SMR) as another Ni-catalyst based chemical process compared to catalytic methanation, it shows similar learning rates of 11 ± 6% [7]. To assess the values for biological methanation, the process of biogas methanation can be referred to, for which similar experience rates of about 12% were found by Junginger et al. [103]. To evaluate overall cost reduction potentials, learning curves resulting from high deployment scenario are used for subsequent analysis, if not stated differently.

The evaluation of scaling effect (in terms of unit scaling) revealed additional cost reduction potentials for future PtG plants. Due to the component-based application of scaling factors the characteristics of these effects vary noticeably between the investigated technologies. Compared to the reference size values, these scaling effects abate relatively fast for AEC and PEMEC based electrolysis systems with an increasing order of magnitude, whereas SOEC based systems provide a more significant dependency of CAPEX to system scale. This is explained by higher cost share of better scalable peripherals (e.g. Power Electronics, BoP) over the Stack module with limited scalability in the SOEC system (cf. Fig. 6). Related to the year of installation, average scaling factors for the investigated electrolysis systems are generally decreasing – resulting in higher scaling effects – as better scalable modules show lower potentials for technological learning compared to the stack modules, which results in increasing cost shares and thus higher influence on scaling for these parts. At least for PEM and alkaline electrolysis systems, these results are close to the few values found in the literature for capacities of about 10 MW$_{el}$ [10,26,42,45]. For scales of 50 MW$_{el}$ and above the literature values [15,44] seem to over-estimate capacity scaling effects. Comparative values for SOEC are fitting according to capacity scaling, but seem to over-estimate learning effects. The high uncertainty in literature on learning rates for SOEC (28% ± 16%) was also discussed in [11], whereas the component-based model derived from comparative technologies is more conservative in this context. However, the resulting learning effects are also dependent on presumed future technology shares (cf. sensitivity analysis in Section 3.3).

For the methanation technologies (cf. Fig. 7), scaling effects are more significant due to rather low scaling factors through all modules (cf. Table 4), being at about 0.81 (2020) to 0.77 (2050) on average, for both technologies. However, these investigations suggest that scale factors < 0.6, which were also found in the literature study [33,34], seem to be rather low. Therefore, we have to assume that these values do incorporate additional effects, like technological learning, besides pure unit scaling. For both technologies, the estimations are similar to the predictions for large scale applications of 50–100 kW that were gathered in the literature study [34–36,59] in terms of capacity scaling. However, acc. to Götz et al. [37], biological methanation may not be an option for a large scale implementation due to the need for large specific reactor volumes and fewer possibilities for waste heat utilization.

3.2. Effects on product costs

Analyzing production costs for corresponding PtG products (hydrogen and SNG), the results show the expected impact of decreasing CAPEX. As shown in Fig. 8, considering PEM electrolysis and catalytic methanation as reference system, SNG production costs are expected to go down by about 75–80% in the high deployment scenario compared to current values based on presumed techno-economic parameters. However, the levelized costs for SNG is still expected to be significantly above the current reference costs for natural gas (EU–28 average price for non-household consumers in 2019 was 3.27 €-cent/kWh [104]).

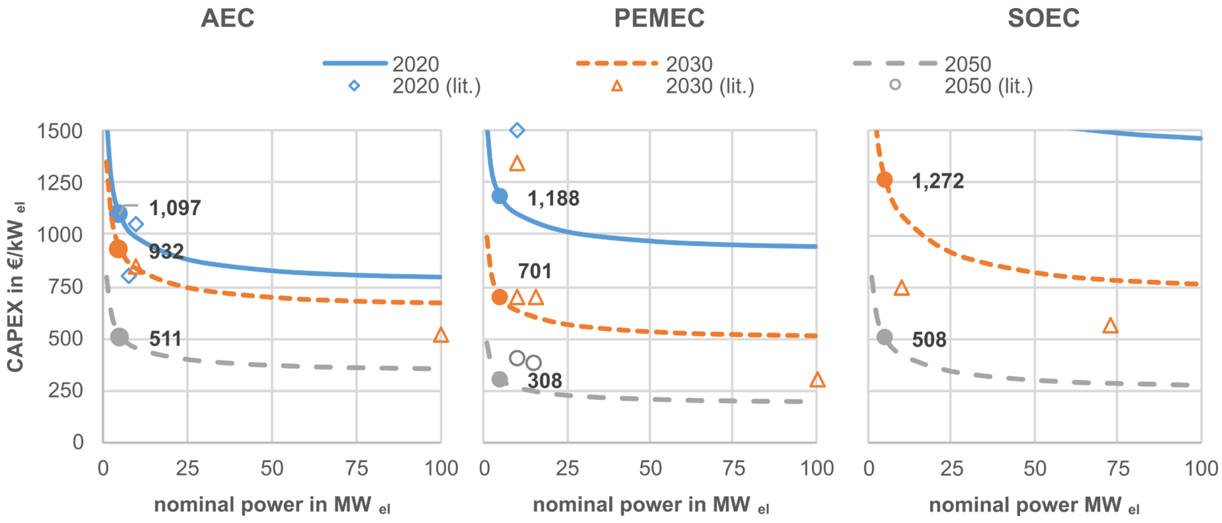

Fig. 6. CAPEX development for electrolysis systems compared to projections from the literature study (lit.) for installations ≥10 MW$_{el}$ (marked and labelled values refer to reference size of 5 MW$_{el}$).

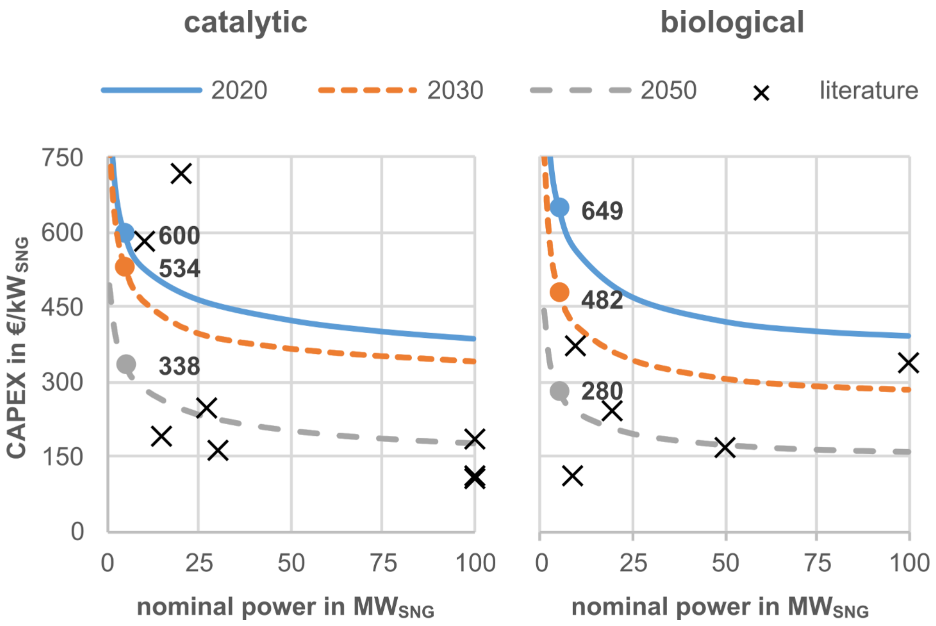

Fig. 7. CAPEX development for methanation systems compared to projections from the literature review for installations ≥10 MW$_{SNG}$ (no target time frames available; marked and labelled values refer to reference size of 5 MW$_{SNG}$).

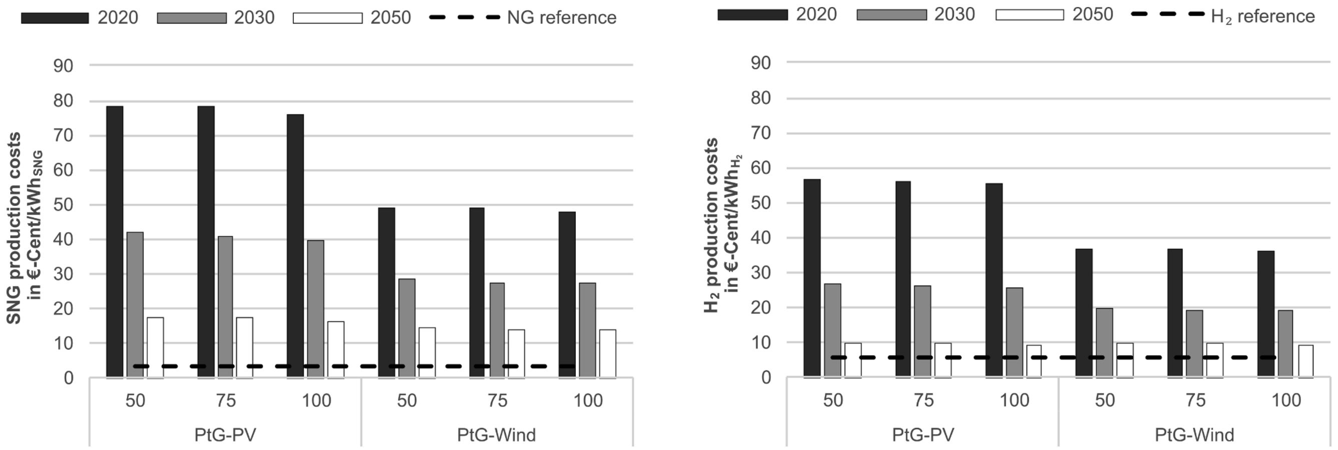

Fig. 8. Resulting production costs for SNG (left) and hydrogen (right) compared to 2019 reference values for fossil production.

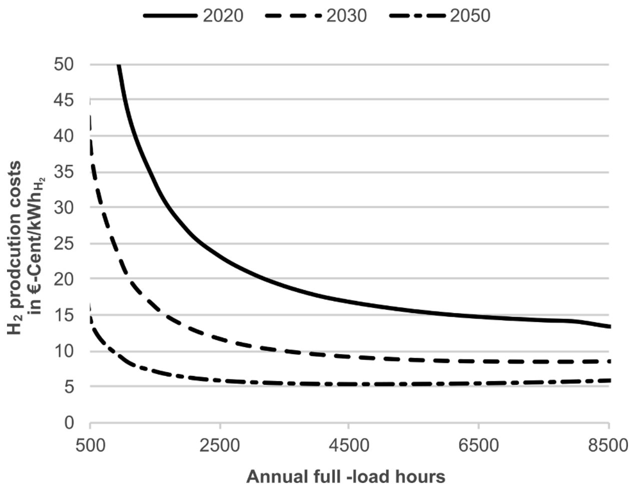

Fig. 9. H_2 production costs related to annual FLH considering spot market electricity costs.

Considering hydrogen production on its own, cost reduction effects from technological learning and scaling tend to provide economic competitiveness of large-scale PtG against current fossil production of hydrogen in the long-term. However, taking CO_2 pricing for fossil products into account, prices of about 120 €/t CO_2 (wind supply) to 160 €/t CO_2 (PV supply) would still be needed to close the gap for hydrogen production, given the according GWP found by Reiter et al. [97].

Evaluating hydrogen production costs and spot market electricity costs (cf. Section 2.4.2) in relation to annual FLH showed that the impact of specific CAPEX is decreasing significantly according to the calculated learning effects. Fig. 9 also shows that the range of optimal economic operation is getting wider and shifted to lower FLH, with decreasing CAPEX (presuming stable volatility of future electricity prices).

3.3. Sensitivity analysis

The results of the learning curve calculations presume that the influence of cumulative production is only modest at the given overall market potentials for electrolysis. Comparing low and high potential scenarios, the differences in expected CAPEX for electrolysis are at

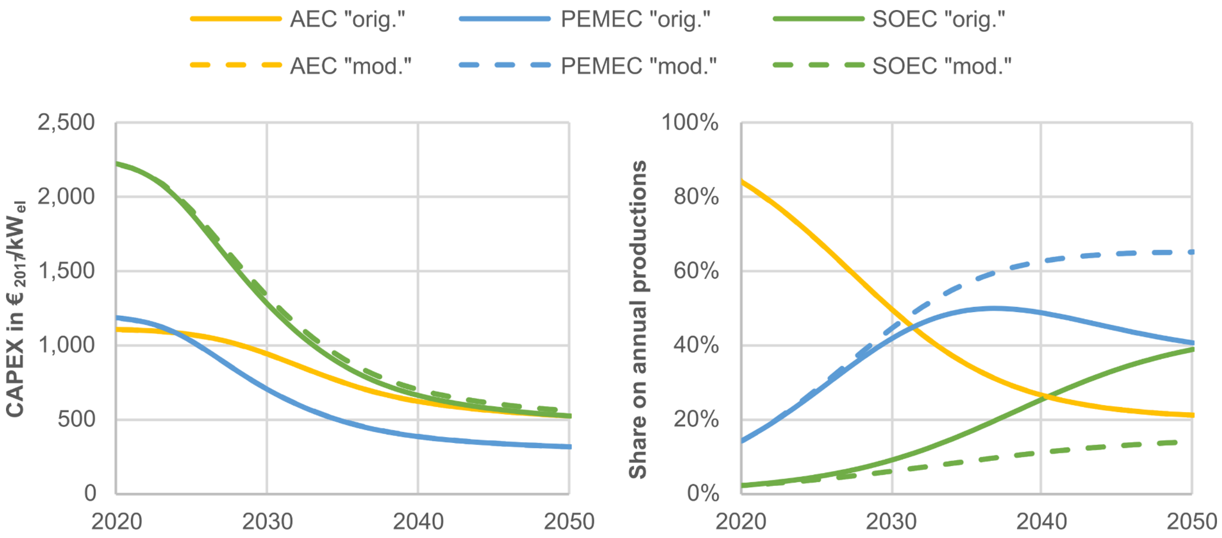

Fig. 10. Impact of presumed production shares (right) on electrolysis learning curves (left) ("orig.": original shares; "mod.": modified shares).

17–19% for a variation of cumulative productions of a factor 3. For methanation these effects are higher according to the higher differences on cumulative productions in the scenarios by a factor of 5, resulting in a variation of methanation CAPEX by 44% (biological) to 57% (catalytical) between moderate and high scenario. Hence, the consideration of system lifetime in terms of replacements, which would increase cumulative productions of electrolysis stack and methanation reactor modules by about 35% and overall system production by $< 1\%$[1], affects target costs in 2050 by $\leq 5\%$.

Application of presumed technology shares to more conservative technologies (higher values for PEM electrolysis and catalytic methanation, lower shares for SOEC) showed that influences of overall cumulative production volumes on evaluated learning curves are negligible at given scales (cf. Fig. 10 for electrolysis and Appendix A for methanation). Beyond that, the presumed learning rates itself are the most relevant impact factor for the development of the calculated learning curves. Sensitivity analysis have shown that a variation of the underlying learning curves for peripheral components (e.g. Gas Conditioning, BoP) by ± 25%, influences the overall learning curves by -13% to $+18\%$ in high volume scenarios depending on the technology. Hence, a continuous observation of cost development over future productions is a mandatory aspect for the validation and adoption of PtG-related learning curves and corresponding measures.

Concerning the influence of the variation of scaling factors on the resulting cost curves, the sensitivity analysis showed, that a general reduction of scaling factors by 10% for peripheral electrolysis system components (scaling effects for stack module are already limited for the investigated scales) would affect costs by up to 4% for 2020 large-scale assessments (50–100 MW_{el}). These effects slightly increase with decreasing base costs, reaching 6–7% for 2050 values. For the investigated methanation technologies these effects are significantly higher, due to scaling effects being lower in general. For 2020 results, a general reduction of the scaling factors by 10% would lead to a further cost decrease by 12–14% for 50–100 MW_{SNG} installations, or up to 20% for 2050 values. However, the component-based analysis of scaling factors based on independent chemical engineering approaches [24,38] is expected to increase accuracy significantly over common rules for estimation (e.g. six-tenth-factor rule [24]). To further improve estimates for large-scale implementations, a reinvestigation of scaling effects and base costs should be considered once additional data from implemented multi–MW plants is available.

4. Conclusions

This study evaluates a cost development of large-scale power-to-gas (PtG) applications in short-, mid- and long-term scenarios, based on a holistic techno-economic assessment. Our investigations on future demands for renewable gases, primarily hydrogen and synthetic natural gas (SNG), has revealed demands of 1310–4530 GW_{el} for electrolysis and up to 1360 GW_{SNG} for methanation capacities till 2050 to achieve a sustainable transition of European and global energy systems to renewable energy sources. The effects of technological learning, caused by the need to significantly increase production rates for corresponding technologies and thus increase the cumulative production by several orders of magnitude, are expected to reduce specific CAPEX by about 30–75% (electrolysis) and 30–60% (methanation), respectively, for future implementations, depending on the technology. However, an upscaling of average plant capacities to multi-MW scales will additionally be necessary for future PtG plants to be economically competitive to incumbent technologies. In that context, presented calculations revealed cost reduction potentials of > 75% for the investigated technologies for capacities of 50 MW and beyond (from a 5 MW reference scale).

In addition to CAPEX development, a variety of parameters could have an impact on the economic feasibility of the PtG process chain. These effects were evaluated by assessing production costs for hydrogen and SNG for different PtG technologies. The calculated results showed that the product costs are mainly driven by electricity supply costs, besides CAPEX and overall plant efficiency. However, the levelized costs of product gas are highly dependent on the annual FLH, despite showing different impacts based on the evaluated scenario. While scenarios with constantly low electricity prices, as presumed in PV and wind supply scenarios, would achieve lowest production costs at high FLH, their supply profiles, especially for peak load coverage, do not support an according operation. For the operation based on the actual spot market prices generation costs are dependent on average electricity prices and their future development. With decreasing CAPEX best cost operation will move from high (> 7000 h/a in 2020) towards a low and wider (3000–6500 h/a in 2050) range of FLH.

Even though short- and mid-term estimations are around 30–80 €-cent/kWh_{SNG}, it could be shown that future generation costs for SNG from PtG can reach values of 15 €-cent/kWh_{SNG} and below in large-scale plants. However, even in the lowest cost considerations and grid supply, resulting production costs are still around 8 €-cent/kWh_{SNG} in a long-term perspective and thus significantly higher than today's energy costs for natural gas, representing an obvious benchmark for future PtG

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[1] presuming a stack/reactor lifetime of 10 years and system lifetime of 25 years based on [40,47].

plants. PtG for hydrogen production is expected to reach production costs of about 10 €-cent/kWh$_{H2}$ by fulfilling the high deployment scenario. However, to reach economic competitivity with fossil technologies, additional ecological measures, like pricing of CO_2 emissions in a range of 120 €/t CO_2 and above, will still be mandatory. Hence, beyond supporting investment and therefore enabling scaling effects for PtG technologies, it will be essential to introduce additional measures and facilitative regulatory frameworks in order to establish power-to-gas as a competitive technology to fossil energy sources.

CRediT authorship contribution statement

Hans Böhm: Conceptualization, Methodology, Formal analysis, Writing - original draft, Visualization. **Andreas Zauner:** Validation, Data curation. **Daniel C. Rosenfeld:** Data curation, Writing - review & editing. **Robert Tichler:** Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This contribution has received funding from the European Union's Horizon 2020 Research and Innovation Program under grant agreement No. 691797 - STOREandGO project (https://www.storeandgo.info). The support by the association Energy Institute at the Johannes Kepler University Linz is gratefully acknowledged.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apenergy.2020.114780.

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Journal of CO_2 Utilization 41 (2020) 101219

Contents lists available at ScienceDirect

Journal of CO_2 Utilization

journal homepage: www.elsevier.com/locate/jcou

Assessing the potential of carbon dioxide valorisation in Europe with focus on biogenic CO_2

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ARTICLE INFO

Keywords:
Carbon capture and utilization (CCU)
Green CO_2
Fossil CO_2
Stationary emissions
Potential analysis

ABSTRACT

This study investigates the theoretical potential and limitations of carbon dioxide sources for technical valorisation approaches. On the one hand the emission of greenhouse gases, especially CO_2, must be rigorously reduced in order to achieve the European and global climate objectives. On the other hand, CO_2 is becoming increasingly valuable as a resource for industries and new disrupting technologies on CO_2 utilization. Therefore, the potential of CO_2 obtained from different biogenic and fossil sources in Europe is discussed for a comparative evaluation. These sources are classified according to their emitting processes and industry sectors, respectively. The resulting valorisation potentials are then calculated from statistical data for CO_2 generating processes in Europe, complemented and verified by relevant papers and reports. This study demonstrates the European potential of capturing and utilizing biogenic and fossil CO_2. In Europe, 69.7 Mt/a CO_2 are estimated to be produced by biogas upgrading, biogas combustion, as well as bioethanol and other fermentation processes. Additionally, 437 Mt/a CO_2 are produced by solid biomass combustion. This accounts for a theoretical potential of up to 506.7 Mt/a biogenic CO_2 currently available, which is nearly seven times the amount of the current European industrial CO_2 demand. The utilization potentials from these theoretical amounts are limited by attainable capture rates, reaching from 10 % to 90 % primarily depending on the dilution of the emitting source.

1. Introduction

Carbon dioxide (CO_2) is mainly seen as a global hazard due to its properties as a greenhouse gas (GHG). In fact, it is also a valuable resource for various state-of-the-art and innovative technologies and processes. There are various CO_2 utilization pathways, some of which were established decades ago while others are still being investigated. Typical direct utilization pathways involve beverage carbonization and horticulture production (greenhouses), using CO_2 as a working fluid. Traditional chemical industries like that of urea, polyurethane (PUR) and various acid and carbonate production processes use CO_2 as chemical feedstock. More recent applications involve micro algae production or new processes to produce well-known products [1,2]. Mikulčić et al. [3] conducted an extensive review of Carbon Capture and Utilisation (CCU) technologies and utilization pathways of captured CO_2.

CCU provides ways to reduce carbon emissions while tapping into this resource. The present paper investigates the theoretical mass potential and limitations of biogenic, and thus green, CO_2 sources for the implementation of such utilization options. Various studies explored the technical opportunities for CCU [4–8], emphasizing the disruptive potential of some of the potential future applications [9]. Possible CCU pathways can be categorized based on the energy supply for activating the stable CO_2, the various synthesis processes or the marketable products (see Fig. 1 for the latter two). Taking the pursuit of a sustainable closed carbon cycle economy and the available CCU pathways into account, it is probable that CO_2 will become a (even more) valuable resource in the future, especially if it is biogenic and thus renewable.

According to Billig et al. [10], global CO_2 demand is estimated to increase from 197 Mt/a in 2013 to 250 Mt/a by around 2026. Chauvy et al. [11] estimated a global demand potential of 590 Mt/a based on a

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Abbreviations: bio – CCU, biogenic CCU; bio – CCS, biogenic CCS; bio-CNG, biological Compressed Natural Gas; CCP, Carbon Capture Project; CCS, Carbon Capture Storage; CCU, Carbon Capture and Utilisation; CHP, Combined Heat and Power; DAC, Direct Air Capture; EBA, European Biogas Association; EIGA, European Industrial Gases Association; EU, European Union; EU ETS, EU emission trading system; FAB, Food and Beverage; GIE, Gas Infrastructure Europe; IPCC, Intergovernmental Panel on Climate Change; ISBT, International Society of Beverage Technologists; MOFs, Metal-Organic Frameworks; PSA, Pressure Swing Adsorption; PUR, Polyurethane; WBA, World Biogas Association; VOC, Volatile Organic Compound

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https://doi.org/10.1016/j.jcou.2020.101219
Received 4 May 2020; Received in revised form 10 June 2020; Accepted 14 June 2020

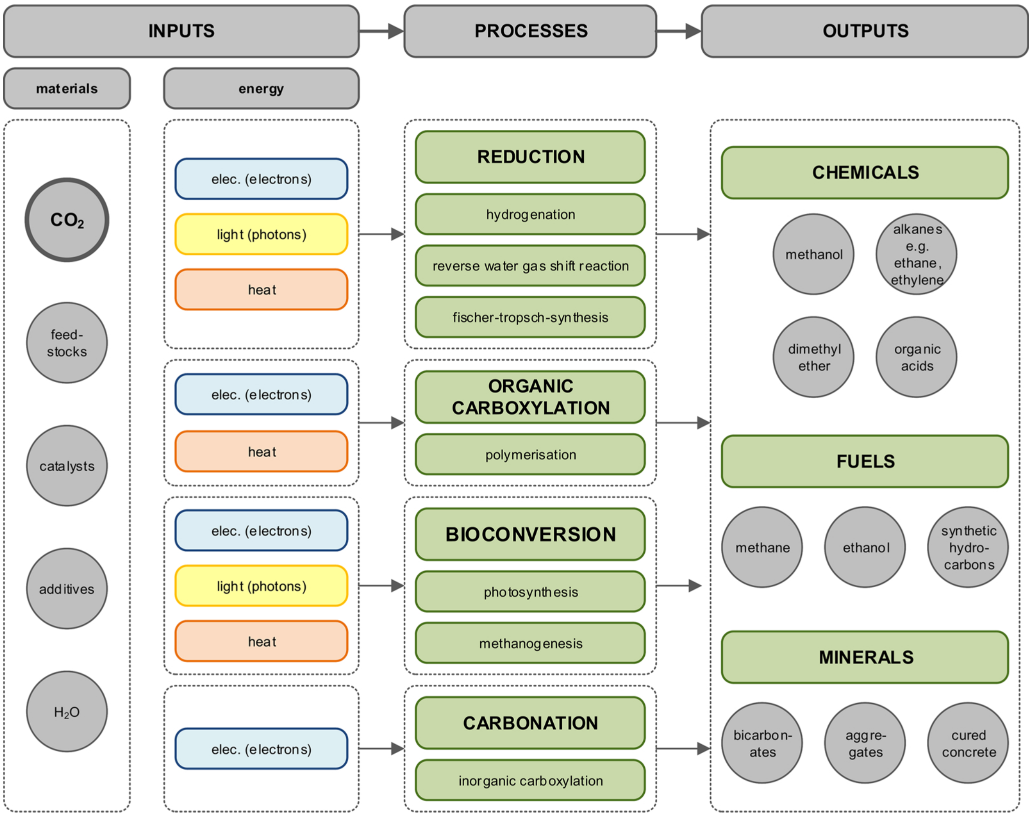

Fig. 1. Classification of potential Carbon Capture and Utilisation pathways.

stoichiometric CO_2-uptake approach, excluding methane, which could account for 3000 to 4000 Mt/a. Patricio et al. [12] presented the current data for the potential CO_2 demand in Europe, which added up to 73 Mt/a for the industrial processes. Accordingly, in order to address the predicted increase in CO_2 demand, a comparative evaluation of the sources and limitations of biogenic CO_2 sources is needed. A comprehensive review on the existing and emerging uses of CO_2 and their demand was published in [13,6,3].

Today, this demand is partially satisfied by CO_2 extracted from natural wells, which is considered to contradict the logical approach to (European) climate goals [14]. According to Naims' CO_2 supply and demand analysis [15], in the USA, approximately 45 Mt/a were extracted from natural wells in 2012 for economic purposes. In addition to the future reduction of fossil CO_2 sources, it is argued that CCU could be used as a complementary technology in mitigation technologies, with a focus on local circular economic approaches. Naims [15] as well as other authors [11,16,17] focus mainly on fossil CO_2 sources presumably because fossil CO_2 adds to the CO_2 content of the atmosphere to some extent, whereas biogenic CO_2 is considered as “carbon neutral”. Aresta et al. [18,19] state that CO_2 recycling technologies, such as renewable fuel production, could become economically and environmentally feasible, with the support of renewable energies. It has been proposed that “spent carbon” emissions should be converted to “working carbon” emissions in order to reduce the total fossil carbon input in our economy and environment. The authors of the present paper are convinced that biogenic CCU (bio – CCU) complements this approach.

To ensure the climate neutrality of the versatile CCU products (e.g. as shown in Fig. 1), the usage of biogenic CO_2 is preferred. Hence, the authors mainly focus on the use of already available biogenic CO_2 from industrial sources, as it has been defined as carbon neutral to the environment [20]. However, *biogenic* carbon capture is mentioned comparatively seldom in literature, although, CCU and Carbon Capture and Storage (CCS) have received increasing scientific attention since the 1990s, as can be derived from a key word search, compared in Table 1.

Table 1
Overview of publication history at ScienceDirect.com for different search items.

Search items	New items in year	
	1996	2019
Carbon Capture	2800	> 31000
Biogenic Carbon Capture	98	942
Bio-CCS	34	480
Bio-CCU	41	140

Within the subject, the possible methods to capture CO_2 from different sources is the most comprehensively discussed topic in scientific publications, followed by the storage and utilization technologies, fossil CO_2 emitting sources, such as the transport sector and the chemical industry and their CO_2 potential. The lower frequency of studies on bio – CCS/bio – CCU can possibly be attributed to the concentration of biogenic CO_2 sources in just a few countries, such as a considerable number of biogas plants in Germany [21]. Another reason for the relatively low research interest in biogenic CCS could be the comparatively simple, far-advanced and relatively low-cost capture and purification process of biogas. Most papers deal with case studies or specific Carbon Capture, Carbon Storage or Carbon Utilization pathways. Moreover, these studies often include new processes, which allow the use of CO_2 as a feedstock material or investigate methods of efficiently capturing CO_2 from fossil sources.

However, the available amount of CO_2 from fossil sources or from primarily biogenic sources at the European or global scale is seldom a core topic [22–24]. More often, the current and future CO_2 demand is discussed [10–12,25,26]. Comparisons of future CO_2 potential (theoretical, technical and economical) from biogenic and fossil sources, combined with the potential CO_2 demand, is out of the scope of most publications. Thus, there is a lack of studies on the holistic potential and sources of CO_2 at the European and global scale.

Table 2
Range of past/current European and global biogenic CO_2 potentials according to literature references.

CO_2 source		CO_2 amount in Mt/year	region	year	Ref.
bioenergy based	bioenergy	73	North America, Brazil	2000, 2003 ref. year	[28]
	bioenergy	73[a]	North America, Brazil	2016	[15]
	biomass + renewable waste combustion	287	Europe	2014	[22]
	municipal non-renewable + industrial waste	81	Europe	2014 ref. year	[22]
	municipal solid waste incineration	2,800	Global	2100 (Forecast)	[29]
fermentation based	fermentation processes	17.6	North America, Brazil	2000, 2003 ref. year	[28]
	fermentation processes	17.6[a]	North America, Brazil	2016	[15]
	fermentation processes	4.4	Europe	2014	[22]
	ethanol production	0.1	Austria	2013 ref. year	[30]
	fermentation processes[e]	0.039	Finland	2017	[31]
	wine production	0.393[d]	Italy	2015 ref. year	[32]
	wine production	2.252[d]	Global	2015 ref. year	[32]
biogas (upgrading)	biogas (CHP) + biogas upgrading	11.95 - 18	Germany	2016	[10,33]
	biogas (CHP) + biogas upgrading	8 - 11.3[b]	Germany	2050 (Forecast)	[10,33]
	biogas upgrading	0.013	Austria	2013	[30]
	biogas production	23	Europe	2014 ref. year	[22]
	landfill gas combustion[c]	1,160	Global	2100 (Forecast)	[29]
Direct Air Capture (DAC)	DAC	500	Europe	Future Scenario	[34]
	DAC	7,000 – 2,000	Global	Future Scenario	[34,35]

[a] Naims refers to IPCC.
[b] Assumption: reduced use of energy crops, less number of biogas plants [36].
[c] alternative to waste incineration, not parallel.
[d] effective CO_2 reduction.
[e] aggregated from traffic ethanol production, beverage production and bakeries.

In this study, we focus on the current potentials to capture and utilize biogenic CO_2. Since the global economy can only become carbon neutral if fossil energy- and fossil resource-based chemical industries adapt to new, renewable energy driven bio − CCU processes, the utilization of CO_2 from green sources, such as biogas upgrading or bioethanol fermentation, is a strategic approach for CO_2 mitigation strategies. The listing of estimates and predictions in Table 2 for the (mainly theoretical) potential of CO_2 capture shows that available data is mostly inconsistent and thus hardly comparable. Future economic and technical potentials are difficult to define, since they strongly depend on further economic and technological developments, such as trends towards decentralized or centralized energy supply systems of industries and municipalities and changing processes in the chemical industry. For completion and their potential value in a carbon cycle economy, fossil CO_2 emissions are discussed and compared to the renewable sources based on the available quantities and qualities of CO_2. Processes involving direct CO_2 capture from air are not the focus of this study, as the required processes are relatively energy and cost intensive at the current development stage [27].

The structure of the paper is as follows: Section 2 gives an overview of stationary CO_2 point sources with a focus on biogenic / green CO_2 in Subsection 2.1. In the following Section 3 Results and Discussion, the Subsections 3.1 to 3.3 present detailed information on European (and global) biogenic CO_2 potentials. Subsection 3.4 focuses on fossil sources. Subsections 3.5 and 3.6 discuss the occurrence of CO_2 point sources as well as limiting factors such as technological capture rates and capture costs. Finally, Section 4 contains the conclusions.

2. Overview of CO_2 sources

2.1. Potential sources of biogenic CO_2

Different sources of biogenic CO_2 are classified and highlighted in Fig. 2. The main biogenic CO_2 sources are combustion of biomass, biogas upgrading to biomethane and industrial fermentation, e.g., brewing and other fermentation processes in the food and beverage (FAB) industry. Although industrial bioethanol is also produced through fermentation [37], it has been treated separately. In contrast to the FAB industry, bioethanol is mostly used as biofuel and as raw material in several industries. Direct capture of CO_2 from the atmosphere has not been considered as a source from existing industrial plants but, rather, as a diffuse source, which would demand a significant technical effort to be separated. Thus, for subsequent chemical conversion herein, ambient air is not considered as a currently affordable source of biogenic CO_2 and not further discussed in this assessment. Nevertheless, CO_2 separation from ambient air may still play an important role in the sequestration of CO_2 from diluted and dispersed sources in the long-term, as the relevant technology has the potential for significant further development and optimization [38].

2.1.1. CO_2 from solid biofuels

Solid biofuels are defined here according to Eurostat:

"Solid biofuels covers solid organic, non-fossil material of biological origin (also known as biomass) which may be used as fuel for heat production or electricity generation. In energy statistics, solid biofuels is a product aggregate equal to the sum of charcoal, fuelwood, wood residues and by-products, black liquor, bagasse, animal waste, other vegetal materials and residuals and renewable fraction of industrial waste." [39].

As can be seen in Fig. 2, flue gases from biomass combustion processes consist of only 3–8 vol.-% CO_2. The flue gases include many other components that make the utilization of CO_2 in a pure form a technically challenging task. Because of this required technical effort, higher financial investments are expected, resulting in a lower economic feasibility of CO_2 utilization from this source in comparison to CCU from industrial bioethanol and biogas production.

An overview of existing and actual initiatives for the development of new CO_2 separation technologies for combustion processes can be found on the CCS browser [40] of the CO_2 Capture Project (CCP) [41]. Further information on CO_2 capture technologies are presented by Cuéllar-Franca and Azapagic [16] and in the IPCC special report on CCS [37].

2.1.2. CO_2 from liquid fermentation processes

2.1.2.1. Liquid biofuels – fermentation of industrial bioethanol. Liquid biofuels are defined here according to Eurostat:

"Liquid biofuels includes all liquid fuels of natural origin (e.g. produced from biomass and/or the biodegradable fraction of waste), suitable to be blended with or replace liquid fuels from fossil origin. […]" [39].

There are several liquid biofuels like biodiesel, biogasoline, bio jet kerosene and bioethanol existing. The latter is produced by

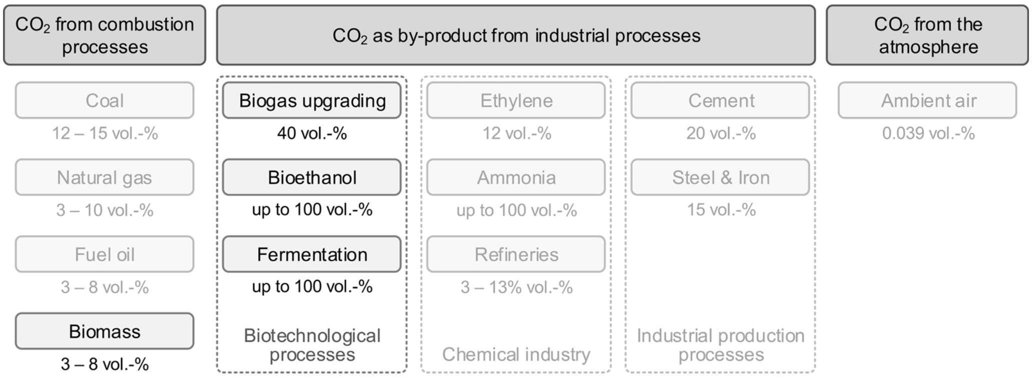

Fig. 2. Classification of potential biogenic CO_2 sources including the available typical CO_2 concentration.
Source: based on [30].

fermentation processes, besides other options, generating a considerable amount of CO_2 as by-product. Concurrent to the stoichiometric equation, the gas produced during the fermentation consists of up to 99–100 % CO_2 [22]. The basic equation of ethanol fermentation is:

$C_6H_{12}O_6 \rightarrow 2\ C_2H_5OH + 2\ CO_2$.

CO_2 from bioethanol production is pure enough to be directly utilized in the FAB industry, e.g., as carbon acid in beverages [42], though quality requirements for CO_2 utilized in the FAB and pharmaceutical industries are very high [3]. Regulations for the quality of food grade CO_2 are released by the European Industrial Gases Association (EIGA) [43] and the International Society of Beverage Technologists (ISBT) [44] (Table 3). Since the aforementioned regulations are strict, hence, this CO_2 source can also be expected to fulfil the fundamental requirements for CCU applications. However, for some impurities such as water, O_2, hydrocarbons, and CO, the limitations are not as demanding as those for CO_2 gases for the chemical industry according to EN ISO 14175: C1. As an example of the purity requirements for CO_2, the product data sheets from the Linde Group could be potentially considered: BIOGON® C flüssig E290 - Kohlendioxid 3.0 [45] for food grade CO_2 and Kohlendioxid 4.5 [46] for chemical industry CO_2. Depending on the requirements for the CCU pathway, further purification of food grade CO_2 or CO_2 for chemical industry may be needed, for example, by activated carbon technologies [47–50].

Table 3
Limiting characteristics for CO_2 to be used in beverages. Source: European Industrial Gases Association (EIGA) (cf. [43]).

Component	Concentration
Assay	99.9% v/v min.
Moisture	20 ppm v/v max.
Ammonia	2.5 ppm v/v max.
Oxygen	30 ppm v/v max.
Oxides of Nitrogen (NO/NO_2)	2.5 ppm v/v max. each
Non-volatile residue (particulates)	10 ppm w/w max.
Non-volatile organic residue (oil and grease)	5 ppm w/w max.
Phosphine (only for CO_2 from phosphate rock sources)	0.3 ppm v/v max.
Total volatile hydrocarbons (calculated as methane)	50 ppm v/v max. of which 20 ppm v/v max. non-methane hydrocarbons
Acetaldehyde	0.2 ppm v/v max.
Aromatic hydrocarbon	0.02
Carbon monoxide	10 ppm v/v max.
Methanol	10 ppm v/v max.
Hydrogen cyanide (only for CO_2 from coal gasification sources)	0.5 ppm v/v max.
Total sulphur (as S for total content < 0.1 ppm v/v)[a]	0.1 ppm v/v max.
Taste and odour in water	No foreign taste or odour
Appearance in water	No colour or turbidity
Odour and appearance of solid CO_2 (snow)	No foreign odour or appearance

[a] If total sulphur content > 1 ppm v/v, then: Carbonyl Sulphide 0.1 ppm v/v max., Hydrogen Sulphide 0.1 ppm v/v max., Sulphur Dioxide 1.0 ppm v/v max.

Notably, the FAB industry offers possible competing utilization pathways for CO_2, especially from industrial bioethanol production. According to ePURE, bioethanol producers in Europe commercialized 0.4 Mt of CO_2 utilization in 2016 [51]. The estimations within this paper are based upon the information available on the producers' websites [52] and on some approximations from ethanol outputs, indicating an amount of 2.03 Mt CO_2/year, which may possibly be commercialized by the European bioethanol producers in the mid-term future. The CO_2 is often utilized as dry ice [53,54], gaseous fertilizer and food packing agent [3,10,55,56] or for carbonating beverages [3,10,42,53].

2.1.2.2. Other industrial fermentation processes. In addition to CO_2 derived from bioethanol industry, the CO_2 from fermentation processes in the FAB industry, such as brewing processes, is of interest. In the beverage industry, beer brewing and wine production lead to considerable amounts of CO_2. Furthermore, the fermentation of acids, e.g., citric acid, produces considerable amounts of CO_2.

According to [57] 41.1 billion litres of beer were brewed in Europe in 2016. Using an average value of 5 vol.-% of alcohol and 5 g/l carbon acid for beer, it can be estimated that 35 g CO_2/l were released during this fermentation process [58–60].

A comparable estimation could be conducted for the CO_2 potential from fermentation in European wine production. The average value of the European wine production is approximately 17 billion litres of wine annually [61]. The average alcohol content is 11 vol.-% while the carbon acid content is quite low, approximately 1 g/l, which corresponds to approximately 87 g CO_2/l released during the production process [62,63]. Marchi et al. [32] estimate 84.5 g CO_2/l_{must} to be released by wine production, which corresponds to the estimations in this paper. Furthermore, the carbon acid amount depends on the type of wine. Red wine in particular has a very low content of carbon acid and is decarbonated very often, whereas the carbonation of white wines and sparkling wines is quite common. Similar to beer brewers, wine producers protect their wine from air using CO_2; in practice most of this CO_2 is not available for chemical syntheses [32,64–66]. The small-scaled structure of producers in this sector is another barrier to the implementation of CCU, for economic reasons [32].

The CO_2 accumulating in the beverage industry is reused nearly completely, especially in larger breweries and wine production sites, e.g. for carbonating wine and beer directly [67–69] or as inert gas to preserve the beverages [3,64,66,70].

2.1.3. Gaseous biofuels

Gaseous biofuels are defined here according to Eurostat:

"Biogas is a gas composed principally of methane and carbon dioxide

Table 4
Selection of possible substrates for biogas plants. Source: based on [73,74].

Category	Possible feedstocks	
	Feedstock type / source	Typical examples
Agricultural	energy crops	maize silage
		sugar beet silage
		silage from different grains
		crop residues in general
	manure	pig
		cow
		sheep
		poultry
FAB industry	liquor industry	grain stillage
		potato stillage
	sugar and starch industry	sugar beet residues
	beverage industry	fruit pomace
	food industry	production residues
		slaughter waste and blood
		dairy residues
Waste industry	different wastes	municipal renewable waste
		industrial renewable waste
		sewage sludge
Textile industry	production residues	leather
		fur
		biological textiles
Wood industry	panels and furniture	wood residues
	paper industry	paper and cardboard residues
		pulp residues

produced by anaerobic digestion of biomass or by thermal processes from biomass, including biomass in waste. In energy statistics, biogas is a product aggregate equal to the sum of landfill gas, sewage sludge gas, other biogases from anaerobic digestion and biogases from thermal processes." [39]

In principle, there are two ways to utilize CO_2 from biogas, independently of the source of the biogas. First, as biogas consists of approximately 60 % methane and 40 % CO_2 ([10,22,71] based on [72]), upgrading biogas to biomethane offers a large potential for the generation of biogenic CO_2. Second, the combustion of biogas and biomethane generates CO_2 as a compound of the flue gas during the generation of heat and power. Both paths are considered separately in the results.

2.1.3.1. *Biogas substrates*. Biogas from anaerobic digestion is derived from biogas plants with highly different biomass feedstock. Table 4 provides an overview of the possible compounds of biogas substrates. Most plants are supplied with varying mixtures of substrates. Some substrates demand special treatment and plant design. Depending on the substrates, the composition of the biogas varies, with regard to the methane concentrations, CO_2 concentrations and trace compounds. Since the ideal CO_2 stream needed for the various CCU synthesis options is highly pure, some biogas substrates may be not suitable for this application without extensive purification processes of the potentially utilized CO_2.

According to the Statistical Report of European Biogas Association (EBA) 2017 [75] the feedstock use for biogas production differs for every country. Using the substrate's mass percentage as an indicator for biogas production (excluding landfill gas), energy crops are the main substrates in Latvia, Austria and Germany, while in Greece, Cyprus, France, Serbia, Poland and Italy agricultural residues are the main feedstock. In the UK, Finland, Sweden, Spain, Denmark and especially in Switzerland [75], sewage accounts for the largest share. In some countries such as Belgium, Croatia and Hungary the distribution is more even.

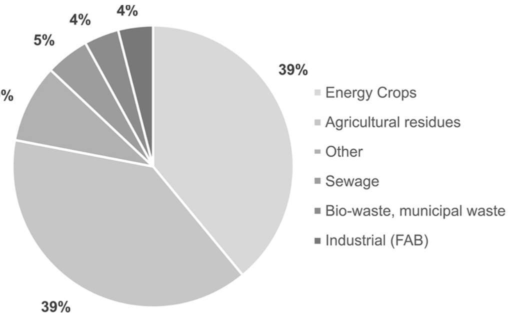

Fig. 3. Estimation of relative significance of each feedstock in the biogas industry in Europe in 2016.
Source: based on feedstock mass percentages and electricity production per feedstock in each country. The share of sewage sludge is underestimated due to missing information on the share of sewage sludge for some countries. Adapted from Figure 9-EU from Statistical Report 2017, EBA [75].

Considering landfill gas, the statistics shift. Approximately one third of Estonia's feedstock origin is landfill waste [75]. In Greece, landfill gas accounts for two thirds of the produced biogas [75], and for Norway landfill gas accounts for nearly half [75]. Portugal is exceptional in using landfill gas; landfill gas accounted for over 95% of produced biogas in 2016 [75]. Other countries making significant use of landfill gas are the UK, Sweden, Romania, Poland, Ireland, France and Finland.

Fig. 3 is compiled based on data from the EBA [75]; it shows the share of feedstock use for biogas according to the substrate's mass percentages and electricity production per substrate in each country for Europe. Agricultural waste and energy crops represent the highest share, with 39% each, followed by "other", which includes organic waste from households and industry, sewage, FAB and bio-waste/municipal waste. According to the EBA, the share of sewage is underestimated.

The main substrates by mass percentage, excluding landfill gas for current biomethane plants, are slightly different than the main substrates for biogases in general. In particular biowaste and municipal waste, agricultural residues and, to some extent, unknown feedstocks are of major relevance. In Germany most biomethane plants are based on energy crops, followed by agricultural residues and bio-/municipal waste, while in the UK "other" (municipal waste, etc.) and agricultural substrates play the biggest role. These two countries are the ones with the most biomethane plants (see Fig. 7). For Sweden, which ranks third among European countries with the most biomethane plants, "other" and sewage are the feedstocks which account for the most biomethane plants [75].

2.1.3.2. *Composition of biogas*. Depending on the substrates, the plant layout, operating temperature and various other parameters, different trace compounds can be found in biogas resulting from biological processes in anaerobic digestion. Typical impurities are water vapour, O_2, N_2, NH_3, H_2, H_2S, siloxanes and biogas specific volatile organic carbons [73,76]. Table 5 provides a more detailed overview of possible impurities and their typical concentrations in biogas.

Rasi et al. [78,79] provide a more detailed overview of biogas components, depending on the utilized substrates for biogas production. Landfill gas, in particular, often contains high amounts of H_2S and Volatile Organic Compounds (VOCs). Additionally, e.g., in industrial wastes, several potentially hazardous trace compounds can be part of the landfill gas, such as fluorinated and chlorinated hydrocarbons, aromatic compounds and higher hydrocarbons [80]. Other common trace compounds are siloxanes, which are also present in sewage sludge gas, since siloxanes originate from ingredients such as cosmetics, soaps and detergents. Depending on the substrate, agricultural biogas plants can also produce biogas with a very high H_2S content, for example with that derived from manure feedstock [79].

2.2. *Potential sources of fossil CO_2*

Although this study targets biogenic, and thus "green", sources of

Table 5
Detailed overview of biogas components. Source: adapted from [77].

Components	Concentration range
Main components	
Methane (CH_4)	50 - 70 mol.-%
Carbon dioxide (CO_2)	30 – 50 mol.-%
Nitrogen gas (N_2)	0 – 3 mol.-%
Oxygen (O_2)	0.0 – 0.5 mol.-%
Hydrogen (H_2)	0.0 – 1.5 mol.-%
Water vapor (H_2O)	1 – 7 mol.-%
Carbon monoxide (CO)	0 – 1 mol.-%
Trace components	
Ammonia (NH_3)	0 – 308 ppm(mol)
Hydrogen sulphide (H_2S)	20 – 850 ppm(mol)
Terpenes	0 – 500 ppm(mol)
Benzene, Toluene, Xylene (BTX)	0 – 7 ppm(mol)
Hydrocyanic acid (HCN)	0 – 0.003 ppm(mol)
Fluorine compounds (R-F, incl. HF)	0 – 1.3 ppm(mol)
Chlorine compounds (R-Cl, incl. HCl)	0.1 – 5 ppm(mol)
Siloxanes (D4 & D5)	0 – 3.4 ppm(mol)

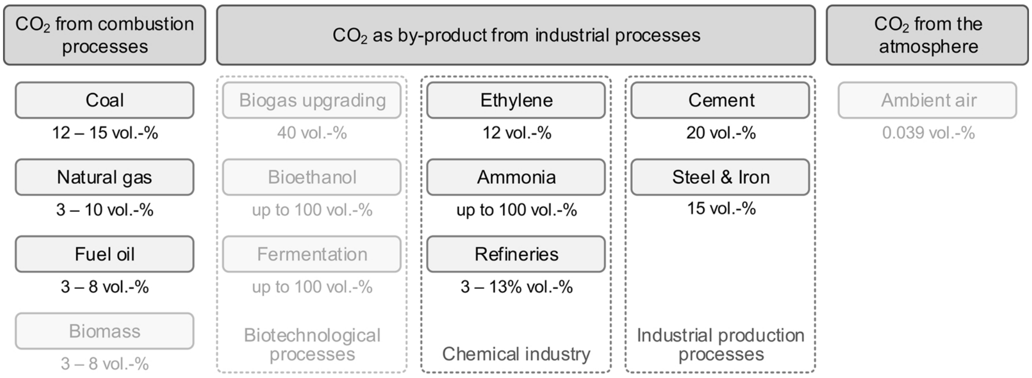

Fig. 4. Classification of potential fossil CO_2 sources including the available typical CO_2 concentration.
Source: based on data from [30].

CO_2 as feedstock material, the main emitters of CO_2 are based on fossil fuels. Because of the increasing interest in the utilization of CO_2 to attain ambitious goals of decarbonisation and closed carbon cycles, these fossil sources must be considered as relevant input sources to some extent as well. Therefore, this section gives a rough overview of potentially available carbon sources and their relevance to CCU process chains.

The potential sources for fossil CO_2 can be classified according to their emitting processes and industry sectors. This classification is shown in Fig. 4.

For the European Union (EU), industrial CO_2 emissions are mainly registered in the EU emission trading system (EU ETS)[1] . The system records CO_2 emissions from power and heat generation as well as from energy-intensive industry sectors, including oil refineries, steel works and production of iron, aluminium, metals, cement lime, glass, ceramics, pulp, paper, cardboard, acids and bulk organic chemicals [81]. Beyond these, commercial aviation is also included in the register; however, owing to the limited capability for the direct capture and separation of CO_2 emissions, these are excluded in the following investigations.

For the analysis of fossil CO_2 potentials in this study, the report data released by the European Commission in April 2018 [82] was used as a primary data source. Since data for 2017 was incomplete at the time of this report, year 2016 data was used as a reference for the subsequent calculations.

The EU ETS database for verified emissions allows for the categorization of registered emissions according to their originating industrial sector. Thus, the available data can be grouped into the following main categories:

- power & heat from fossil fuels; energy industry
- chemical industry
- iron & steel
- (other) metals processing
- cement, clinker, lime, ceramics
- production of glass & glass fibre
- pulp, paper & board
- other installations
- aircraft operator activities

In addition to these main categories, the chemical industry sector has been further subdivided into refinery, ammonia production and other chemicals. Within the category of cement, clinker, lime and ceramics, the manufacturing of ceramics by firing has been separated from the production of cement clinker and lime.

Adding additional detail categories in the other sectors according to the activity type codes provided in the data source is also possible; however, other than providing a clearer classification for large parts of the register, further categorization of the other sectors does not add any beneficial value to the investigations executed in the context of this potential analysis.

3. Results and discussion

3.1. CO_2 potential from solid biofuels

Solid biofuels are predominantly utilized by combustion but some amounts also statistically contribute to biogas production. However, the combusted amount is responsible for the highest biogenic CO_2 emissions in Europe, which account for approximately 437 Mt CO_2/year according to Eurostat statistics of 2016 and IPCC 2006 emission factors for stationary combustion [28,83]. Solid biofuel are not only combusted at large plants but also at various small-scale facilities such as household fireplaces and central heating systems of buildings. Hence, it can be concluded, that if the global statistics for solid biofuels and the fraction of direct heat use is considered, the amount of CO_2, which could be utilized is therefore significantly lower than the theoretical potential of 437 Mt CO_2/year.

According to the Global Bioenergy Statistics 2017 of the World Biogas Association (WBA) [84], municipal waste, industrial waste and primarily solid biomass account for 54.72 EJ/year (for 2014)

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[1] The EU ETS operates in 31 countries. This includes the 28 EU countries plus Iceland, Liechtenstein and Norway. These 31 countries are indicated as EU in the following context, if not stated otherwise.

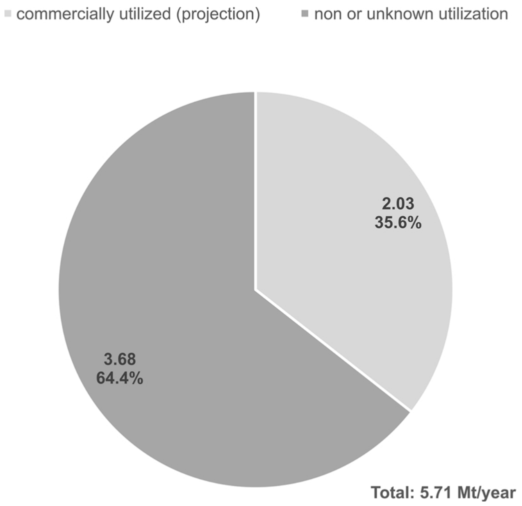

Fig. 5. Amount of CO_2 in Mt/year produced in the EU bioethanol industry in 2016 including amount of possibly commercialized CO_2.
Source: Own projections based on company data and data from [51,52].

worldwide. Municipal waste and industrial waste include non-renewable fractions. 42.88 EJ of this solid biomass energy are converted to direct heat, meaning direct consumption of energy sources in the residential, agriculture and commercial sectors (not combined heat and power (CHP), heat or electricity-only plants) [84]. Therefore, only CO_2 derived from 11.84 EJ[2] could be utilized for CCU, but only 6.86 EJ[3] of solid biomass are utilized in CHP, electricity-only and heat-only plants. The respective CO_2 amounts are 1184 Mt and 686 Mt per year[4] .

3.2. CO_2 from liquid fermentation processes

3.2.1. CO_2 potential from bioethanol industry

The total amount of CO_2 produced during bioethanol fermentation approximates to 5.71 Mt CO_2/year based on production statistics from 2016 [51]. Ericsson estimates 4.4 Mt CO_2/year for Europe [22]. Most European bioethanol fermentation plants are based in France (17 plants) followed by Germany (8 plants) and the UK (5 plants). Other European countries have [51] three plants or less. In total, there are approximately 56 plants in Europe, of which 43 are located in the EU [based on 51,52,83]. The total installed production capacity for bioethanol in Europe is approximately 9.2 billion litres a year (of which 6.3 billion litres a year account to ePURE Members, whereas total production of ePure Members accounted for 5.2 billion litres in the year 2016, i.e. 82.5 % of capacity) [51]. It is assumed that the European share of real production is similar, i.e. 7.57 billion litres ethanol in 2016.

The approximated theoretical amount of CO_2 produced in the European bioethanol industry in 2016 (based on 7.57 billion litres ethanol) is summarized in Fig. 5.

According to ePURE [51], the FAB industry as one group and other industries as another group each represent 11% of European industrial bioethanol consumption. Fuel accounts for the remaining 78% of consumption. The possibilities for yielding CO_2 “end-of-pipe” from the ethanol utilization pathway in the FAB and other industries is complex and thus relatively unattractive. Similarly, while gaining CO_2 from ethanol flue gases would theoretically be possible, it is technically and economically not currently feasible, as bioethanol is usually used as a component of vehicle fuel.

However, the global bioethanol production is much higher; it accounted for 90.5 billion litres of bioethanol produced in 2014 [85]. According to these figures, the European fraction in 2014 was only 7 %. Industrial bioethanol from the US accounted for the highest amount, with 60 % of global production, while Brazil produces around 30 %. Due to the high purity of CO_2 from bioethanol and hence simple capturing, establishing CCU production in North and South America, where large bioethanol production sites can be found, is an option for further consideration.

3.2.2. CO_2 potential from other industrial fermentation processes

Based on the average alcohol content, approximately 1.44 million tons of CO_2 are released from annual European beer production. The CO_2 amounts which can be utilized are smaller, since approximately 5845 out of 8130 European breweries in 2015 were so called microbreweries, with an annual beer output of, at maximum capacity, 1000 hl [57], which corresponds to approximately 3.5 tons of CO_2/year on average per microbrewery. Therefore, the technical effort needed to gather large amounts of CO_2 is quite high. Furthermore, some breweries already utilize their own CO_2 as protective gas for filling [67,69,86,87]. Therefore, most of the CO_2 from this fermentation process is not applicable for other CCU processes.

For the EU wine industry, the annual CO_2 amount estimated from the average values presented in Section 2.1.2.2 is 1.48 million tons CO_2. Marchi et al. [32] estimate an effective annual CO_2 potential of 1.065 Mt/a for Spain, Italy and France, who are the main wine producers in Europe.

3.3. CO_2 potential from biogas

3.3.1. CO_2 from biogas upgrading

At the end of the year 2017 17,783 biogas plants were operational in Europe [88] of which 497 had upgrading facilities installed in early 2017 [21,88]. At the end of 2017, approximately 540 biomethane plants were operational in Europe [88]. Fig. 6 gives an overview of the number of biogas plants per country in Europe at the end of 2017. Since then the distribution has changed only marginally.

In 2018, EBA and Gas Infrastructure Europe (GIE), in collaboration with several partners, published the European Biomethane Map 2018 [21], which includes detailed data from all known European biogas upgrading plants. According to the map, Germany, the UK and Sweden are the pioneers of the field, in terms of the number of upgrading plants, as can be seen in Fig. 7. By comparing Figs. 6 and 7 it can be seen that there is still a significant potential for biogas upgrading. This indicates a significant potential for CCU from biogas.

Since not only the number of biogas and biomethane plants, but also the specific CO_2 potential of each are of interest, annual CO_2 amounts derived from biogas and biomethane production in Europe are presented here. The numbers include the CO_2 potential from the raw biogas (~ 40 vol.-% CO_2) and upgraded biomethane (~ 99 vol.-% CO_2) and exclude the CO_2 emitted during the combustion of biogas in CHP facilities or utilization of the biomethane. Figs. 8 and 10 show the approximated cumulative CO_2 potential in 2016 in the EU for biogas and biomethane plants. Fig. 9 shows the CO_2 potential from biogas production per country for EU-28. Germany is leading in biogas production except for landfill gas, for which the UK accounts for the largest current amount. Biogas from thermal processes accounts for a very small amount of CO_2 and is mostly derived from Finland.

As illustrated in Fig. 10 the approximated CO_2 potential from biomethane upgrading represents only 14 % of the cumulated CO_2 from biogas production, still excluding the CO_2 emitted during utilisation (combustion of biogas in a CHP facility or utilization of upgraded biomethane in different applications, e.g., heat, electricity or transport).

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[2] Difference of 54.72 EJ and 42.88 EJ

[3] Summed up the energy utilized 2014 in CHP (2.4 EJ), heat only plants (0.48 EJ) and electricity only plants (3.98 EJ), Source: [84].

[4] Conversion from energy to CO_2 with IPCC emission factors [28] Approx. 100,000 kg CO_2/TJ

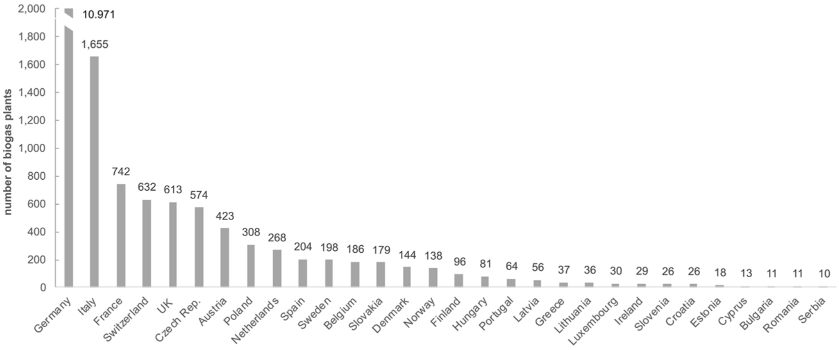

Fig. 6. Biogas plant distribution in Europe at the end of 2017. Note: Data underlie constant change and are only an orientation guide for the development of European biogas economy.
Source: based on Figure EU-2 from Statistical Report 2018, EBA [88].

According to the data from the WBA [84] in 2014, approximately 1.27 EJ of biogas were produced globally. This accounts for a potential of approximately 42.3 Mt CO_2 from biogas production, assuming a CO_2 content of 40 % and a biogas heating value of 21.6 MJ/Nm^3 or 6 kW h/Nm^3, respectively. About 50 % of global biogas is produced in Europe, whereas Asia accounts for one third and America for roughly 17 %.

3.3.2. *CO_2 from combustion of biogas and biomethane*

During the combustion of biogas and biomethane, other than some trace compounds, mainly CO_2 and water are produced. Since exact fractions depend on the composition of the fuel gas and the air supply during combustion, CO_2 amounts in the flue gas can only be approximated.

3.3.2.1. Estimations based on stoichiometry. In 2016, approximately 695 PJ of biogas were produced in the EU according to Eurostat [83]. Considering data from the EBA [88], 62 PJ biomethane were produced in 2016. This is equal to 8.9 % of total biogas production. Therefore, 633 PJ, or approximately 26.5 billion m^3, of biogas[5] are utilized in CHP and other plants, whereas the remaining amount is upgraded. This is equal to 52.5 Mt of CO_2 per year[6], considering a density of 1.98 kg/Nm^3 for CO_2 and including 40 vol.-% CO_2 content of the biogas.

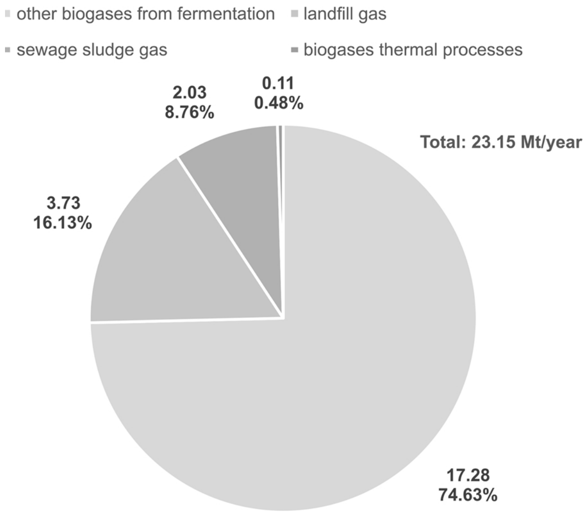

Fig. 8. Cumulative CO_2 potential in Mt/year from biogas production for the EU-28 countries.
Source: based on Eurostat data on EU biogas production in 2016 [83].

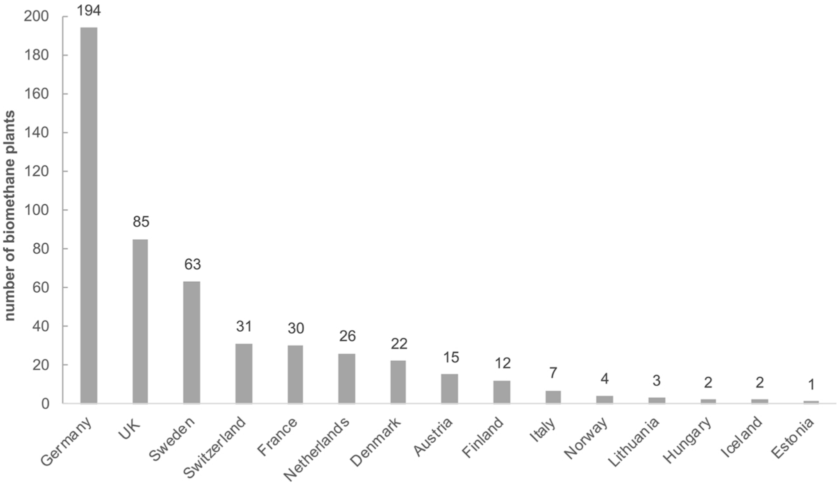

Fig. 7. European country ranking according to the number of biogas upgrading plants in early 2017.
Source: based on a diagram from European Biomethane Map 2018 [21].

3.3.2.2. Estimations based on emission factors. Emission factors can also be used in order to estimate specific CO_2 emissions. Considering a CO_2 fraction of 40 vol.-% of the biogas and the CO_2 from combustion, an emission factor of 91.5 g CO_2/MJ_{LHV} can be determined. This leads to an emission of 57.9 Mt/a.

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[5] Converted with higher heating value of 6.64 kWh/m^3

[6] Stoichiometric calculation with pure oxygen as reagent. Source: Own calculation.

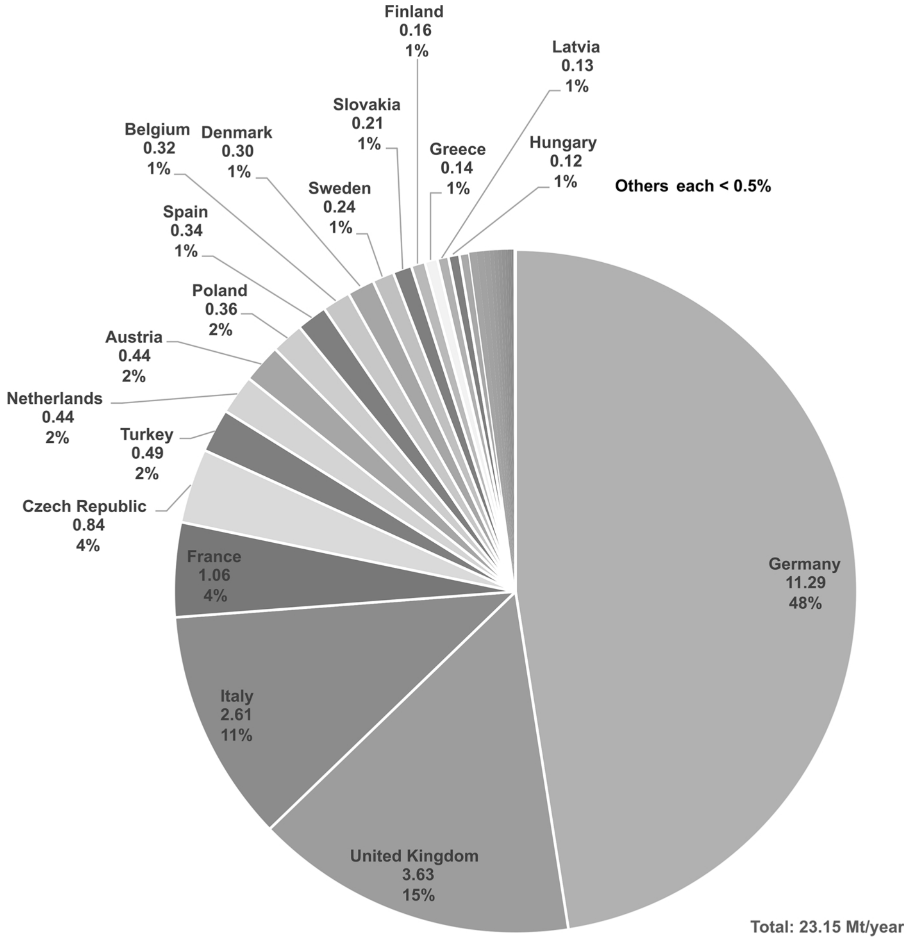

Fig. 9. CO_2 potential from biogas production per EU-28 Member State.
Source: based on Eurostat data on European biogas production in 2016 [83].

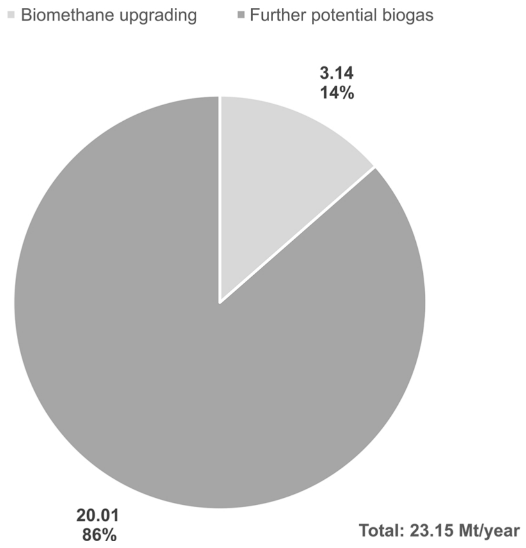

Fig. 10. CO_2 potential in Mt/year in 2016 from biomethane upgrading as part of total CO_2 potential from biogas production for the EU-28.
Source: based on data from [21,83].

Basing the calculations on the emission factor from IPCC 2006 [28], which is equal to 54.6 g CO_2/MJ_{LHV} and is based on the calorific value, 34.6 Mt CO_2/year are released. The 34.6 Mt CO_2/year do not include the 40 vol.-% CO_2 fraction from biogas production, but only the CO_2 from combustion of biogas (approx. 60 vol.-% CH_4). Summing up the latter and the CO_2 amount from theoretical biogas upgrading, which corresponds to the 40 % CO_2 fraction, it can be calculated, with the comparable order of magnitude, that approximately 55.6 Mt CO_2/year are emitted. This is nearly the same as using 91.5 g CO_2/MJ_{LHV} as the emission factor, and roughly higher than the stoichiometric result.

3.3.2.3. Limitations in CCU potentials. In most cases, the upgraded biogas is injected into the gas grid. Therefore, in practice, the CO_2 derived from the combustion of biomethane is not directly available for CCU, unlike the CO_2 separated during the upgrading process. In 2017, 73 of 497 biomethane upgrading plants in Europe were not connected to the gas grid [21]. At these plants, the biomethane is directly utilized, which means that in some cases the CO_2 from combustion could be potentially harnessed for utilisation, except for places where the biomethane is used as biofuel for motor vehicles. In the latest Statistical Report from the EBA [75] the several incentives are presented to increase the use of upgraded biogas as a fuel. Especially in Sweden (88 %) and Finland (25 %) large amounts of the national production are already used as fuel for vehicles. Other countries, e.g., Estonia, Norway and Italy, are planning to strengthen this utilization pathway in the upcoming years.

Considering the global biogas production of 1.27 EJ for 2014, approximately 116 Mt CO_2/year are released by biogas combustion, using an emission factor of 91.5 g CO_2/MJ_{LHV}. Basing calculations on the emission factor from IPCC 2006 [28], 69.3 Mt CO_2/year are released from the combustion of the methane content. Adding the basic CO_2 content of 40 % of the biogas, approximately 112 Mt CO_2/year are emitted.

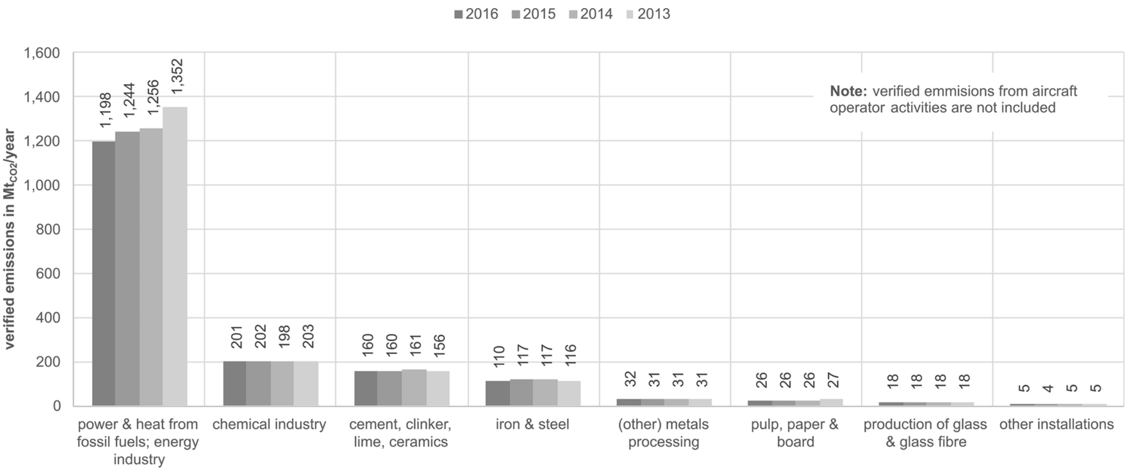

Fig. 11. Total verified CO_2 emissions in the EU per industry sector.
Source: based on data from [82].

3.4. CO_2 potential from fossil sources

The verified emissions registered in the EU ETS are allocated to their respective categories and their development in recent years compared, as illustrated in Fig. 11. This figure shows that, on the one hand, the energy industry, and therefore the production of power and heat from fossil fuels, is by far the main emitter of fossil CO_2 in the EU. On the other hand, the energy industry is the only sector which continuously shows significant reductions in absolute emissions over recent years in the EU. Nevertheless, to achieve the goals of GHG emission reduction by 80–90% until 2050 compared to 1990 levels as stated by the European Commission [89], the use of fossil fuels in the energy sector must be reduced substantially or, rather, completely avoided. Therefore, the utilization of fossil CO_2 from the energy industry is excluded from further analysis.

In this context, it must also be stated that the mobility sector has not been investigated according to its potential for serving as a carbon source for CO_2-based process chains. This is justified by the consideration that efficient capturing from the source in this sector is not expected to be feasible, with acceptable capture rates, in the mid-term. Additionally, the high decentralization of the emitters makes the industrial usage of captured CO_2 unviable.

3.5. Incidence as point sources

To establish CCU applications on an industrial scale, the centralized availability of resources is an important aspect. To maximize the economic and ecological advantages of such CO_2-based applications and reduce costs and efforts for transportation and storage, their operation near to the carbon emitting process is highly preferable. For an overview of which industry sectors provide highly centralized emissions of CO_2, the average per site emissions have been evaluated in this study based on the categorizations and data described above. The results are shown in Fig. 12.

Fig. 12 illustrates the highly centralized CO_2 emissions from iron and steel and the refinery industry, which provide an average amount of approximately 420 and 330 kilotons of CO_2 per year and site, respectively. Another remarkable sector is represented by the cement industry (including clinker, lime and ceramics production). While average emissions per site are well below the two major ones, the number of sites in total is significantly higher, which allows a more distributed installation of CO_2 utilizing technologies, and thus is advantageous in site selection. In contrast to the fossil industrial emissions, per site emissions from biogenic sources are rather low, especially for biogases, which would propose a more decentralized implementation of CCU activities.

With regard to these high amounts of centrally available CO_2, along with the fact that many industrial processes are highly established and efficient and are expected to still be available in the mid-term, it is reasonable to expect that these carbon sources be considered for renewable products. This especially applies to the steelmaking and cement industry, where a decarbonisation would imply a complete revision of the process chain. Therefore, when investigating resource potentials for future CO_2-based process chains at an industrial scale, these point sources should be considered as well.

3.6. Efforts and costs of CO_2 capture

3.6.1. Upgrading technologies for biogas

Focussing on the two biogenic “low hanging fruit” CO_2 sources, namely bioethanol and biogas, the latter can contain significant amounts of impurities. These cause the need for further (costly) purification and hence can influence CO_2 capture costs substantially. Thus, some upgrading technologies are presented here. In contrary, CO_2 from bioethanol needs no or very little purification to be further utilized in other processes, which is why it is not in focus here.

There are several technologies available for biogas upgrading: water scrubbing, amine scrubbing, pressure swing adsorption (PSA), physical scrubbing, chemical scrubbing, membrane separation and cryogenic separation [21,76,91]. Since biogas upgrading mainly aims at the separation and purification of the methane content in the biogas, trace compounds are often removed together with the CO_2 stream as can be seen in Fig. 13. Consequently, the CO_2 stream may contain considerable amounts of impurities and is potentially not suitable for most utilization applications for chemical synthesis without further treatment.

According to the European Biogas Association [75], most current biogas upgrading plants use water scrubbers, followed by chemical absorption, PSA, membrane separation and physical absorption. In Germany, the European country with the most upgrading plants, all upgrading technologies are represented; however, chemical absorption and water scrubbing are each implemented in 30% of the plants, and PSA in 22% of the plants. In Sweden, ranked third among European biomethane countries, 69% of the biomethane plants use water scrubbers, followed by chemical absorption, PSA and membrane separation [75]. For the UK, ranked second, no information on upgrading technologies is available [75]. The only European countries known for implementing physical absorption are Germany, Norway and Switzerland. Combining this information with the paths for impurities according to Fig. 13, most CO_2 from upgrading plants requires further purification before it can be utilized for CCU pathways.

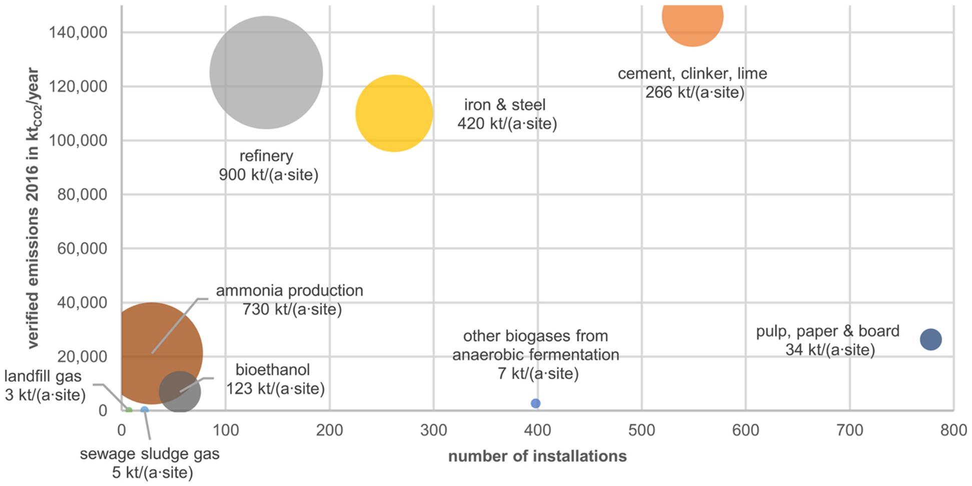

Fig. 12. Average CO_2 emissions in the EU per site and industry sector.
Source: based on data from [51,52,82,83,90].

The available CO_2 concentration in biogas upgrading off-gases depends on the applied upgrading technology. In processes like membrane separation, amine scrubbing and pressure swing adsorption, relatively high CO_2 contents are reached in the off-gas streams (typically up to 99 vol.-%, CH_4 being the balance). Depending on the remaining impurities and downstream processes, the resulting off-gases can be readily used for CCU options.

In contrast, processes that involve stripping with air, such as with pressurized water scrubbing, produce more diluted CO_2 off gases. This means that CO_2 from such processes would need further gas upgrading steps to remove air components and increase the CO_2 content.

A review on upgrading technologies for biogas to biomethane from Vijayanand and Singaravelu [92] gives an overview of CO_2 separation techniques for biogas; additionally, Singhal et al. [93] gives an overview of the transformation of biogas to biological compressed natural gas (bio-CNG). A comprehensive review on biogas generation factors, enhancements of biogas production techniques, upgrading and cleaning techniques are given by Al Mamun and Torii [94] as well as Andriani et al. [95] and Sun et al. [80]. Pellegrini et al. [96] give an overview of the purification costs of biogas, depending on the source of biogas.

Examples of emerging CO_2 separation technologies for biogas

A relatively new technique for biogas upgrading is the cryogenic CO_2 separation, which involves many different process steps using very low temperature processes. One example, which results in partially food grade CO_2, is the CO_2 Wash® process developed by US-based Acrion Technologies. It applies the effect of impurities solubility in liquid CO_2. After H_2S removal and drying, the biogas is mixed with liquid CO_2. This process results in biomethane, food grade CO_2 and a CO_2–VOC mixture as the products [91]. Depending on the requirements for the CCU pathways, further purification of the food grade CO_2 may be needed, for example, by activated carbon technologies [47,48]. Yousef et al. [97,98] improved cryogenic liquid CO_2 separation from Biogas and Tan et al. [99] present a system review and property impacts.

A new technique for CO_2 capture at room temperature using aqueous Na_2CO_3 has been presented by Barzagli et al. [100]. Chaemchuen et al. [101] and Cavenati et al. [102] presented metal-organic frameworks (MOFs) for upgrading biogas. Lim et al. [103] investigated clathrate-based CO_2 capture from biogas.

Because of the recent increase in popularity of CCU, but also due to the quality requirements of the chemical and FAB industries, purification processes of CO_2 are of high interest. For example, some impurities have corrosive properties, which is a problem in transport and long-term storage [104]; other impurities are harmful to chemical processes or toxic in terms of the FAB industry. For CCU, not only because of the transport but also because of the chemical conversion of the CO_2 and the possible poisoning of conversion equipment, very low impurity

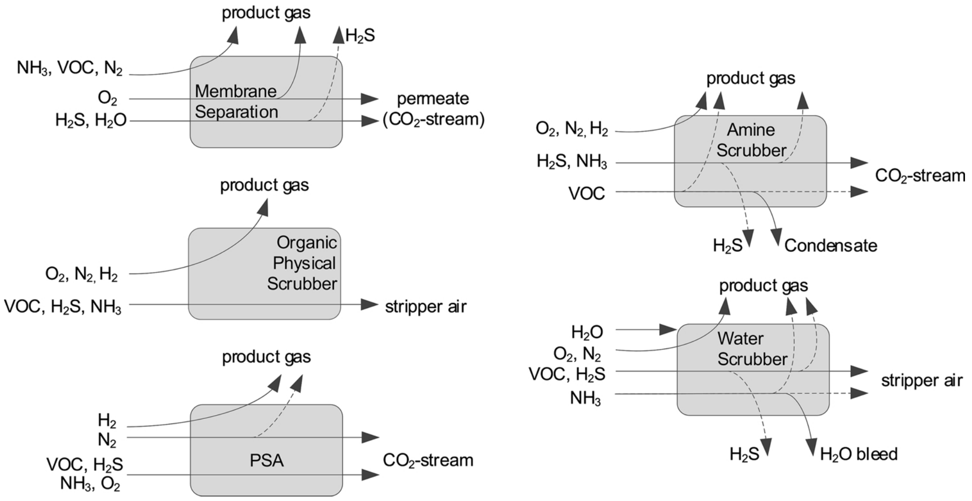

Fig. 13. Some exemplary paths of impurities from biogas upgrading technologies.
Source: based on Figure 12 from Hoyer et al. [76].

Table 6
Categorization of CO_2 providing industrial processes.

Main industry sector	Sub-category
power & heat from fossil fuels; energy industry	coal
	natural gas
	biomass
chemical industry	refinery
	ammonia production
	other chemicals
iron & steel	iron & steel
(other) metals processing	(other) metals processing
cement, clinker, lime, ceramics	cement, clinker, lime
	ceramics by firing (bricks, tiles, …)
production of glass & glass fibre	glass & glass fibre
pulp, paper & board	pulp, paper & board
biogenic processes	biogas upgrading
	bioethanol (fermentation only)
	bioethanol (fermentation & cogeneration)

concentrations are required. Despite CO_2 purification processes' state-of-the-art nature, much development work is ongoing.

3.6.2. Technological capture rates

The amounts of CO_2 discussed before provide an overview of the overall direct emissions from different sources. To evaluate the real potentials for utilizable CO_2, the appropriate efficiencies of certain technologies used for separation, which limit the amounts of CO_2 that can be captured and used, must be considered. In the following section, affordable capture rates and resulting utilization values are analysed.

To obtain an overview of the technically affordable capture rates for industrial processes, an appropriate literature review was conducted. This review closely mimics the industrial sectors discussed in Section 2. As some of these sectors include various technological processes, the analysis was performed on the even more fine-grained level of sector sub-categories, resulting in the categorization shown in Table 6.

The industrial processes covered by the categorization in Table 6 are significantly different from each other in terms of volume flows and purity of CO_2 in their flue or by-product gases. Additionally, each process allows the suitable use of one or more different capture technologies for efficient separation. This is particularly notable because efficiency often requires a compromise between the two, i.e., the process and capture efficiency. Consequently, highly different capture rates are technically and economically achievable, and thus may be actually implemented in existing processes and considered state of the art. These results are summarized in Fig. 14, showing the determined ranges and averages for the selected processes.

As can be seen in Fig. 14, for some sub-categories listed in Table 6, appropriate values cannot be provided. On the one hand, this is caused by the rough definition related to the available data for CO_2 potentials, which does not further specify the underlying process (e.g., other chemicals, (other) metals processing). On the other hand, some processes and industrial sectors do not provide sufficient data to estimate appropriate capture rates (e.g., glass and glass fibre production, ceramics by firing) or do not allow feasible CO_2 capturing at all.

These capture potentials significantly reduce the amount of CO_2 that is utilizable from the total amounts emitted. Presuming the average capture rates as shown in Fig. 14, the resulting potentials for CO_2 from industrial sources are reduced to the amounts shown in Fig. 15.

The capture efficiency also has an impact on the utilizable CO_2 emissions per site as shown in Fig. 16, according to the categorization given in Table 6. If fossil sources are considered, chemical industry processes, followed by iron and steel and cement production, would provide the highest amounts on utilizable CO_2 per site. Despite high capture rates, per site amounts for biogenic sources are still rather low in comparison. Therefore, for the large-scale application of CCU, fossil processes could be considered as a potential source for CO_2, as long as there are no environmental drawbacks and doing so does not support or elongate the deployment of processes that can and should instead be substituted by renewable approaches.

3.6.3. Carbon capture cost

Generally, the investment costs for CO_2 sequestration are not easy to define. It is reasonable to set a reference for specific costs according to the CO_2 source being used. Affordable sequestration rates strongly depend on the concentration of CO_2 in the (generally gaseous) source stream and the underlying emitting process. As the CO_2 sources and the reference values for assessing investment costs exhibit significant variance, it seems more practical to determine the value of the required CO_2 as an operating supply and therefore represent its costs as per ton (€/t) CO_2, depending on its source and sequestration technology, respectively.

Table 7 summarizes the gathered carbon capture costs for CO_2.

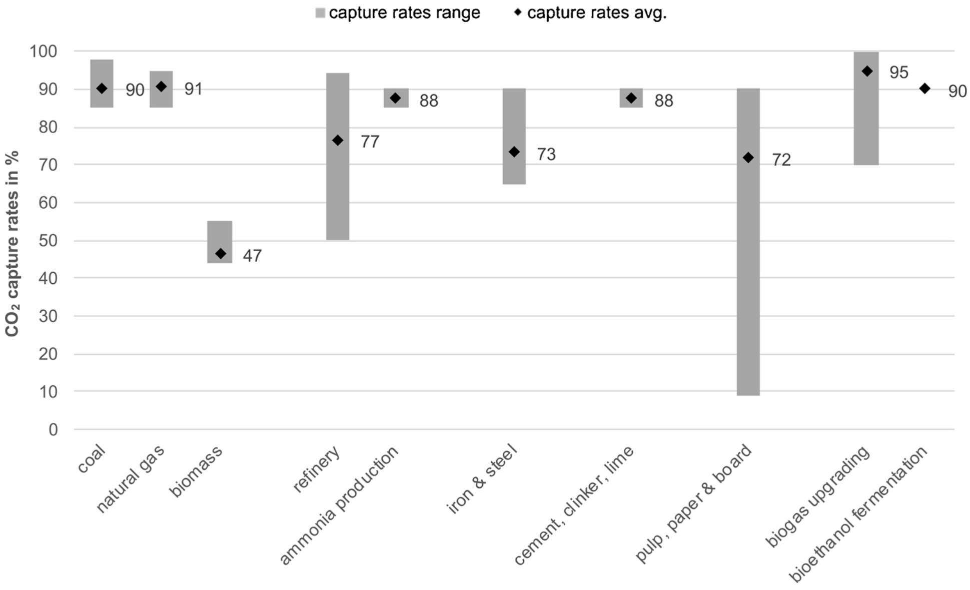

Fig. 14. Affordable capture rates for various industrial processes (ranges and average).
Source: based on data from [17,30,90,105–113].

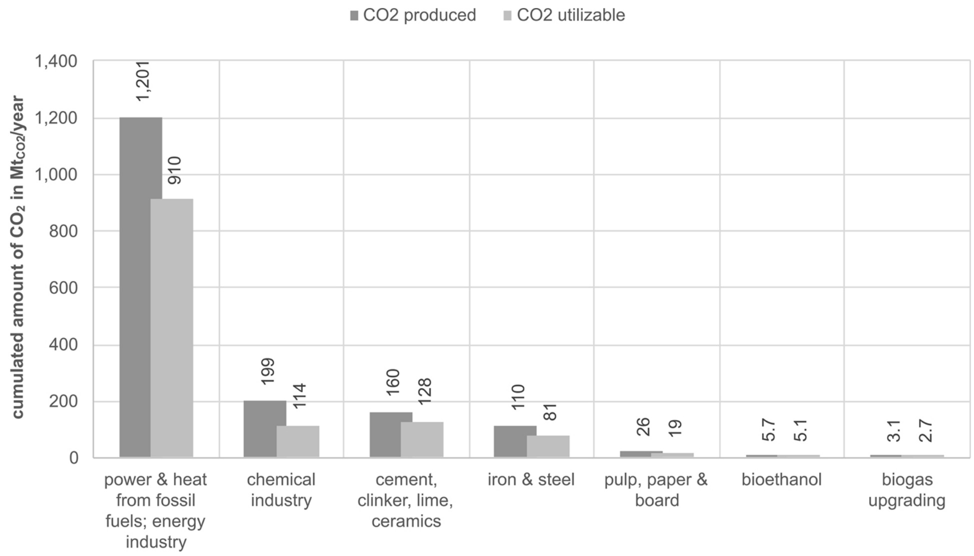

Fig. 15. Comparison of produced and theoretically utilizable CO$_2$ per industrial sector. Note: for biogas only the CO$_2$ output of existing upgrading plants is included. Source: based on emission data from [21,51,52,82,83,90] and capture rates from [17,30,90,105–113].

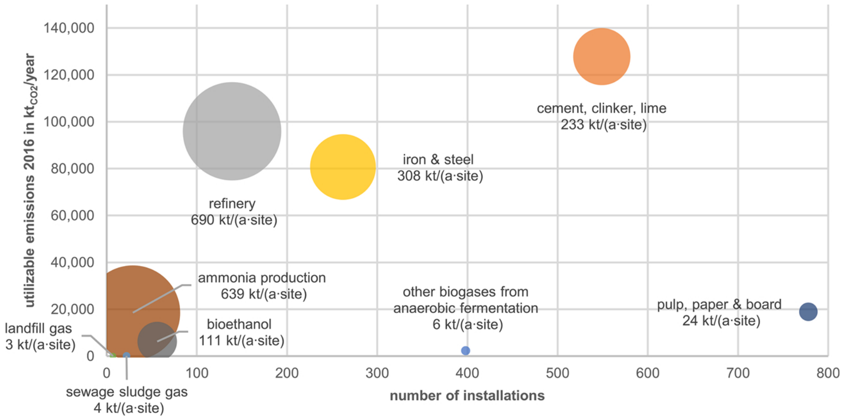

Fig. 16. Utilizable CO$_2$ potentials from various industrial processes.
Source: based on emission data from [21,51,52,82,83,90] and capture rates from [17,30,90,105–113].

Compared to the previous sections, the data herein was extended with CO$_2$ from fossil sources, though the acceptability of CO$_2$ from fossil sources for CCU must be further discussed (e.g., CO$_2$ may originate from waste gases from industrial processes which cannot be shifted to use as a renewable energy source, and therefore fossil CO$_2$ cannot be avoided).

As can be seen, capture costs for CO$_2$ are highly dependent on the source used. Whereas capturing from diluted industrial flue gases (combustion of natural gas or solid biomass, refinery) ranges from €50–100 per ton, efforts for sources with high concentrations (biogas upgrading, industrial bioethanol fermentation, ammonia production, etc.) are substantially lower, reaching values clearly below €50 per ton. Because of low concentrations of CO$_2$, direct air capture (DAC) shows the highest costs in conjunction with high uncertainties on account of the low maturity of DAC technology.

Besides general energy, investment and operation costs, the CCU cost structures are highly dependent on the properties of the CO$_2$ stream. Higher cost applications are those where waste CO$_2$ is produced in a more diluted form and potential harmful substances must be extracted prior to any utilization. Due to the technology readiness level of CCU there are large uncertainties, too, concerning scalability, the permanence and demand set of the capture as well as the cleanness of the future energy mix being used to power certain technological pathways.

Biogas plants which feed-in to the natural gas grid lend themselves as a source of otherwise unused CO$_2$ as characterized in Subsection 2.1.3. Specific costs for CO$_2$ sequestration in such biogas plants are approximately €12 cents per standard cubic meter of methane. Assuming a CO$_2$ fraction of 40 % in the raw gas flow, this unit price would lead to the cost of approximately €90 per ton CO$_2$ (for 2012). However, the sequestration, as well as the removal of impurities (like sulphur), is normally done for the retrieval of biomethane which can be fed into the gas grid, and hence costs are assigned to the methane production. In this aspect, the sequestration of CO$_2$ is neutral in terms of costs [30,105].

The costs of CO$_2$ from a bioethanol plant, as the source, would behave in a similar manner. In the fermentation process, a high-quality stream of CO$_2$ is accumulated as a by-product. If only this method is considered as a potential source, then the sequestration costs would be limited to the costs necessary for the compression of the gas, which can be assumed to be approximately €12–25 per ton CO$_2$ [17,30,106,107]. If the bioethanol plant uses cogeneration for energy provision and the CO$_2$ capture from the cogeneration process is also considered, then the costs would be between €42 [108] and €111 [106] per ton CO$_2$ for capturing and compression, respectively.

4. Conclusions

The present study revealed a significant potential for utilizable CO$_2$ emissions from already existing biogenic sources. Up to 23.15 Mt CO$_2$/

Table 7
Average capture costs for CO_2 related to industrial sectors.

CO_2 source		capture costs in €/t CO_2	year	ref.
energy industry; power & heat from fossil fuels	coal	34 – 42 $^{3)}$	2017	[17]
		19 – 47	2015	[30]
		20 – 63 $^{2)}$	2015	[109]
	natural gas	63 – 83 $^{3)}$	2017	[17]
		54 – 101	2015	[30]
		35 – 75 $^{2)}$	2015	[109]
	biomass	54 – 101	2015	[30]
chemical industry	refinery	29 – 83 $^{3)}$	2017	[17]
		44 – 94	2015	[30]
		48 $^{1)}$	2012	[110]
		97 $^{3)}$	2014	[107]
	ammonia production	12 $^{3)}$	2017	[17]
		23 – 54	2015	[30]
		22 $^{3)}$	2014	[107]
	other chemicals	12 – 52 $^{3)}$	2017	[17]
		21 $^{3)}$	2014	[107]
iron & steel production		19 – 33 $^{3)}$	2017	[17]
		16 – 41	2015	[30,110]
		81 – 83 $^{3)}$	2014	[107]
cement, clinker & lime production		22 – 35 $^{3)}$	2017	[17]
		33 – 69	2015	[30,110]
		17 – 37 $^{1)}$	2012	[110]
		82 $^{3)}$	2014	[107]
pulp, paper & board production		18 – 27 $^{2)}$	2003	[108]
		57 – 87	2017	[112,113]
biogenic CO_2 sources	biogas upgrading	0 – 90	2012	[105]
		5 – 9	2015	[30]
	bioethanol fermentation	12 $^{3)}$	2017	[17]
		0 – 18	2011	[106]
		25 $^{3)}$	2014	[107]
		5 – 9	2015	[30]
	bioethanol fermentation (incl. cogeneration)	83 – 111	2011	[106]
		42 $^{2)}$	2003	[108]
direct air capture		150 – 320	2012	[105]
		22 $^{1)}$	2012	[105]
		150 $^{2)}$	2010	[114]
		331 – 423 $^{2)}$	2011	[115]
		268 – 309 $^{2)}$	2013	[116]
		341 – 475 $^{3)}$	2014	[117]
		81 – 201 $^{3)}$	2018	[118]
		18 – 90 $^{1)}$	2019	[119]

$^{1)}$ long term prediction.
Exchange rates USD/EUR:
$^{2)}$ 0.72 – 0.8.
$^{3)}$ 0.81 – 0.86.

year could be derived from today's biogas production as a by-product [83], of which approximately 3.14 Mt CO_2/year are already separated during biogas upgrading [21]. Summing up, biogenic CO_2 from solid biofuel combustion (437 Mt CO_2), bioethanol fermentation (5.71 Mt CO_2), wine and beer production (1.48 and 1.44 Mt CO_2), implemented biogas upgrading (3.14 Mt CO_2) and combustion of remaining biogas (up to 57.9 Mt CO_2) amounts to approximately 506.7 Mt CO_2 produced annually in Europe. The assessment performed herein showed that, in reality, only part of this CO_2 potential is available for valorisation. Nevertheless, the amounts are vast.

Limitations of this study potentially are in the availability of up-to-date raw data, as comprehensive statistical data is difficult to access for some CO_2 sources due to missing recording obligations. Data was validated and completed via desk research. Additionally, conversion factors from statistical raw data (for example "sewage sludge" in TJ) to annual CO_2 mass amounts might differ to other studies. Conversion factors were presented to preserve comparability to a maximum extent.

There are several limiting factors for green CO_2 sources: 1) Solid, liquid and gaseous biofuels are distributed to many small applications, e.g., household fireplaces, motor vehicles and gas heating systems. The CO_2 emitted from these small consumers cannot be reasonably utilized, because of the high technical and economical effort. It is unlikely that the elaborate capture process would result in an ecological and sustainable system, which contradicts the intention of CCU. 2) CO_2 is already utilized as a raw material in the FAB and other industries (water purification, pulp and paper, metal industry, welding, electronics, refrigerant gas, fire suppression technologies etc.). 3) CO_2 from bioethanol fermentation (including wine and beer industry) is already utilized to some extent, e.g. as dry ice, gaseous fertilizer and food packing agent or for carbonating beverages.

Consequently, the major biogenic potential is derived from biogas upgrading plants, the remaining CO_2 from bioethanol production and flue gases from biogas combustion. The latter is combusted with air and the derived flue gas represents a lower CO_2 concentration (approx. 8–15 vol.-%). Consequently, this requires intense flue gas purification and separation, whereas biogas upgrading offers large amounts in high concentrations, but provides the side effect of potentially harmful trace compounds. The associated capture costs for CO_2 are highly dependent on the source used. Whereas capturing from diluted industrial flue gases (combustion of natural gas or solid biomass, refinery) ranges from €50–100 per ton, efforts for sources with high concentrations (biogas upgrading, industrial bioethanol fermentation, ammonia production, etc.) are substantially lower, with values substantially below €50 per ton.

Though the present paper provides a comprehensive overview of theoretic potentials of biogenic and fossil CO_2, the technical and economical usability is of significant relevance for appropriate implementation projects. In addition, the impact of such CCU efforts on the fulfilment of fossil energy/resource reduction goals and the increase of renewable energies has to be topic of further research. Lastly, the economic feasibility of different CCS/CCU pathways will be a decisive factor for their long-term competitiveness and final demand of CO_2 as a resource, dependent on processing efforts and regulatory aspects.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Funding: The authors would like to express their gratitude to the European Commission for the financial support of this research within the European Framework Programme for Research and Innovation Horizon 2020 (project CO2EXIDE, Grant No. 768789) and the Association Energy Institute at the Johannes Kepler University Linz as well as the Fraunhofer Institute for Interfacial Engineering and Biotechnology, Straubing.

Furthermore, the authors would like to thank all direct and indirect contributors to this paper, such as all project partners from the CO2EXIDE project and all authors, whose work was referenced in this paper.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jcou.2020.101219.

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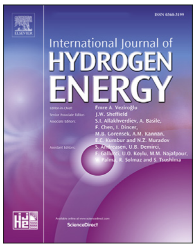

Power-to-hydrogen & district heating: Technology-based and infrastructure-oriented analysis of (future) sector coupling potentials

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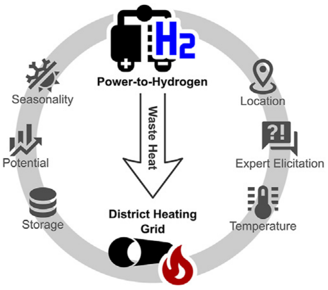
HIGHLIGHTS

- Discussion of practical synergies of power-to-hydrogen and district heating.
- By 2030 up to 12% of Austrian DH demand could be covered by electrolysis waste heat.
- 2.5–4% of EU heat demand <100 °C is discovered as electrolysis waste heat potential.
- Electrolysis is likely to be recognized equivalent to today's waste heat sources.
- Quantification of found synergies is important for future research.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 11 April 2021
Received in revised form
11 June 2021
Accepted 29 June 2021
Available online 17 July 2021

Keywords:
Power-to-hydrogen
Electrolysis
District heating
4GDH
Waste heat

ABSTRACT

Numerous studies suggest that power-to-hydrogen (PtH_2) will take a decisive part in future sustainable energy systems. District heating (DH) networks are also assigned a crucial role for the overall efficiency of such. In this regard, heat flows resulting from PtH_2 may lead to synergies with the heat supply of DH systems.

This paper discusses the potentials of PtH_2 as a relevant heat source for DH (with focus on Austrian system conditions). Technology-specific efficiencies, heat flows and temperatures are put in context with today's and future DH system specifications and synergies are analyzed. A qualitative analysis summarizes the opportunities and challenges that arise from a system perspective, e.g. electrolyzer location, user type, and user-specific operation (i.e. generation load).

It is found that high-temperature electrolysis is likely to be fully integrated in industrial utility operations and heat utilization corresponds to the well-known challenges of integrating industrial waste heat into DH networks. The location of low-temperature electrolysis is subject to infrastructure limitations and the economics of utilizing by-products. Operation is likely to be more electricity-market-oriented and may seasonally differ from

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https://doi.org/10.1016/j.ijhydene.2021.06.233

heat demand. However, its waste heat is sufficient to feed modern low-temperature DH networks and by 2030 could cover up to 12% of Austria's current DH demands and up to 4% of the EU demand for heat below 100 °C.

Introduction

Hydrogen and district heating are seen as two essential pillars of a future, sustainable energy system. This paper aims to identify technological synergy potentials between power-to-hydrogen (PtH_2) and district heating (DH) and to present infrastructure-related or system-inherent barriers and opportunities.

In the transition to renewable energy systems, hydrogen is expected to play a major role as a future energy carrier. In the recent past, various national governments have announced intentions to integrate hydrogen and power-to-gas (PtG) technologies to achieve climate goals [1]. The European Union has also announced their hydrogen strategy for a climate-neutral Europe in mid-2020, which targets the ambitious goal of implementing up to 40 GW electrolyzer capacities within the EU, alongside with additional 40 GW of electrolyzers in Eastern and Southern neighborhood, until 2030 [2]. In addition to the use as energy carrier, hydrogen is widely seen as feedstock material needed to decarbonize CO_2-intensive industries. This includes chemical industries such as ammonia production, which accounts for about one quarter of today's global hydrogen demand [1], or the future production of hydrocarbons and synthetic fuels in various power-to-X and synthesis processes [3]. Moreover, other energy-intensive industries, e.g. steel production, are expected to significantly increase their hydrogen demand when required to reduce carbon emissions [1], either due to novel approaches (e.g. Ref. [4]) or by decarbonizing existing process chains (e.g. Ref. [5]). Without claim to completeness, these given perspectives make clear that hydrogen from electrolysis will be of considerable relevance in future energy systems.

Despite growing demand for renewable hydrogen, the significant energy losses of the water electrolysis process are undisputed. Using low-temperature electrolysis, approx. 60–70% of the electricity input is transferred to the product; the residual energy is dissipated to heat [6]. Logically, that waste heat can be used for heating purposes; conceptual studies propose its usage (e.g. Ref. [7]) and demonstration projects have started in new-built quarters (e.g. in Esslingen [8]). The potentially utilizable waste heat output of different electrolysis technologies (alkaline electrolysis – AEL, proton exchange membrane electrolysis – PEMEL, solid oxide electrolysis – SOEL) and its development over the next decades is quantified by the Danish Energy Agency and Energinet [9], showing an increasing relevance for utilization. Presently, the waste heat is not used in commercial systems, but future utilization is technically feasible. However, an increase of the efficiency of electrolyzers goes along with a decrease of utilizable heat. Saxe and Alvfors [10] conducted a similar but more detailed study on calculating the waste heat potential of electrolyzers. Waste heat potential, composed of process excess heat, cooling of stack and product gases, radiation and convection losses, can reach up to 29% of the energy input. With temperatures of 50–60 °C, it could be utilized for preheating the return line of the DH network. The additional utilization of by-products beside waste heat such as oxygen further increases the economic viability [11].

Regarding the increasing relevance of renewable gases in energy networks, Jensen et al. [12] analyzed the role of PtG in the electricity and DH market. They showed, that if hydrogen is used for methanation to synthetic natural gas (SNG), the waste heat resulting from electrolytic hydrogen production or the downstream methanation reaction can be of significant relevance for internal recycling or feed-in to a DH network. Of course, appropriate DH networks must be available and economical in order to actually absorb electrolysis waste heat [13]. Ikäheimo [14] conducted a study of the economic viability of a PtG plant integrated in a future decarbonized DH system. The system is built up on a heat pump, heat boiler, heat storage and a biomass CHP plant utilizing carbon, and leads to the result of being a profitable option in the future. Howard and Bengherbi [15] go further by proposing to transform the gas grid in general into a hydrogen running grid and to modify existing gas boilers to achieve a low carbon heat supply.

DH networks will also play a crucial role in the macroeconomic energy efficiency of a future, sustainable energy system. DH can use low-grade renewables and absorb waste heat in order to provide customers with thermal energy and hot water at the exergetically appropriate temperature level [16]. However, there is broad consensus in the scientific literature that DH networks need to evolve, especially with regard to lowering the flow and return temperatures [17]. With the so-called 4GDH (4th Generation District Heating), Lund et al. propose a vision of the overall future DH system [18]. The lowering of the network temperatures is relevant as it enables additional and new heat sources to be (easier) integrated into the network: these include solar thermal energy, geothermal energy, heat pumps [19,20], but also waste heat sources that provide lower temperature levels [21].

Despite an obvious potential for heat utilization from electrolysis-based hydrogen production, the topic is hardly covered in available literature. Therefore, this paper aims to investigate from a practical and systemic perspective, if realizing synergies between electrolysis and DH is realistic. Thus, this paper focuses on whether a positive interaction between water (and steam) electrolysis and DH networks is possible in the overall energy market/system, taking into consideration the mutual effect of current and future technology characteristics as well as the effect of the enabling infrastructure. In order to clearly delimit the paper, electrolysis (i.e. the production of hydrogen with electricity) is considered exclusively as a source and DH as a sink (see

Fig. 1). This system perspective analysis is based on a technical analysis of resource (PtH_2) and demand (DH grid) side as well as a qualitative conclusion based on the elicitation of expert responses. To figure theoretical future waste heat potentials, up-to-date projections of PtH_2 capacities based on Austrian and European energy and climate plans applied on common electrolyzer characteristics. Therefore, the paper is considered as the first to discuss potential synergies of emerging electrolytic hydrogen production and evolution of DH networks from a systemic perspective.

It is known that fuel cells can be a substitute for conventional CHP systems and that these can be used specifically to generate heat for process or heating purposes, or that the waste heat from fuel cells generating power should be used for heating purposes [22]. While these technologies are expected to have a decisive part in future (hydrogen-based) energy systems [23,24], and thus enforce the role of hydrogen production, they are excluded herein for direct heat integration. Same accounts for hydrogen-related downstream processes, such as methanation. Additionally, the individual use of waste heat from electrolysis for heating purposes in general, e.g. directly in individual buildings or micro-networks [8], is excluded. It is not intended to describe or elaborate any ideal technology combinations or potentials settings for the realization of synergies.

Materials and methods

In order to derive the potential synergies between electrolysis and DH, the paper is using two different approaches. The first approach analyses the status and development of electrolysis technologies and their characteristics (see sections Electrolysis technology review and Technology demand and waste heat potentials) as well as DH networks and potential routes of development (see section District heating systems). The future synergies of electrolysis technologies and associated waste heat potentials and demand are elaborated in an Austrian context under consideration of national energy and climate plans. In addition, an estimation is made for the EU based on their recent hydrogen strategy.

The second approach uses a qualitative elicitation of expert responses to discover *actual* opportunities, i.e. opportunities that are realistic considering energy system and market functioning, and infrastructure boundaries (see section Qualitative system analysis). Opportunities and obstacles are summarized in a SWOT analysis.

Definitions and assumptions

Before explaining the methods applied in the respective analyses, definitions and assumptions are made and denominations are clarified in order to avoid ambiguity. The following definitions and assumptions are made as a starting point for general understanding:

- A future sustainable energy system is defined as follows: in the long term no fossil CO_2 is released into the atmosphere and the energy system is provided entirely by renewable energies. This definition of renewable energy systems is consistent with appropriate contributions in this field [25–27].
- In addition, a future sustainable energy system shall be defined to imply a highly efficient use of energy. This results from the fact that many studies suggest a shortage of renewable energy if today's demand would have to be covered [28,29].
- Renewable *electricity* represents the majority of the *energy* generated in a future, sustainable and renewable energy system [30].
- Renewable hydrogen is defined to be produced from renewable sources (renewable electricity) or without fossil CO_2 emissions (biogas reforming or pyrolysis).
- Synthetic natural gas (SNG) is understood to mean CH_4 produced from the combination of renewable (electrolysis) hydrogen and carbon.

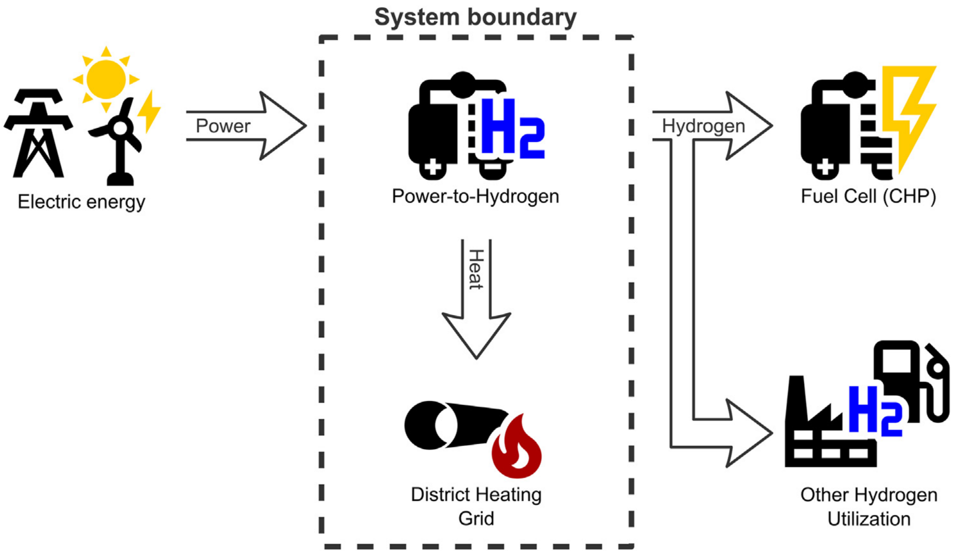

Fig. 1 – System boundary of the evaluation in this paper.

- Power-to-X, in the context of this paper, covers all technologies and processes that generate energy carriers from electric power, such as gas (hydrogen, methane), fuels, chemicals, etc.
- The DH demand is assumed for the Central European context, using Austria's largest networks as exemplary cases. This means that the heating and hot water demand is high in the winter months and significantly lower in the summer months, but still present (about 10–15%).

Electrolysis technology review

To serve future demands for hydrogen, electrolysis powered from renewable energy sources is considered as a major technology for renewable hydrogen production. Depending on the centralization of hydrogen production, electrolysis systems with capacities in multi-MW or GW range will be needed. Thus, their thermal integration within the future energy system is an obvious target in terms of sector coupling and overall energy efficiency. However, potential heat integration and temperature levels mainly depend on the underlying technology and mode of operation. Table 1 provides an overview on today's state-of-the-art electrolysis technologies.

Low temperature electrolysis

Low-temperature electrolysis describes technologies supplied by feeding liquid water and operating at temperatures usually below 100 °C. These include alkaline and proton exchange membrane (PEM) electrolysis, which represent today's most mature and commercially available technologies. Low-temperature electrolyzers are operated above their thermoneutral voltage[1] to compensate high internal losses and overvoltages [6]. Thus, the stack is generating waste heat during operation, which requires external cooling and could therefore be utilized. Due to the low temperature levels (50–80 °C for PEMEL, 60–90 °C for AEL [6]), options for waste-heat recovery are limited (see Fig. 2). Taking additional losses of heat transfer and transportation into account, spatially close applications and direct integration are considered to be the preferred paths.

High temperature electrolysis

Compared to low-temperature, high-temperature electrolysis can be supplied with steam and thus reducing the total energy demand by the heat of evaporation (see Fig. 3). Beyond that, depending on the operating temperature, high-temperature electrolysis offers to replace significant parts of the electric energy input by thermal energy and therefore allows for external heat integration [6]. This implies that, depending on whether the cell is operated below or above thermoneutral voltage, it either acts as a heat sink or source. However, typical operating temperatures of 650–1000 °C are significantly above common DH temperature levels and partly contradict the intentions towards lower heat supply temperatures [17] (cf. section District heating systems).

Technology demand and waste heat potentials

Future demand potentials for renewable hydrogen from electrolysis are widely dependent on the respective scope and the implementation of national and international hydrogen strategies. From a global perspective, the immediate demand can roughly be estimated according to the growing demands in the chemical and refinery industry, but also in the production of iron and steel, presuming a sustainable decarbonization of these sectors until 2050 [1]. According to Böhm et al. [35], these would result in an annual electricity input related electrolysis demand of 6640–7590 TWh_{el}. Taking storage capacities for fluctuating energy sources into account, Pleßmann et al. [36] suggest that an additional demand of 2360 GW_{el} could be needed globally. From a European perspective, the EU has targeted in their recent Hydrogen Strategy an increase of electrolysis capacities of 6 GW_{el} until 2024, and 80 GW_{el} until 2030 [2].

However, to consider non-industry sectors, including mobility power and heat supply, a global evaluation is not constructive, due to regional differences (e.g. in climate, energy generation and use, etc.). In combination with the potential of heat integration, the regionality of DH grids has to be taken into account. Thus, a national consideration of the Austrian conditions and projections is reasonable. Based on the national energy and climate plans, *Fuel Cells and Hydrogen Joint Undertaking* (FCH JU) has evaluated the *Opportunities for Hydrogen Energy Technologies* in Austria [37]. The study presents a demand of hydrogen generation capacities, namely electrolyzers, of 550–1960 MW_{el} for an annual hydrogen production of 1590–5620 GWh by 2030. These include demands of the four relevant sectors, industry, buildings, transport and power.

With respect to the technological differences in the available electrolysis technologies (see section Electrolysis technology review) the potential heat output, both in energy amounts and temperature levels, is dependent on the according technology shares. Based on the future production shares suggested in recent studies [35,38], the following capacity-related installation shares are presumed for the year 2030: 84% for AEL, 14% for PEMEL and 2% for SOEL technology. Based on Li et al. [7] and DEA [9], AEL and PEMEL are presumed to provide a waste heat potential of 20–30% of their nominal capacity, while the potential for SOEL is neglected, due to their operational characteristics (see section High temperature electrolysis).

District heating systems

DH networks, as they are mainly used in large cities in Central and Northern Europe, correspond to the second or third generation of the categorization according to Lund et al. [18]. Table 2 shows the typical temperatures of the DH networks. For the analysis carried out here, it is relevant that the supply temperatures of these two network generations are around 100 °C. For example, the Austrian DH networks in the provincial capitals Linz, Graz and Salzburg show flow temperatures of 120–130 °C. The return temperatures are around 50–60 °C; these temperatures should enable domestic hot water to be processed without additional resources (heating rod, booster heat pump).

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[1] Thermoneutral cell voltage describes the minimum voltage required for electrolysis in an ideal cell without heat integration.

Table 1 – Key characteristics of state-of-the-art water electrolysis technologies; based on [6,31,32].

Technology	Alkaline (AEL)	Proton Exchange Membrane (PEMEL)	Solid Oxide (SOEL)
Technology status	Commercial (mature) TRL 9	Commercial (mature) TRL 8-9	Pre-commercial (R&D) TRL 5-6
Operating temperature	60–90 °C	50–80 °C	650–900 °C
Typical stack capacity	<10 MW	<5 MW	<100 kW
Electric efficiency (LHV)	63–71%	60–68%	100%[a]
Load flexibility	20–100%	0–100%	−100%/+100%
Cold start-up time	1–2 h	5–10 min	Hours
Warm start-up time	1–5 min	<10 s	15 min

[a] Operation at thermoneutral voltage.

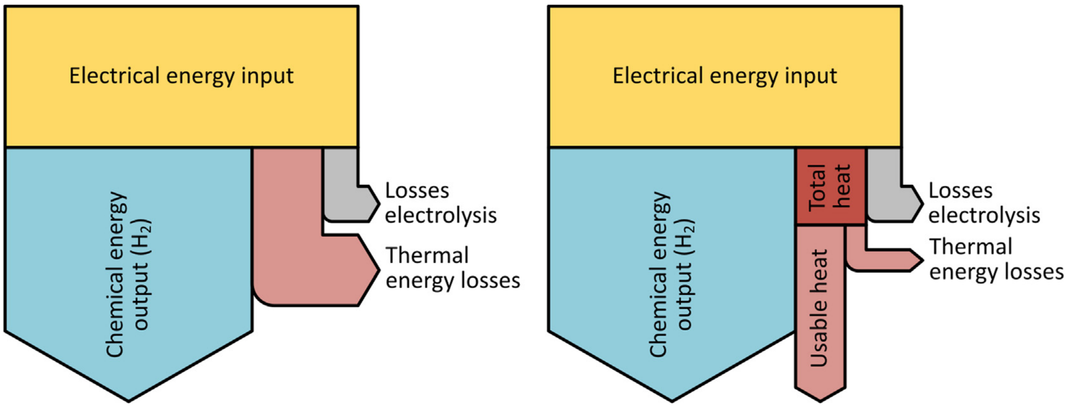

Fig. 2 – Energy flow diagram for electrolysis with (right) and without (left) heat extraction; based on [33].

The temperatures around or above 100 °C can be traced back to the fact that many DH networks replaced oil boilers and had to supply at least at the same temperature in order to further use the rest of the building's heating system. At elevated temperatures (but unchanged pipe diameters and costs) corresponding amounts of energy can be transported while on the supply side the necessary exergy, especially from fossil fuels, is available inexpensively [42].

Lower network temperatures cause lower losses and allow better integration of low-carbon energy sources, contribute to the reduction of greenhouse gas emissions and provide energy-economic benefits [43]. DH networks are an integral part of a sustainable energy system as they make a contribution to the efficiency of the overall energy system and can use low-exergy sources (e.g. solar energy, waste heat) [18]. Therefore, as a general statement, it can be derived that newly constructed DH networks should belong to this fourth generation (reasons for postponing the fifth generation can be found in Ref. [44]). A technical analysis of potential implementation was done by Volkova et al. [41]. Due to the options arising with low temperatures, sources like solar, geothermal and heat pumps are investigated [45,46]. For the reasons mentioned, existing DH networks should also be transformed towards the fourth generation. This transformation primarily concerns the reduction in network temperatures (flow, return) as pipes are already installed (cf. Fig. 4). The lowering of the temperatures of existing DH networks is particularly possible when new buildings are connected or larger buildings/blocks are comprehensively renovated. Therefore, one strategy is to lower the return temperatures by setting up low-temperature subnetworks [41]. Comprehensive renovation measures allow buildings to get by with lower temperatures and volumes [47], but customer behavior is also decisive for a transition [48]. New business models are expected to support temperature reduction e.g. by overcoming split incentives [49]. A different approach towards future DH networks is the interconnection of individual grids to supra-regional DH networks as suggested by Moser and Puschnigg [50].

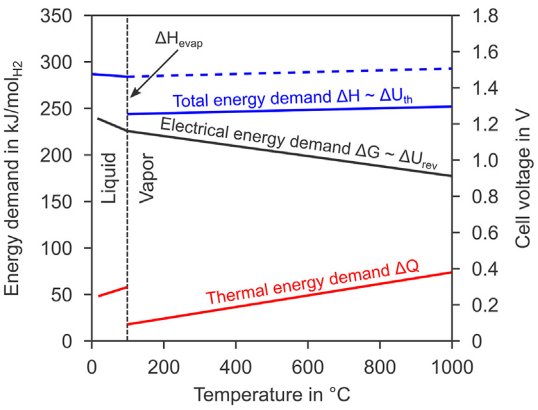

Fig. 3 – Total (ΔH), thermal (ΔQ) and electrical (ΔG) energy demand of an ideal electrolysis process as function of the temperature; based on [6,34].

Above-mentioned transition pathways and potential solutions are confronted with many barriers, humbling the transition of existing DH networks towards 4GDH. These include:

Table 2 – District heating network temperatures and heat carrier [18,39,40]. The temperature range of 4GDH is an indication as other authors, e.g. Ref. [41], slightly deviate.

Generation of district heating	Heat carrier	Temperature	Framework
1GDH	Steam	Steam	Dangerous heat carrier
2GDH	Pressurised hot water	Mostly over 100 °C	Individual pipe construction
3GDH	Pressurised hot water	Often below 100 °C	More lean components and prefabrication
4GDH	Low-temp. water	30–70 °C	Intensify prefabrication and pre-insulation
5GDHC	Water	0–30 °C	Heating and cooling, booster heat pumps

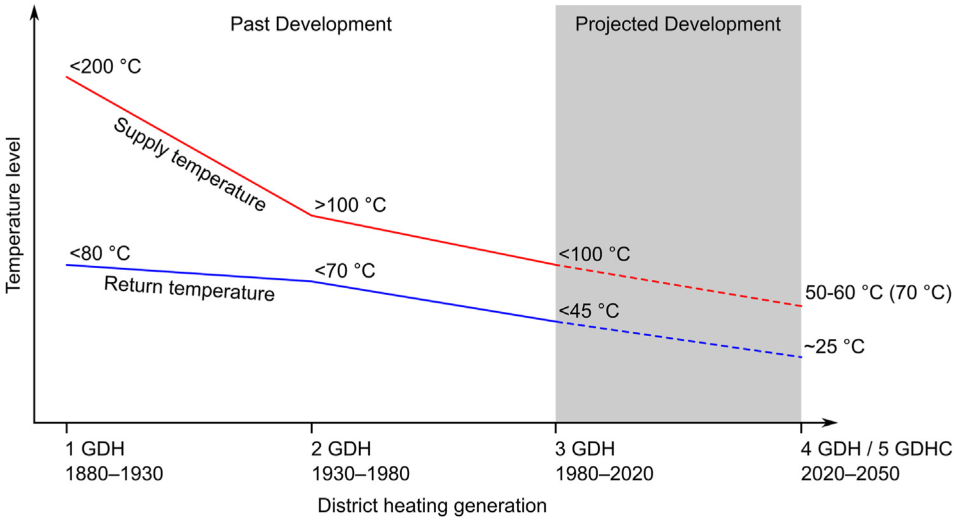

Fig. 4 – Past DH temperature development and projected transition pathway for today's 2GDH/3GDH networks towards 4GDH network in a sustainable energy system. Based on Table 2 and timeline based on [18].

- Network hydraulic factors decrease reliability or increase pump costs [51].
- Potential to guarantee supply as network capacity decreases with temperature delta and pressure might be limited [52].
- Supply flow temperatures must satisfy the building's requirements (undersized substations) [49].
- Supply flow temperatures must satisfy the building installation's requirements (undersized radiator surfaces) [49,51].
- Technical restrictions (valves, bypasses, sensors, etc.) [49] and more complex management of the overall system [53].
- Contracts (including temperatures) must be complied with [52].
- No substation monitoring data [49].
- Split incentives [49].
- No direct supply of domestic hot water [44].
- Lacks of economic efficiency in the short/middle term [44] as business model is still not developed [53].
- Difficult adaption in dense areas [44].

These obstacles may result in a poor diffusion of low-temperature DH networks [53]. Thus, it is reasonable to calculate with close to 100 °C today, but to assume that some existing and larger networks remain at flow temperature levels slightly below 100 °C also in the long run.

Qualitative system analysis

This paper aims to elaborate the applicability of the synergies, given an energy system and infrastructural framework. Thus, the practical applicability of waste heat from electrolysis in DH networks means that a number of challenges have to be solved. These challenges are systemic; they are interdisciplinary and not linearly linked. Before systemic relationships can be quantified, it is relevant to understand the causal relationships. The method of expert interviews enables internal validity, but is suited to be able to derive actual correlation. The illustration of argumentative-based and comprehensibly-presented qualitative results should not be misunderstood as descriptive or general. A description or elaboration of any ideal technology combinations for the realization of synergies is not in the scope of this study.

Practical issues that have a direct effect on the usability of the PtH_2 waste heat in DH networks arise primarily from the economic considerations of the operators. These system questions include:

- Where will PtH_2 be located at all? Is the electrolyzer positioned at a sufficiently short distance from the DH network?
- Are the operating temperatures of electrolysis suitable for feeding into the local DH network?

- Is there a sufficient seasonal overlap between the DH network's heat demand and electrolyzer operation?
- Can the operating mode of the electrolyzer be sufficiently predictable for the DH network operator or can interruptions be handled through storage and backup capacities?
- If the electrolyzer is positioned at a third party (e.g. industrial companies), what options are there to intervene and what contractual options and barriers result from this?

As a basis for the expert interviews, profound literature search is conducted, identifying and investigating the interaction between electrolysis and DH. In doing so, reference is made to fundamental statements made in scientific papers on the characteristics of energy technologies, energy markets and energy infrastructure. Then, interviews are conducted with 11 experts; they are chosen as their expertise is virtually located in the intersection of hydrogen and DH. The experts come from research institutions, energy suppliers and energy-intensive industries. They cover the areas of DH, power supply, hydrogen technologies, energy-intensive industrial processes and energy system aspects. Due to the urgent desire of some experts for complete anonymity, names and companies are not provided and the statements are not assigned. This is possible with the expert interview method given the focus on the argumentation; of course, the authors are aware of the associated scientific restriction but the results are thoroughly presented and if experts deviate from a common line of argument, this is clearly indicated.

The interviews took place via phone or video conference. The expert survey was conducted as a guided interview. The individual questions in the guideline are clustered into subject areas. The clusters are “technologies and technological developments”, “expediency and applicability with regard to demand and provision”, “implications from infrastructure” and “questions regarding the position of the synergy between hydrogen and DH in the overall energy system”.

The result is a categorized overview, which summarizes beneficial and prohibiting aspects of the interaction/synergies of electrolysis and DH. The empirically supported analysis has a qualitative character and represents a summary as complete as possible with a focus on the infrastructural and systemic aspects.

Results and discussion

The following sections summarize the results of the present study, both for technical and potentials analysis and the qualitative expert elicitation.

Technical potentials in future energy systems

An increasing demand for renewable hydrogen will be accompanied by the installation of electrolysis and therefore raise the question of utilization of corresponding waste heat potentials. Based on the demand potentials for electrolysis as described in section Technology demand and waste heat potentials, the aggregated waste heat potentials from low-temperature electrolysis in Austria are expected to reach 530–2810 GWh_{th} annually by 2030, presuming a utilization potential of 20–30% of the nominal power [7]. Even though with temperature levels of 50–90 °C a direct integration in today's common DH systems (second and third generation; see section District heating systems) is mainly possible in the return line. However, continuously decreasing temperature levels in future DH networks [41,42,54] is about to improve utilization potentials. Beyond that, electrolyzer waste heat could be utilized as an input for heat pump applications to minimize the efforts for domestic and also process heat supply.

Taking the EU hydrogen strategy into account, the associated waste heat potentials would be in a range of 16–24 GW_{th} by 2030. Presuming a cost optimized operation of these electrolysis capacities at approx. 3500 full-load hours (as proposed in Ref. [1]), this would result in an annual supply of thermal energy of 56–84 TWh_{th} linked to the European renewable hydrogen supply.

To put these figures into context, the DH sales in Austria relate to approx. 22 TWh annually [55]. Thus, 2–12% of the Austrian DH demand could be supplied by renewable hydrogen production until 2030. Beyond that, there is an additional demand potential of about 21 TWh from industrial space heat and hot water supply and process heat at temperatures below <100 °C [56]. Taking into account that electrolyzers could preferably be placed in close distance to industry sites, a significant amount of this heat demand could be directly supplied from electrolysis in the future. Similar considerations can be done for the EU scope, which accounts for an annual DH demand of 437 TWh [55]. Besides, a demand potential for industrial low temperature heat (<100 °C) of approx. 608 TWh for process heat, space heat and hot water exists [57]. Based on the EU electrolysis distribution plans (under consideration that only half of the capacity is planned within the EU), 6–10% of the DH demands could be covered by electrolysis waste heat utilization, or 2.5–4% in total, respectively, if industrial demands are considered.

When looking at high-temperature electrolysis, an integration in conventional DH is controversial, since its operating temperature is conflictive with before mentioned developments to lower grid temperatures. Quite contrary, with increasing demand for hydrogen and electrolysis in energy-intensive industries (cf. [1,35]), an endothermal operation and appropriate heat integration is expected in industrial use cases to maximize the electric efficiency of the hydrogen production. Consequently, if these industrial waste heat sources are rather used in internal electrolysis processes, this could take away essential heat resources from future DH networks. However, based on current technology status and the projected market shares (cf. [38]), the heat integration of high-temperature electrolyzers in the context of DH is expected to be neglectable compared to low-temperature counterparts until 2030.

For completeness, it has to be mentioned that the future demand for renewable hydrogen is to a large extent generated by the need as a feedstock material for further downstream synthesis processes (e.g. Refs. [58,59]). While the present study focuses on power-to-hydrogen the subsequent treatment to produce higher value hydrocarbons often comes with additional waste heat potentials. In addition, exothermal catalytic processes, such as methanation or Fischer-Tropsch synthesis, provide these at elevated temperatures (i.e. >150 °C, [60,61]). However, due to the variety of potential options for hydrogen upgrading, these are out of the scope of this study and have to be treated individually.

Qualitative system analysis

The technological requirements (i.e. suitable performance and temperatures) for leveraging the synergies between hydrogen and DH were dealt with in the previous section. As it turns out, from a technological point of view, the possibilities are given. From the perspective of hydrogen, the use of the waste heat to increase overall efficiency is clearly desired. This leads to the question whether and how these synergies can be practically realized in the energy system considering infrastructures and markets. Due to the fact that the analysis looks at a (i) complex and (ii) future system, a qualitative analysis was chosen. Thus, in addition to literature research, these system aspects and their effects on the realization of the synergies between hydrogen and DH are qualitatively analyzed through expert interviews (see method in section Qualitative system analysis). Please note that all findings, which result from the literature research, are referenced as usual, while any other statements are based on the expert interviews.

The results of the analysis are described in the following sub-sections. To summarize the most relevant results from the qualitative assessment a SWOT analysis is provided in section SWOT-oriented summary.

General parameters for the integration of waste heat from electrolysis

From the point of view of the DH operator, waste heat from electrolysis is in competition with conventional heat supply; it also competes with other waste heat sources. This means that the following known requirements for the use of waste heat also apply to hydrogen technologies' waste heat [62–64]:

- Low cost of recovery
- Sufficient temperatures and/or low preparation costs
- Low costs of supply infrastructure (pipelines, transfer station)
- Short-term constancy (low production fluctuations, i.e. low need for storage)
- Long-term availability (no risk of bankruptcy, no serious process modifications that avoid waste heat, no relocation to more efficient locations)
- Presence of backup systems
- Low value of alternative use of waste heat (i.e. that there is no cost-efficient use, e.g. on the site or for electricity generation)

If the electrolysis is located in an industrial company, local industrial heat use (e.g. from an exergetic point of view, when process steam is required, or when high-temperature waste heat is available for high-temperature electrolysis) can be more efficient than external use. If electrolysis is located in an industrial company, those barriers that already today characterize the connection between industry and DH networks also apply to leverage the synergies from industrial electrolysis and DH [63,64].

Seasonality

The question of seasonality discusses when electrolyzers are operated (and thus the waste heat is available) in the course of the year and whether this is congruent with the common DH network requirements.

Power-to-hydrogen demand. Power-to-Hydrogen is considered to be essential for mid-to long-term energy storage, as well as hydrogen production for industry – as an energy carrier and feedstock (sometimes in connection with the capture and storage of carbon) – and mobility, especially in the heavy load sector [65]. No clear seasonality of hydrogen demand can be derived from this.

Using hydrogen as a heating energy source for decentralized boiler units is generally possible and it can rely on existing infrastructure (gas distribution grid, gas boiler). However, if other heat energy sources are available (as there are on the market), using hydrogen for heating is often considered inappropriate due to the high conversion losses. Of course, this demand would result in a seasonal consumption profile that is aliquot to the DH demand. DH itself may need hydrogen or derivatives for peak demand (see the experts' arguments in 3.2.4.1) [66,67].

Power-to-hydrogen supply (electrolyzer operation). From a ceteris paribus perspective, electrolyzer operation is more economical if (i) the system has many operating hours per year and (ii) is operated with low electricity costs [1,68]. Since electricity costs are generally varying over the year, there must be a case-specific optimal trade-off between (i) and (ii) [1,35]. With a broader consideration of other decision parameters, e.g. the environment of the electrolyzer, it is expected that other results will be obtained: For example, the electrolyzer could also be operated continuously for the supply of industry, especially when processes are continuous and the electrolyzer can use industrial waste heat as an electric-efficiency enhancing input (see 3.2.5.2). Depending on the electrolysis technology, dynamic operation (see Table 1) considering real-time electricity price or the provision of system services (control energy) is possible, especially with low-temperature technologies [69]. Higher discontinuity can be assumed if hydrogen storages or networks are available or hydrogen is specifically generated for storage [70,71].

Batteries and pumped storage power plants function as efficient [72] hourly to weekly storage [28] and thus compensate for short-term fluctuations in PV and wind power. It is to be expected that they will stabilize electricity prices over these short-term periods. Therefore, it becomes clear that electrolyzers react less to acute price changes than to prices averaged over several days/weeks in order to achieve a high degree of utilization.

Assuming that a certain proportion of the electrolyzers operates depending on the current electricity price, the following questions arise: Will the times of low electricity prices in a future sustainable energy system show a seasonality and if so, how strong will it be? In other words: Will the proportion of PV electricity, which is primarily available in summer, be so dominant that there is a large summer-winter storage requirement? Or is the proportion of wind power high enough to have such a small difference between power generation in summer and winter that the electricity prices seasonally differ little enough that electrolyzers will also be operated in winter?

Consequently, there is disagreement not whether but how great the role of PtH_2 as seasonal storage is in a sustainable energy system. If the role of wind power was dominant, hydrogen would be less relevant as a seasonal storage medium, since it can also be produced economically in winter. Literature clearly states that a long-term or seasonal demand for storage capacities in form of PtH_2 exists if a high proportion of volatile renewable energy is in the system [73–76], but this is true also for back-up reasons. However, recent studies also show that this seasonality of power-to-hydrogen energy storage is mostly independent of the underlying energy demand profile (residential vs. commercial) [77].

Comparison of the scenarios. Since literature does not allow to dissolve the above-posed questions on seasonality of operation, the supply of waste heat is considered for the two scenarios (summer and winter). Remember the assumption that a certain proportion of the electrolyzers operates depending on the electricity price and that short-term fluctuations are buffered by other storage types and demand response.

- With seasonally lower prices in summer, the waste heat from electrolysis is likely to be mainly available in the summer months. This waste heat could cover summer heat demands but is confronted with low heat demand and competing waste heat sources (see further arguments on that in 3.2.3.2). However, since the relevance of wind power in a sustainable energy system is undisputed, the low-temperature electrolysis will also have operating hours in the winter half-year, but the amount remains unclear and thus the economic feasibility of connecting to DH.
- Without seasonal fluctuations in the price of electricity, the electrolysis would also be in use in winter. If the waste heat meets the winter temperature requirements of the DH network, it can be fed in. Uncertainties in production would be a shortcoming (known from the feed-in of industrial waste heat, see 3.2.1) and are likely to be considered in terms of lower remuneration.

Criteria for the position of electrolyzers

The future location of an electrolyzer depends on many factors. The experts' responses support the hypothesis that the actual allocation can hardly be estimated. The actual positioning of the electrolyzer will be a techno-economic case-by-case decision, weighing up the following factors mentioned by the authors.

According to the experts, these factors include.

- Position and type of electricity generation.
- Position and composition of the consumers.
- Availability and capacity of the local electricity network or the electricity network along the way of energy transport.
- Costs of setting up a hydrogen network or the availability and capacity of the local gas network or the gas network on the way of energy transport.
- On-site or close-to-site usability of by-products from electrolysis such as oxygen or heat can also be critical.

Position criterion: usability of the by-product oxygen. In addition to heat, oxygen is also generated as a by-product of electrolysis. If cost-efficient synergy potentials arise, this increases the economic efficiency of the electrolyzer [11]. This includes use in industrial or communal processes. For example, in combination with oxyfuel combustion processes, NO_x emissions can be avoided and the process emissions contain purer CO_2, which makes carbon capture technically easier and more economically efficient [78,79].

Position criterion: usability of the by-product heat. Not only in total (in the entire energy system), but also in individual cases, efficiency losses in the production of hydrogen are a highly relevant issue for profitability. It is obvious that if the electrolyzer is positioned away from DH networks, the waste heat cannot be used. As an illustration of such a situation, experts cite a potential position of the electrolyzer close to electricity generation, e.g. near PV plants in the Sahara or wind farms in the North Sea.

The proximity of the electrolyzer to DH networks does not automatically mean that the waste heat is used. The waste heat competes with other heat generators, which have to be assessed in terms of heating costs, controllability, reliability, temperature, amount of energy, integration costs, etc. [62]. This competition also includes waste heat from industry or waste incineration. The place and time of the electrolysis essentially determine the competitiveness; however, experts agree that the key decision criterion for positioning the electrolysis will be the conversion from electricity to hydrogen (keyword "core business" [64]). Although, the development of supra-regional heating networks as proposed by Moser and Puschnigg [50] could support the integration of decentralized electrolysis capacities.

Position criterion: type of consumer and type of electrolyzer. Experts state that one ideal location of a low-temperature electrolyzer are larger sewage treatment plants (see also [80]). There, the waste heat can be used directly and oxygen can also be used effectively in the treatment process.

For the potential integration of high-temperature waste-heat a spatial proximity of high-temperature electrolyzers to industry or other sources, e.g. waste incineration plants, is to be expected favorable, although the direction of heat exchange is dependent on electrolyzer operation (see section High temperature electrolysis). If industrial companies provide high-temperature waste heat potentials and a need for hydrogen, experts consider the industry's site a logical location for high-temperature electrolysis.

Industrial companies can choose whether hydrogen is produced on-site, produced by a third party on-site, or produced elsewhere. Combinations are also possible: it is also an alternative that an electrolyzer is on-site while additional hydrogen is obtained. The existing infrastructure (connection to the electricity and heating network) and its expansion are also expected to play an important role here.

Energy system aspects

CHP plants and/or boilers for the security of heat supply. There will be significant surpluses, especially in summer, from waste heat, solar heat, waste incineration, or waste heat from unavoidable power plant operation [81]. Heat storage systems are therefore a central component in the future DH network in order to bring these summer surpluses into winter. For reasons of cost efficiency, the experts see a techno-economic threshold for the sizing of seasonal storages, at which the remaining heat required in winter is covered by (renewable) fuel, i.e. the storage of hydrogen or derivate fuels for the DH network must be weighed against the storage of heat in large seasonal heat storage system. In the same context, the integration of hydrogen in CHP-based heat supply is considered as a valuable addition to balance peak loads in terms of decreasing operation costs in multi-carrier energy systems [66,67].

Waste heat as a traditional element of district heating. Experts point out the relevance of inexpensive energy for DH networks. It is their “original nature” to use inexpensive heat (waste heat, formerly especially from the power plant), also to be able to stem the high network (pipeline) investments (see also [16]). For reasons of the required overall system efficiency in a future sustainable energy system, waste heat will only be available at lower temperature levels, since higher temperature levels should be supplied to exergetically suitable applications. Large water-based heat storage systems are also more economically efficient at low network temperatures due to their temperature limitations (<100 °C) and thus the higher temperature spread [81].

From this it follows that the DH network (and also the buildings heated via DH) must join/follow/contribute to the temperature reduction [82]. According to the experts, this transition to low-temperature DH networks should be started proactively today: it can already bring advantages today, and may have a supportive effect on the positioning of electrolysis, as it can absorb the waste heat.

Possible application of hydrogen technologies

Low-temperature electrolysis. Experts expect that, for economic reasons (high annual utilization of the asset) and due to seasonality, low-temperature electrolysis is likely to be operated almost continuously over the summer half-year (the amount of electricity available is higher in summer due to PV generation and hydrogen storages are charged) (see the restrictions in section Seasonality). This means that low-temperature electrolysis is a reliable supplier of waste heat in the summer months and can be used, for example, in DH to cover the summer base load. Due to the regular use of low-temperature electrolysis in summer, fluctuations in heat output are likely to be low. It follows that the low-temperature electrolysis would be well suited to cover the base loads of the DH network. Due to temperature levels, use in next-generation DH networks (4GDH [18]) all year round is conceivable, if DH networks remain at high temperatures (especially second generation district heating), which is possibly due to a lack of ambition to convert (as stated by an expert). This potential may remain unused in winter.

With low-temperature electrolysis, 20–30% losses occur in the form of waste heat between 50 °C and 90 °C (see sections Electrolysis technology review and Technology demand and waste heat potentials). The temperature of the waste heat is hardly suitable for feeding in typical second-generation DH networks or for charging water-based seasonal storage tanks. From a technological point of view, higher operating temperatures (e.g. around 100 °C) would be preferred from the low-temperature electrolysis. While this is a subject of recent research (e.g. Ref. [31]) it is considered complicated due to component and operating material limitations in PEM and alkaline electrolyzers (gas-tight and ion-conductive membranes or liquid electrolytes).

If it is true that low power prices (excess energy) will mainly be available in summer and (especially low-temperature) electrolysis will thus mainly be operated then (see the corresponding option in 3.2.2.3), this scenario implies either low prices or low quantities:

- Due to the number of sources and quantities of excess heat in summer [81], which are competing for application in limited summer sinks, the summer value of waste heat from the electrolysis is expected to be low.
- Moreover, typical current DH networks (2GDH, 3GDH; cf. [18]) operate at high/higher temperature levels in winter. Feeding into the return line might be an option. However, in this scenario, the heat amounts produced by the electrolyzer are expected to be low due to the focus on summer operation.

High-temperature electrolysis. High-temperature electrolysis offers advantages compared to low-temperature processes, especially if inexpensive heat input is available at elevated temperature levels. With high-temperature heat being available as input, electrical efficiencies >100% can be achieved (see section High temperature electrolysis). Even at lower heat input temperatures, significantly better efficiencies can be achieved compared to low-temperature electrolysis, at least if providing the heat of evaporation for the steam input (see Fig. 2). The high-temperature electrolysis can efficiently utilize heat sources available at a sufficiently high temperature level [83,84]. This includes, for example, waste heat from energy-intensive industry or heat from waste incineration plants.

If high waste heat temperatures are available in the energy-intensive industry, these can be used directly for high-temperature electrolysis on site. Energy-intensive industrial companies that have their own on-site high-temperature electrolysis can benefit from this, i.e. they can use their own waste heat in the electrolyzer and use the remaining heat on-site (e.g. steam) and without the risks and complexities of external cooperation.

Thermal management is already a key topic in high-temperature electrolysis research. The heat is a major input here, so minimizing waste heat is an efficiency objective [85].

Although the waste heat is circulated, it can be assumed that relevant waste heat temperatures >200 °C are remaining from the product gases [86]. Thus, depending on the system design and utilization, an industry-internal use e.g. in the steam network or for other industrial applications, as well as re-use in the DH network are technically possible.

SWOT-oriented summary

The elaborated analyses brought up technical opportunities and expert positions for the integration of renewable hydrogen production technologies in today's and future DH systems. These findings from the previous subsections are compiled and summarized in the following SWOT (Strengths, Weaknesses, Opportunities, Threats) analysis.

Strengths – Technology-specific advantages

- From a technical perspective, electrolysis offers sufficiently high temperatures to feed into the DH network, at least in the return line.
- Some degree of freedom on the final position within an area and thus potentially located close to DH sites.
- High-temperature electrolysis is likely to become an integral part of future industrial processes, envisioning a good (at least company-internal) use of their waste heat.

Weaknesses – Technology-specific disadvantages

- From a market perspective, waste heat temperature, especially from low-temperature electrolysis, might be too low to compete with other alternative or conventional sources.
- The operation of electrolyzers and the demand in DH networks could diverge seasonally.
- Electrolysis is one source of waste heat among others – they compete with other sources and cannot really offer better characteristics (e.g. due to volatile production, potential relocation).
- Electrolysis faces the same problems on feed-in into DH systems as today's waste heat sources.

Opportunities – System advantages

- Electrolysis will be indispensable in a sustainable energy system and demand will be enormous. Hence, the waste heat potential will exist and DH operators (facing goals towards renewable energy [55]) may actively look to integrate such sources.
- The integration of industrial sites in DH networks already before electrolysis is installed will ease the feed-in of its waste heat.
- Seasonal heat storages can be a summer heat sink and thus an advantage for some hydrogen technologies.

Threats – System disadvantages

- Infrastructure (electricity grid) limitation may enforce the allocation of electrolyzers near renewable power generation and away from heat consumption.
- Oxygen as another by-product of electrolysis could be given more weight for positioning than heat sinks.
- Some DH system may not manage to decrease flow/return temperatures, decreasing techno-economic feasibility to feed in.

Conclusions

This paper shows that there are several synergies and efficient interactions between power-to-hydrogen and (district) heating systems. The analysis of current power-to-hydrogen technology status and their projected deployment according to recent hydrogen strategies has revealed a significant waste heat potential. Especially for temperature levels below 100 °C as intended in modern DH networks, 2–12% of today's DH demand in Austria could be covered by electrolysis waste heat by 2030 (or 6–10% for the EU). Taking the additional industrial demand for space heat, hot water supply and process heat <100 °C into account, electrolysis could still serve 1.2–6.5% of the Austrian or 2.5–4% of the EU demands, respectively. While the experts highlight the importance of using the waste heat in order to achieve a high macroeconomic primary energy efficiency, the topic of waste heat from hydrogen electrolysis seems to be barely taken into account by the DH sector. The discussed topics show, that the integrability of power-to-hydrogen in modern DH networks is basically possible, especially for low-temperature electrolysis, but strongly depends on various additional systemic parameters. Besides an industry-beneficial placement to optimize techno-economic synergies of by-product use and reduce transportation efforts, the seasonality of operation can significantly affect the potentials for waste heat utilization. Summer-oriented operation due to lower electricity costs could aim to serve base loads of DH, but is expected to have to compete with existing heat sources. Furthermore, the qualitative analysis has affirmed the presumption that high-temperature electrolysis is preferred to be applied in close interaction with heat-intensive industry and thus providing low potentials for DH integration.

There is a need for further research, particularly in order to quantitatively substantiate the qualitative results obtained here. Moreover, effective planning tools could be developed, mapping the expected demand for hydrogen, heat and oxygen.

In view of the kW-scale performance or the technology status of demonstrators, waste heat is not yet a criterion for the position of electrolyzers. Various arguments can be put forward for the geographical or infrastructural location of electrolysis technologies; however, a clear conclusion on positioning is not possible at this stage, demanding further research. A positioning far-off potential heat sinks is inevitably associated with technical energy losses of 30–40%. Effective macroeconomic positioning is desirable, but further research is also required.

CRediT author statement

Hans Böhm: Writing - Original Draft, Visualization, Investigation, Validation. Simon Moser: Conceptualization, Methodology,

Formal analysis, Writing - Original Draft, Supervision. Stefan Puschnigg: Investigation, Writing - Review & Editing. Andreas Zauner: Validation, Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This paper was prepared within the Austrian participation in the IEA DHC Annex TS3 “Hybrid Energy Networks” (contract # 876727), which is financed by the Austrian Research Promotion Agency (FFG).

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frontiers
in Sustainability

ORIGINAL RESEARCH
published: 27 September 2021
doi: 10.3389/frsus.2021.726332

Techno-Economic Assessment of Thermally Integrated Co-Electrolysis and Methanation for Industrial Closed Carbon Cycles

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Energy-intensive industries still produce high amounts of non-renewable CO_2 emissions. These emissions cannot easily be fully omitted in the short- and mid-term by electrification or switching to renewable energy carriers, as they either are of inevitable origin (e.g., mineral carbon in cement production) or require a long-term transition of well-established process chains (e.g., metal ore reduction). Therefore, carbon capture and utilization (CCU) has been widely discussed as an option to reduce net CO_2 emissions. In this context, the production of synthetic natural gas (SNG) through power-to-methane (PtM) process is expected to possess considerable value in future energy systems. Considering current low-temperature electrolysis technologies that exhibit electric efficiencies of 60–70%$_{el,LHV}$ and methanation with a caloric efficiency of 82.5%$_{LHV}$, the conventional PtM route is inefficient. However, overall efficiencies of $>$80%$_{el,LHV}$ could be achieved using co-electrolysis of steam and CO_2 in combination with thermal integration of waste heat from methanation. The present study investigates the techno-economic performance of such a thermally integrated system in the context of different application scenarios that allow for the establishment of a closed carbon cycle. Considering potential technological learning and scaling effects, the assessments reveal that compared to that of decoupled low-temperature systems, SNG generation cost of $<$ 10 c€/kWh could be achieved. Additional benefits arise from the direct utilization of by-products oxygen in the investigated processes. With the ability to integrate renewable electricity sources such as wind or solar power in addition to grid supply, the system can also provide grid balancing services while minimizing operational costs. Therefore, the implementation of highly-efficient power-to-gas systems for CCU applications is identified as a valuable option to reduce net carbon emissions for hard-to-abate sectors. However, for mid-term economic viability over fossils intensifying of regulatory measures (e.g., CO_2 prices) and the intense use of synergies is considered mandatory.

Keywords: co-electrolysis, methanation, power-to-gas, techno-economy, oxygen utilization, CCU, cement industry, steel industry

OPEN ACCESS

Edited by:
Andrea Lanzini,
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Reviewed by:
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Specialty section:
This article was submitted to Circular Economy, a section of the journal Frontiers in Sustainability

Received: *16 June 2021*
Accepted: *27 August 2021*
Published: *27 September 2021*

Citation:
Böhm H, Lehner M and Kienberger T (2021) Techno-Economic Assessment of Thermally Integrated Co-Electrolysis and Methanation for Industrial Closed Carbon Cycles.
Front. Sustain. 2:726332.
doi: 10.3389/frsus.2021.726332

INTRODUCTION

Based on its goal of achieving climate neutrality by 2050 and reducing greenhouse gas emissions to at least 55% below 1990 levels by 2030, the European Union has just recently tightened its climate target plan (EC, 2020). While major efforts toward these goals have been put forth by the extension of renewable power sources, the requirement for alternative energy carriers such as renewable gases remains evident (Jensen et al., 2020). Particularly for industrial production processes, decarbonization by electrification is not always possible due to the required energy densities or the process-related demand for specific materials or gases. Additionally, individual production processes induce carbon emissions that are not fuel-related, and thus, they are still present in scenarios with a completely sustainable energy supply. For example, this applies to mineral industries such as cement production, where up to two-thirds of the CO_2 emissions are related to carbon that is bound to raw materials. Hence, carbon capture and storage (CCS) or utilization (CCU) is an inevitable measure of according roadmaps to carbon neutrality (CEMBUREAU, 2020). Similar requirements apply to heavy industry processes such as crude steel production that currently rely on long-term optimized and well-established processes, where a transition to renewable processes requires the establishment of fundamentally different production routes (Rechberger et al., 2020). Intermediate alternatives based on the methanation of steel gases that are available in conventional steel production processes have been previously investigated (Rosenfeld et al., 2020).

In this context, power-to-gas or power-to-methane can be considered as a valuable option for utilizing these process-related carbon emissions and generating a re-integratable substitute fuel (or process gas). Based on the implied requirement for carbon capture and internal reuse, closed carbon cycles can be created to thus omit net carbon emissions. However, the available low-temperature electrolysis technologies only allow for moderate system efficiencies of 46–60%$_{\mathrm{LHV}}$, related to the lower heating value (LHV) (Buttler and Spliethoff, 2018). In combination with catalytic methanation, the overall power-to-methane efficiency is ∼40–50%$_{\mathrm{LHV}}$ when assuming complete conversion (Frank et al., 2018). With solid oxide electrolysis (SOEL), higher electric efficiencies can be achieved due to operation at temperatures of 600–1,000°C (Zheng et al., 2017), particularly when supplied with steam (Buttler and Spliethoff, 2018). The appropriate heat demand can be supplied from external sources or, in the case of power-to-methane, from the exothermal methanation process that can be operated at temperatures of 250–700°C (Götz et al., 2016; Rönsch et al., 2016). Additionally, SOEL technology provides the ability to perform co-electrolysis of H_2O and CO_2, thus allowing for the generation of a suitable syngas composition for the downstream methanation process (Banerjee et al., 2018; Biswas et al., 2020). These synergies allow to significantly increase the overall system efficiencies by a high thermal integration of the electrolysis and the methanation process (see **Figure 1**). In particular, the waste heat emitted in the methanation reactor is directly utilized to evaporate and preheat the water supply for the steam-driven co-electrolysis without external heat supply. Thus, the external energy demand, primarily electricity, is reduced by the heat of evaporation or beyond depending on individual operating conditions. By implementing these systems at or in close proximity to relevant industries efforts for storage, transport and distribution could additionally be reduced and thus efficiencies increased even further.

The resulting gross reaction equations for an appropriate combination of co-electrolysis and CO methanation are provided in Equations (1 and 2) according to a previous study (Krammer et al., 2021). With thermal integration, an overall thermal efficiency of 83%$_{\mathrm{LHV}}$ could be achieved for the power-to-methane process.

$$H_2O \rightarrow H_2 + 0.5\,O_2 \qquad \triangle H_R^{1023K} = 248.1\ \frac{kJ}{mol} \tag{1}$$

$$CO_2 \rightarrow CO + 0.5\,O_2 \qquad \triangle H_R^{1023K} = 282.3\ \frac{kJ}{mol} \tag{2}$$

$$CO + 3\,H_2 \rightarrow CH_4 + H_2O \qquad \triangle H_R^{553K} = -215.9\ \frac{kJ}{mol} \tag{3}$$

Considering these elevated efficiencies, such a system is also expected to provide higher economic competitiveness in regard to operation than are conventional power-to-gas systems. The general positive effect of heat integration for high-temperature electrolysis in terms of hydrogen production was reported by Buttler et al. (2015). The significant increase in the efficiency of power-to-X processes by thermal integration of co-electrolysis and downstream synthesis, like Fischer-Tropsch, was also found by Herz et al. (2018). More recently, Zhang and Desideri (2020) determined that co-electrolysis-based power-to-methanol implementations can reach payback times of 3–5 years for optimized systems. While former studies emphasize the impact of electricity prices on product generation costs, the competitiveness of such a system is highly dependent upon the costs of the SOEL stack. Since high-temperature electrolysis technology is still in a lab-scale and R&D status (Grigoriev et al., 2020), current investment costs are expected to decrease considerably. Hence, the present study considers the impact of learning and scaling effects for future implementation scenarios as projected by Böhm et al. (2020). Additionally, recent studies have demonstrated that the utilization of by-product oxygen significantly impacts the techno-economic performance of power-to-gas plants in certain application scenarios (Röben et al., 2021). Therefore, this impact was closely managed within this study.

To ensure sustainable production and reutilization of synthetic gas from fossil carbon, the use of renewable power is mandatory in addition to the establishment of a closed carbon cycle. Therefore, the direct integration of renewable energy sources such as wind or photovoltaics (PV) is a major topic for future power-to-gas plants. The flexibility to follow intermittent production and to operate in a grid-supportive manner is an important factor in successfully applying power-to-gas processes for decarbonization (Schreiber et al., 2020). However, due to comparably long start-up times and high standby energy demand (Buttler and Spliethoff, 2018; Smolinka et al., 2018), high-temperature electrolysis is often considered as less suitable for operation with intermittent energy sources. This aspect was thus

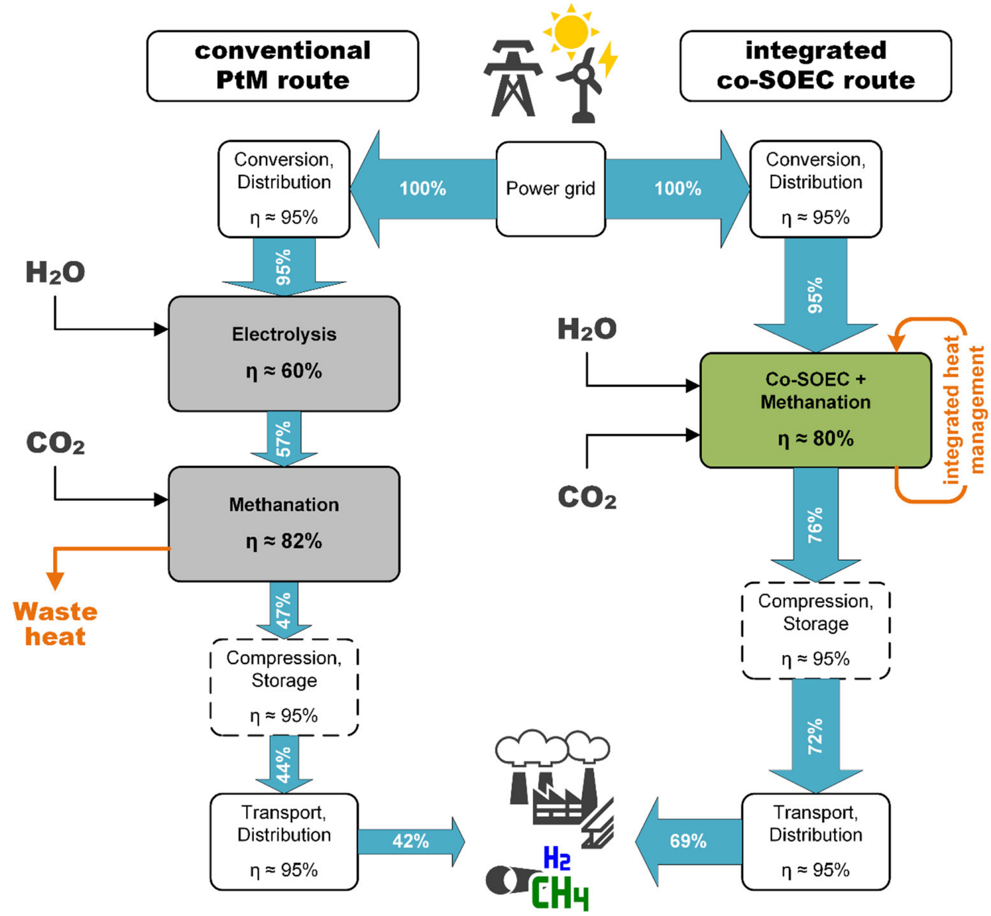

FIGURE 1 | Comparison of efficiency chains of power-to-methane routes using conventional technologies vs. the thermally integrated co-electrolysis approach.

given attention to in the evaluation of electricity supply in the investigated implementation scenarios.

The present study aims to analyze the competitiveness of co-electrolysis-based power-to-gas implementations in application scenarios that are not expected to become completely carbon neutral in the mid- or long-term based on their origin. Therefore, a comprehensive techno-economic assessment was performed under the presumption of the creation of closed carbon cycles. Hence, appropriate costs for CO_2 capture and emission abatement savings are taken into account. Additionally, to increase the techno-economic performance, the potential for internal and external utilization of by-products is considered. While focusing on two apparent implementation scenarios, the study does not intend to cover all relevant use cases. Instead, it identifies economic performance of the technology that come with an exploitation of synergy potentials.

MATERIALS AND METHODS

System Boundary and Process Definition

The analyses in this study focus on the integrated power-to-gas system that includes co-electrolysis and catalytic methanation. The system is presumed to be implemented in close proximity to an industrial carbon source (industrial site) that simultaneously utilizes the produced gas as a substitute natural gas (SNG). Thus, the system enables a closed carbon cycle without net carbon emissions. Process streams such as electricity, CO_2, and O_2 from and to the power-to-gas system are only considered according to their energetic and economic values and are otherwise outside of the system boundary. The process scheme is illustrated in **Figure 2**.

Due to the thermal coupling of the co-electrolysis and methanation subsystems, the power-to-gas system is expected to achieve an overall electric efficiency of $> 79\%$ that is related to the LHV of the product gas. This type of process is currently under development in the Austrian flagship research project HydroMetha that targets (FFG, 2018) an overall electric efficiency of $>80\%$ without external heat integration. Thus, the projections for future scenarios presume an increase in efficiency up to 82.5% by 2050. The general feasibility of these efficiencies was confirmed by a previous study (Wang et al., 2019). Although the industrial scenarios investigated in the techno-economic study aim for high annual loads of the appliance, the investigations examining direct supply from wind power imply the occurrence of part-load operation. However, as appropriate studies on the part-load operation of high-temperature electrolysis have

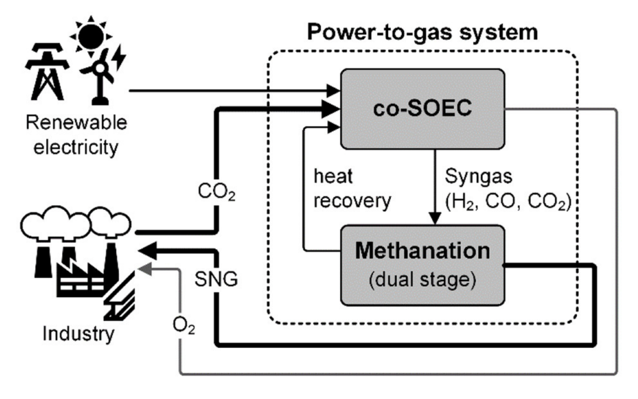

FIGURE 2 | System boundary of the power-to-gas system establishing a closed carbon cycle with the integrating industry (bold streams).

suggested (Buttler et al., 2015; Sanz-Bermejo et al., 2015), an operation exhibiting constantly high efficiencies is possible and presumed as feasible for the analyses. Additionally, for large-scale applications, part-load operation will be less of an issue due to the incorporation of modularization (Smolinka et al., 2018).

The methanation subsystem is considered as a two-stage and dual pressure level methanation process. This configuration was investigated by Krammer et al. (2021) with relation to integration with co-electrolysis as part of the HydroMetha project. Their analyses revealed that a two-stage process with intermediate compression to up to 10 bar resulted in the highest methane concentrations in the product gas. For the calculations herein, a product gas composition with a methane content of 89.0% and a hydrogen content of 9.5% was assumed, resulting in an LHV of the product gas of 9.13 kWh/m^3. Although a hydrogen content of >4 vol% does not allow for a direct feed-in to Austrian public gas grids without further processing, the product gas is presumed to be suitable for industrial combustion processes as a natural gas substitute (Krammer et al., 2021). The substitution of conventional fuels is considered equivalent to the LHV.

For dual-level pressure methanation, an intermediate compression of the co-electrolysis synthesis gas is necessary. Therefore, isothermal compression with an electric energy demand of 0.153 kWh per m^3 at an efficiency of 75% according to a previous study (Campbell et al., 2014) was presumed. Based on the industrial application scenarios and similar load flexibility and thermal integration of the electrolysis and methanation processes, the requirement for additional (intermediate) gas storage in the investigated use cases was omitted.

Scenario Definition and Implied Parameters

Based on the constraints inherent to the decarbonization of these well-established industries and their local value in the Austrian energy transition, cement production and primary steelmaking were chosen as relevant use cases for this techno-economic CCU assessment. Furthermore, due to the evaluated process temperatures, both applications provide the potential for additional heat integration. The different integrations of the power-to-gas process for these scenarios are discussed below. However, both scenarios share certain implications such as utilization of oxygen or participation in the European emission trading system (EU ETS).

For the evaluations presented in the scope of the present paper, only the economic effects of synergetic utilization of products (SNG and oxygen) and recycling of carbon emissions are considered in relation to the defined use cases. Thus, the underlying process configurations and potentially required adaptions are considered to be available and feasible. The parameters related to the application scenarios are summarized in **Table 1** and are justified in subsequent chapters.

Cement Production

The production of cement results in the emission of 0.54 t CO_2 per ton cement (IEA, 2018). According to Rodin et al. (2020), the average per-site emissions of cement plants under the EU ETS account for ~266 kt per year. As 60–65% of these emissions are related to the calcination process (CEMBUREAU, 2020) and thus are not directly affected by switching to renewable fuels, approaches for CCU/CCS are mandatory for the decarbonization of this industry sector.

In addition to amine-based CO_2 capture, calcium looping, and membrane-based technologies, oxyfuel combustion is a widely discussed option to capture CO_2 from the calcination process (Rolfe et al., 2018; Ditaranto and Bakken, 2019; Voldsund et al., 2019). This would allow for a high concentration of CO_2 (> 95 vol%) to be present in the flue gas and thus reduce the efforts required for separation and conditioning for utilization. Therefore, the combustion air is substituted with oxygen to eliminate the presence of inert nitrogen. The required oxygen is commonly produced by dedicated cryogenic air separation (CAS). With the integration of electrolysis, the generated by-product oxygen could (partially) substitute the need for external production, thus reducing the operational costs of the oxyfuel process. According to a previous study (Carrasco-Maldonado et al., 2016), the O_2 demand for oxyfuel in a medium-sized cement production plant is estimated to be ~0.25–0.35 t O_2 per ton clinker or 162–228 kg_{O2}/t_{cement} at a clinker to cement ratio of 0.65 (IEA, 2018).

Based on Austrian industry data, (VÖZ, 2017) the fuel demand per ton cement produced is ~750 kWh, and ~78% (i.e., 588 kWh) of this is covered by alternative fuels. The residual non-electric energy demand is supplied by fossil fuels (coal, fuel oil, coke, and natural gas). Despite the relatively low content of natural gas (~0.66 m^3/t_{cement}), it is presumed that the use of solid and liquid fossil fuels is driven by economic aspects rather than by technical reasons, and thus, a substitution by SNG is possible.

Steel Production

Although the direct reduced iron (DRI) process using hydrogen is a current topic of extensive research for renewable hydrogen production (Rechberger et al., 2020), it requires a complete adaptation of well-established and highly efficient processes. As these transitions of production require long-term efforts, alternative pathways for the integration of renewables and internal utilization of carbon emissions have been investigated

TABLE 1 | Scenario-specific calculation parameters.

Parameter	Unit	Cement industry	Steel industry
CO_2 source	–	Kiln off-gas	Steel gases
CO_2 capture costs	€/t	39.1	38.1
Annual full load hours of production	h/a	8,000	8,500
Fuel demand	kWh/t_{cement} or kWh/t_{steel}	162–750	528
Emission factors of substituted fuels	t_{CO2eq}/TJ_{SNG}	*2020:* 67 *2030:* 59 *2050:* 56	56
Oxygen demand potential	kg_{O2}/t_{cement} or kg_{O2}/t_{steel}	162–228	40–79

(Rosenfeld et al., 2020; Medved et al., 2021). Additionally, natural gas is still required for post-processing within integrated steelworks such as hot and cold rolling. These demands could be substituted by SNG from the methanation process in the mid-term while capturing and utilizing carbon emissions. Additionally, high-temperature electrolysis would already be established for a long-term transition to hydrogen-based direct reduction.

The scenario evaluated herein presumes the integration of the power-to-gas plant into a conventional integrated steel plant implementing blast furnace (BF) and basic oxygen furnace (BOF) according to a previous study (Rosenfeld et al., 2020). The natural gas demand in this configuration is ~528 kWh per ton of crude steel. Depending on the individual configuration of the steelwork, additional utilization paths for SNG may be available within the steelwork.

To provide oxygen for the steelmaking process for use in the converter in the conventional routes or in the subsequent electric arc furnace (EAF) process in the DRI route, integrated steelworks possess a dedicated oxygen production plant for cryogenic air separation (CAS). This oxygen demand can be partially supplied by the integrated electrolysis process, thus reducing the operational costs of the overall steelmaking process. According to a previous study (Kirschen et al., 2011), the oxygen demand for DRI-based steel production is 28 m^3/t_{steel}, while it is ~55 m^3/t_{steel} for the BOF route (Bieda, 2012).

Oxygen Supply

In addition to the main product hydrogen, the co-electrolysis process produces oxygen as a by-product (see Equations 1 and 2). Although it is available as a separate stream from water and carbon dioxide splitting, it is typically emitted to the atmosphere without further use. Based on stoichiometric reaction and full conversion, 2 mol oxygen per mol methane (4 kg_{O2}/kg_{CH4}) are produced. Potentially higher amounts that are caused by intentional hyperstoichiometric hydrogen production as a means to ensure complete carbon conversion are neglected in relation to methane as the target product.

As previously mentioned, oxygen used for large-scale supply (>200 t/d) is conventionally produced by CAS units (Luo and Liu, 2018). However, separation from ambient air is conducted with high electric energy demands of ~250–300 kWh per ton O_2 (Wu et al., 2018). To evaluate the economic impact of by-product oxygen use in the investigated scenarios, the costs of conventional oxygen supply were assessed in comparison to those that have been previously published. The available data range from ~50 €/t_{O2} (Kuparinen and Vakkilainen, 2017; Rosenfeld et al., 2020) to 150 €/t_{O2} (Guilera et al., 2018). Consistent with other data within that range (Breyer et al., 2015; Parra et al., 2017), a reference price of 85 €/t_{O2} was used.

CO_2 Certificates and Capture Potentials

As previously mentioned, the investigated application scenarios provide significant amounts of process-related CO_2 emissions. For cement production, the average emission of 0.54 t_{CO2}/t_{cement} with up to 65% from mineral origin (cf. section Cement Production) results in ~350 kg of non-fuel-related CO_2 emissions per ton cement. According to previous reports (Bains et al., 2017; Rodin et al., 2020), the appropriate capture costs for carbon emissions in this industrial sector are 22–35 €/t_{CO2}, with a capture efficiency of up to 90%. However, this range is related to post-combustion capture and lower CO_2 content in the flue gas compared to that of the oxyfuel process. Thus, for the consideration of an oxyfuel scenario, the input costs of CO_2 for the power-to-gas process are presumed to be at the lower end of this range. The fuel substituted in the cement scenario represents a mix of fossil fuels (coal, fuel oil, natural gas) and refuse-derived fuels. Based on the sustainability reports from the Austrian cement industry association (VÖZ) and common emission factors, the average emissions relate to ~67 t CO_2 per TJ of fuel used today (VÖZ, 2015, 2017; Jurich, 2016). Presuming that the use of coal and fuel oil will be omitted predominantly to achieve future emission targets, and the used refuse-derived fuels reach emission levels in the range of natural gas (cf. VÖZ, 2015), and the average emissions of the fuels to be substituted by the produced SNG are expected to be equivalent to the emission factors of natural gas at 56 t_{CO2}/TJ (Jurich, 2016).

According to a previous report (IEA, 2020), the current average direct CO_2 emissions relate to 1.4 t per ton of crude steel. Although the majority of these carbon emissions originate from the use of coke in the BF and BOF routes, production by natural gas-based DRI-EAF still results in direct emissions of ~1.0 t_{CO2}/t_{steel}. However, the SNG produced by the investigated power-to-gas system is intended to primarily substitute natural gas that is used energetically or as a process gas for finishing or use in the DRI route. Thus, the emission factors for the substituted fuels are related to those of natural gas at 56 t_{CO2}/TJ (Jurich, 2016). Referring to a previous report (Rodin et al., 2020), the capture costs for CO_2 in iron and steel production would be in the range of 19–83 €/t_{CO2} at capture rates of up to 90% without further specification of the underlying steel production process. As a further specification of the steel plant is not intended in the present study, an average value of 38 €/t_{CO2} is presumed to be the input cost for CO_2 in the techno-economic evaluation.

In comparison to the conventional processes (for cement and steel production), the implementation of a closed carbon circle is presumed to reduce the direct emissions generated by the substituted fossil fuel. Therefore, the costs for emission-related certificates according to the European emission trading system (EU ETS) are reduced. The avoidance of CO_2 certificate costs is a major benefit of the power-to-gas concept beyond the conventional processes and is thus considered as a reduction of SNG generation costs in the techno-economic assessment. Currently, CO_2 certificates are traded at a price of ∼25 €/t_{CO2} (EEX, 2021). However, these are expected to increase significantly in the future as a regulatory measure to effectively reduce industrial carbon emissions.

Learning and Scaling Effects

To allow for a projection on cost developments for future implementations up to the year 2050, an appropriate cost reduction effect in regard to the capital expenditures (CAPEX) of the power-to-gas technologies was considered. Therefore, a component-based approach was used to evaluate the technological learning effects based on well-known technologies and processes. This model was previously developed and applied to the most common electrolysis and methanation technologies (Böhm et al., 2020). The details of the mathematical description can be found in a previous report (Böhm et al., 2019).

$$C\left(X_t\right) = \sum_{i=1}^{m} \left\{ C_{0i} \cdot \prod_{j_i=1}^{n_i} \left[\left(\frac{P_{0j_i}}{P_{tj_i}} \right)^{ex_j} \right] \left(\frac{X_t}{X_0} \right)^{-r_i} \right\} \tag{4}$$

with:

C_{0i} … costs of component i at time $t = 0$
$C\left(X_t\right)$ … total costs at time t
X_0 … cumulative number of productions at time $t = 0$
X_t … cumulative number of productions at time t
r_i … learning parameter for component i (where $lr = 1 - 2^{-r}$)
P_{0j_i} … initial value of property P_j of component i at time $t = 0$
P_{tj_i} … value of property P_j of component i at time t
ex_j … influence exponent for property P_j

In the present study, the learning curve models evaluated in a previous article (Böhm et al., 2020) for SOEC and catalytic methanation were used. Though these two models use different references for their specific CAPEX values, the electrolysis model refers to the electric input power and the methanation model refers to the power of the product gas. As the power-to-gas system considered herein is assessed as a coherent system, the reference was harmonized to the electric input power of electrolysis. Thus, a potential change in efficiency exerts an impact on the conversion of the methanation learning curve. Therefore, an additional "Learning Property" (Böhm et al., 2019) was introduced for the methanation portion that represents the development of the electrolysis efficiency in relation to the cumulative production development:

$$P_t = P_0 \left(\frac{X_t}{X_0} \right)^{-r_p} \tag{5}$$

The reference value P_0 and the learning rate $lr = 1 - 2^{-r_p}$ were evaluated based on the overall electric efficiency of the power-to-gas system of 79% today and on a presumed development to 82.5% until 2050. This resulted in a learning rate for the property of $lr = -0.003$. The influence exponent in Equation (4) is $ex = 1$ (linear dependency).

Based on the cost structures defined in Böhm et al. (2020) for SOEC and methanation that were both confirmed to be widely applicable to the integrated co-SOEC and methanation approach used in the HydroMetha project (FFG, 2018), the CAPEX of the investigated power-to-gas system was evaluated for different scales ranging from 1,000 to 10,000 kW_{el}. The resulting CAPEX characteristics in relation to the plant scale and the time of implementation are shown in **Figure 3**. In this context, it should be noted that these learning effects are not directly related to the year of installation but rather to the actual development of cumulative production. Therefore, the given year dates rather have a designative character to represent the impact of CAPEX reduction potentials than to an actual time value.

Electricity Supply

According to the chosen scenarios that represent energy-intensive industrial use cases, a high load of the application throughout the year as assessed by a high number of annual full load hours (FLH) is presumable. Thus, a grid-based power supply could be considered as the most practical source of electric input with respect to the base load operation, costs, and security of supply. However, as power-to-gas systems are often considered as energy storage and load balancing options, the applicability of the system in combination with volatile renewable energy sources (RES) was evaluated. To achieve this, three different scenarios for power supply were investigated and included *(i) direct supply from wind parks*, *(ii) hybrid supply from wind and grid*, and *(iii) spot market grid supply*.

Direct Supply From Wind Parks

To evaluate the suitability of the power-to-gas system for grid-supportive operation in the context of peak load operation from RES, a direct supply from a representative wind park was investigated. Therefore, the production of an existing Austrian wind park possessing a nominal capacity of 21.35 MW (VERBUND AG, 2021) was simulated based on meteorological data using www.renewables.ninja (Pfenninger and Staffell, 2016; Staffell and Pfenninger, 2016). The plant was chosen due to its proximity to the Eastern Austrian industry regions and to several biomethane plants that can be utilized as potential additional sources for biogenic CO_2 (European Biomethane Map, 2020). Furthermore, the power plant size is at an appropriate scale for the investigated electrolyzer capacity range of 1–10 MW. The

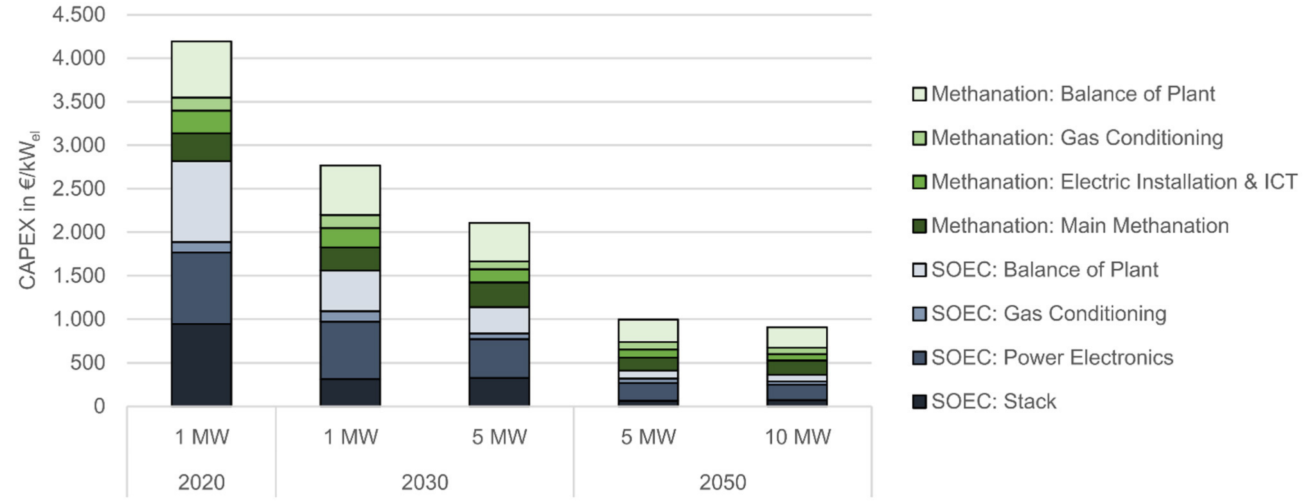

FIGURE 3 | CAPEX development of the power-to-gas system due to technological learning and scaling effects.

TABLE 2 | Direct supply operation parameters.

Parameter	Unit	Value
Nom. capacity wind park	MW	21.35
Min. load electrolysis	% of nom. power	20
Hot-standby demand	% of nom. power	5
Min. time of operation	hours	2
Min. time for shutdown	hours	30

analyses are based on a full year of data to exclude seasonal effects, and we refer to the year 2019 as the latest available dataset.

Depending on its scale, the power-to-gas system is intended to be operated with an appropriate peak load of the electricity produced from the wind park. According to the co-SOEC characteristics, minimum operation times and standby demands were considered (cf. **Table 2**). Hence, co-electrolysis is started up when the intended peak load exceeds the minimum load for at least the minimum operation time. When there is no appropriate peak load supply available from the wind power plant, the electrolyzer is set to hot-standby mode or is turned off if no operation is expected for a longer time period. The parameter values were chosen with respect to the general characteristics of high-temperature electrolysis according to cold and warm start times and flexibility (Buttler and Spliethoff, 2018; Smolinka et al., 2018). The electricity demand for the hot-standby mode is drawn from the grid at the spot market price according to the appropriate EXAA data (EXAA, 2019). **Figure 4A** presents the resulting electricity supply profile for the 1st month of the simulation year.

Hybrid Supply From Wind and Grid

To extend the annual time of operation for the electrolyzer, to reduce CAPEX-related SNG production costs but also allow for operation in an industrial context, a hybrid electricity supply is considered. In addition to the direct wind supply, the system is operated using electricity purchased from the spot market. Depending on the requested number of FLH, hours with the lowest prices in the year are preferred while also accounting for co-SOEC characteristics as described in the previous section. First, the times at which the system is already in operation through direct supply are prioritized until the nominal load is reached. Then, the cost limit for electricity from the spot market is increased continuously until the requested number of FLH is reached. The resulting electricity supply profile mix for the power-to-gas system is presented in **Figure 4B** for the first month of the simulation year.

Spot Market Grid Supply

The system is operated using electricity from the spot market with preference for hours with the lowest prices of the year while also accounting for co-SOEC characteristics as per section Direct Supply From Wind Parks. For consistency with the other scenarios, the historical EXAA spot market data for the reference year 2019 was used (EXAA, 2019).

Although EXAA also offers designated green electricity at the spot market price, the data demonstrate that there is apparently no trade based on it and that periodically recurring prices are not market-driven. Thus, even though the use of renewable electricity was preferred, the present analysis refers to gray electricity prices to evaluate the influence of spot market prices.

Techno-Economic Evaluation

The techno-economic assessment in this study primarily analyzes the specific production costs for SNG from the power-to-gas plant arising in the individual implementation scenarios. Therefore, the energy output-related *levelized costs of product* (LCoP) were calculated according to Böhm et al. (2020). The calculation is based on the LCoE approach that is often used to evaluate the costs of electricity production (de Visser and Held, 2014; IEA et al., 2020).

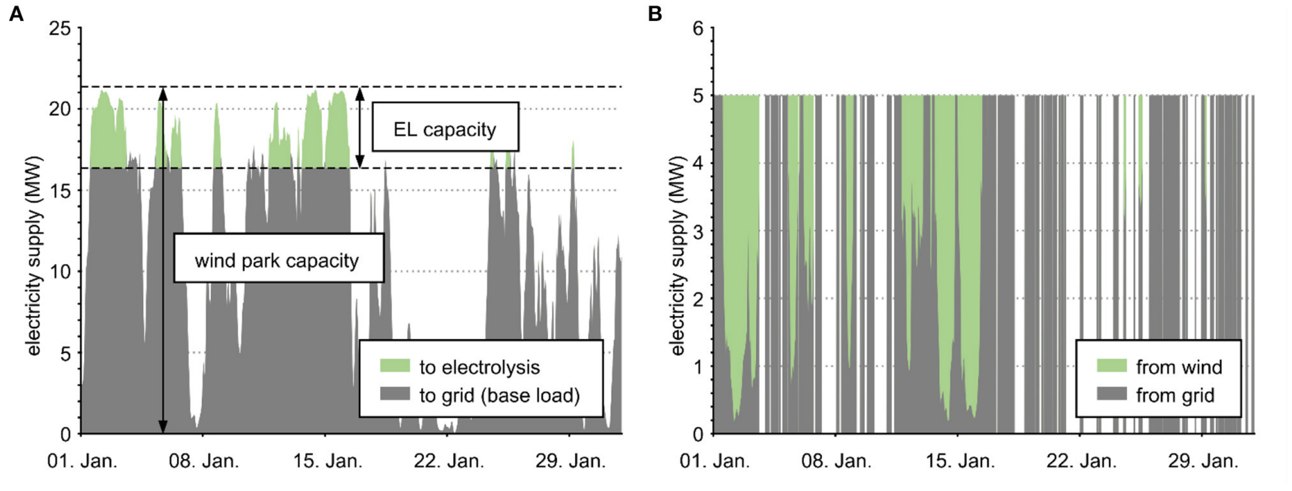

FIGURE 4 | Co-SOEC electricity supply profiles (one month). **(A)** peak load supply from wind park, **(B)** electrolyzer supply profile for hybrid supply operation.

$$LCoP = \frac{-A + \sum_i C_{var,i}}{E_{out}} \tag{6}$$

The assessment considers a full year of operation in which the total annual costs are calculated using the annuity method (VDI, 2012). In Equation (6), A represents the annuity of the fixed total annual payments such as capital- and operation-related (e.g., insurance and maintenance) costs. C_{var} represents variable costs and revenues that are dependent upon the corresponding material and energy streams and thus related to the annual time of operation and potential variable load conditions such as part-load efficiencies. These include the demand-related costs for resources, such as electricity, CO_2, and water, and they also include by-product sales. E_{out} is the total annual energy output of the power-to-gas plant corresponding to the SNG produced.

Capital-related annuity considers the investment costs of the main equipment of a power-to-gas plant. Additionally, it includes future costs for the expected replacement of individual components within the set observation period. It is defined as:

$$A_C = (I_0 + I_1 + \cdots + I_n - R) \cdot a \tag{7}$$

where I_0 is the initial investment cost, and $I_1 \ldots I_n$ represents the first to nth replacement investment. R represents the residual value of the plant at the end of the observation period. The cash values of the replacement investments and residual value are calculated considering the interest rate factor $q = 1 + i$ (where i is the interest rate) and the presumed deprecation period T_N:

$$I_n = \frac{I_0}{q^{n \cdot T_N}} \tag{8}$$

$$R = I_0 \cdot \frac{(n+1) \cdot T_N - T}{T_N \cdot q^T} \tag{9}$$

The annuity factor a in Equation (7) is calculated based on the interest rate factor and observation period:

$$a = \frac{q^T \cdot (q-1)}{q^T - 1} \tag{10}$$

As only investment costs for the main equipment of the power-to-gas plant are directly included in the capital-related annuity, additional costs that arise such as those for engineering, construction, and commissioning of the plant were considered in the form of appropriate overhead factors (Peters and Timmerhaus, 1991; Perry et al., 1999; Sinnott and Towler, 2020) (see **Appendix Section 1.2**).

The annuity of operation-related costs (maintenance and insurance) was considered as a fixed factor related to the initial investment I_0 and was likewise provided with the annuity factor a. The actual values are listed in **Table 3** along with the other calculation parameters relevant for the techno-economic assessment.

RESULTS AND DISCUSSION

Optimization of Electricity Supply Costs

As electric power is the main input energy for the power-to-gas system, the relationship regarding this power type in the investigated application scenarios is analyzed independently of the other plant costs. This includes the costs of the direct supply of peak electricity production from local wind parks. Operation at peak production is considered preferable in terms of grid service and allows the electricity supply to the grid to be more base-load oriented while preventing curtailment or shutdown of electricity production. Thus, the supply costs for the electrolyzer are expected to be lower than the average generation costs from onshore wind. To estimate the potential electricity supply costs for this peak load operation, the appropriate spot market prices for the times of available wind power supply were evaluated.

TABLE 3 | Calculation parameters for the techno-economic assessment.

	Unit	2020	2030	2050	References
General					
Interest rate	%	4.0	4.0	4.0	Steinmüller et al., 2014
Deprecation period	years	20	20	20	Steinmüller et al., 2014
Power-to-gas system					
Efficiency (LHV)	$\%_{LHV,SNG}$	79	81	82.5	FFG, 2018, Hydrometha project goals
LHV of product gas	kWh/m^3	9,319	9,319	9,319	Krammer et al., 2021
OPEX	% of CAPEX	5	4	2	FCH 2 JU, 2018
Electricity demand auxiliaries	% of nom. power	1	1	1	own assumption
Water supply costs	€/$m^3_{H_2O}$	1.15	1.15	1.15	Böhm et al., 2020
Lifetime electrolysis stack	hours	20,000	40,000	90,000	Smolinka et al., 2018
Lifetime methanation catalyst	hours	25,000	60,000	150,000	based on Grond et al., 2013 and own assumptions
Lifetime BoP	years	15	20	20	based on FCH 2 JU, 2018 and own assumptions
Standby operation					
Energy demand hot-standby	% of nom. power	5	5	5	Smolinka et al., 2018
Start-up time warm	Minutes	10	5	1	Smolinka et al., 2018
Start-up time cold	minutes	600	180	30	Smolinka et al., 2018
Additional expenditures					
Insurance	% of CAPEX	0.5	0.5	0.5	Steinmüller et al., 2014
Administration	% of CAPEX	2	2	2	Steinmüller et al., 2014

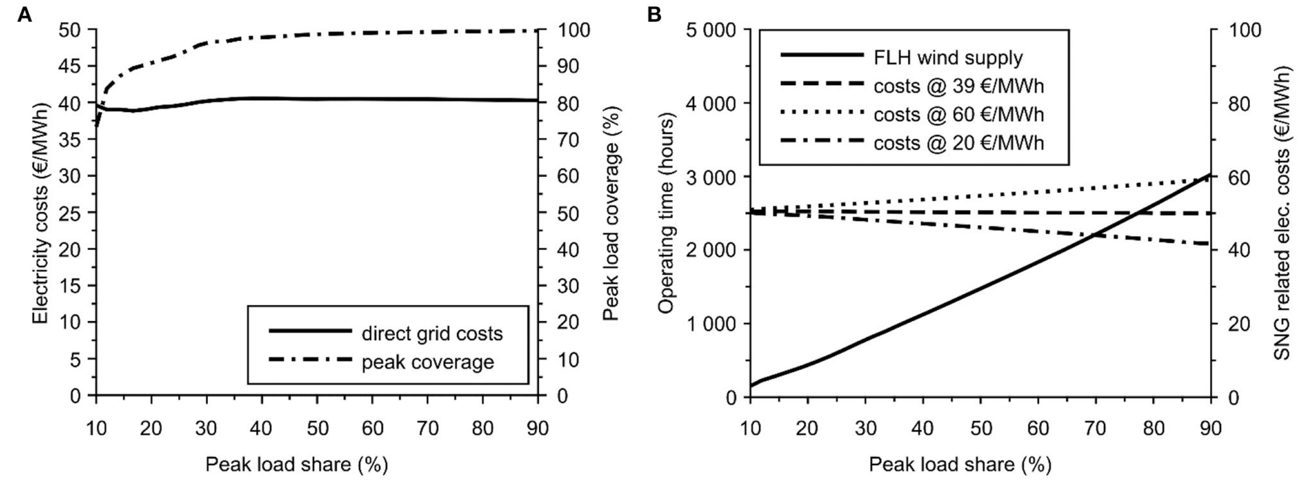

FIGURE 5 | Influence of the electrolyzer to wind park capacity ratio (peak load share) on **(A)** peak coverage and supply costs and **(B)** on full load hours and production costs.

The analyses revealed that these average costs for an alternative direct grid supply would be no lower than 38.85 €/MWh for the investigated production site and the Austrian spot market (cf. **Figure 5A**). Although this value is lower than that of the average annual spot market prices of 40.16 €/MWh in 2019 (EXAA, 2019), it is still higher than 44% of the quarter-hourly values. Therefore, operation at peak load wind production does not necessarily correlate with the times of lowest spot market prices.

As the definition of peak electricity production in the context of this analysis is dependent upon the electrolyzer capacity, the impact of this parameter on peak load coverage, costs, and the available FLH of electrolyzer operation was analyzed. Depending on the value of the ratio of electrolyzer capacity to wind park capacity, which is referred to as peak load share, this analysis has shown that at a ratio of 19% (4 MW electrolyzer in the case of the 21 MW wind park), up to 90% of the resulting production peaks could be utilized by the electrolysis (**Figure 5A**). Additionally, with higher peak load share up to 3000 FLH per year could be reached from the direct supply operation (**Figure 5B**). Therefore, even under the limitations of

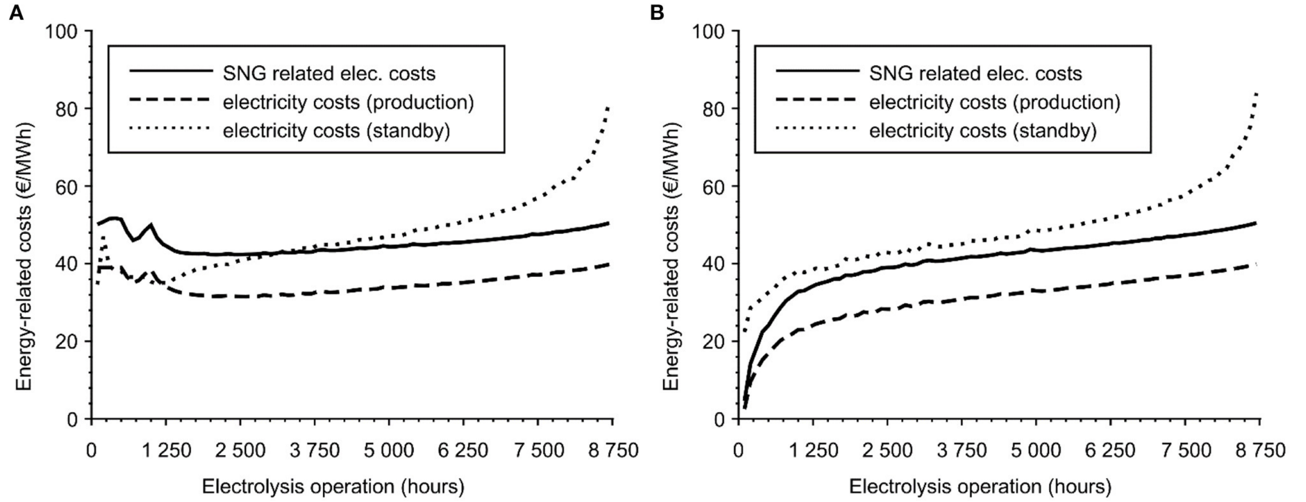

FIGURE 6 | Electricity supply costs and electricity-related production costs for **(A)** the hybrid electricity supply and **(B)** the grid-only electricity supply.

the co-SOEC in terms of load flexibility, this technology offers significant potential for operation with volatile renewable energy sources for providing both grid services and dedicated supply.

Based on these results, the electricity supply costs for direct supply from wind were presumed to be 39 €/MWh for the subsequent calculations so that they did not compete with grid supply costs in the hybrid supply scenario. The resulting electricity supply costs for production and standby operations were evaluated in relation to the annual full load hours of operation. In regard to the investigated electricity supply scenarios (cf. section Electricity Supply), the scenario for direct wind supply is identical to the hybrid supply scenario up to the maximum amount of available FLH as shown in **Figure 5B** and is therefore not discussed separately.

As presented in **Figure 6A** for the hybrid supply scenario, the electricity costs related to SNG production are discontinuous for low annual FLH. Starting with constant supply costs for production but increasing standby costs, electricity-related production costs become larger until the maximum amount of FLH for direct wind supply is reached (∼550 FLH for the 5 MW electrolysis vs. 21 MW wind park case). Subsequently, when maximizing the system load at times that the power-to-gas system is already in operation, the costs initially decrease before increasing according to the bound grid electricity costs. After this, the supply costs again decrease with increasing annual FLH due to prioritization of hours with the lowest spot market prices, and they ultimately reach a minimum of 42.3 €/MWh$_{SNG}$ at 2,100 FLH for the 5 MW electrolysis. Due to the higher standby demand, the minimum for a 10 MW electrolysis is slightly higher at 45.8 €/MWh$_{SNG}$ and 4,700 FLH. Considering the cost-optimized operation with electricity supply only from the spot market price (**Figure 6B**), production costs continuously increase with an increase in annual full load hours. Even though electricity costs for hot-standby operation increase significantly (up to 84 €/MWh), this is compensated for by the decreasing time of operation in this mode.

Evaluating the results in relation to the implementation in an industrial context, the most relevant range is from 7,500 FLH and above. In this range, both supply scenarios (hybrid and grid-only) result in similar electricity-related production costs of 47.4–50.5 € per MWh SNG produced. Additionally, the dependency on the peak load share (ratio of electrolyzer to wind plant capacity) is insignificant and within the range of 50.0–50.5 €/MWh (90%—10%) for the set wind supply costs. Thus, without considering the significant incentives for providing grid services by peak load coverage of wind power plants, an operation has no significant impact on the electricity supply costs in terms of industrial applications. Additionally, for low peak load shares, the impact of direct wind supply costs was less significant (cf. **Figure 5B**). However, these direct supply costs become more relevant for higher ratios, and this, in turn, reduces the available base load produced for the grid and thus counteracts the idea of peak-shaving. Therefore, a grid-supportive implementation of an industrial power-to-gas plant can be achieved in a manner that results in low economic impacts on electricity producers and consumers.

SNG Generation Costs

The evaluation of the resulting SNG generation costs for the two investigated industries (**Figure 7**) revealed that the application scenario itself exerts only a minor impact on the product costs. Both scenarios reach product cost levels of 20–26 c€/kWh for current state implementations with expected cost reduction potentials of 50 to 80% for future installations. The slightly lower product costs for the current state (2020) steel industry scenario are primarily related to the higher number of annual full load hours compared to the cement industry (8,500 vs. 8,000). However, this advantage almost completely disappears for mid- and long-term implementation due to the significant reduction in fixed cost factors (CAPEX and OPEX) that occurs as a result of projected technological learning and scaling effects. Accounting for up to 59% of the overall generation costs for a

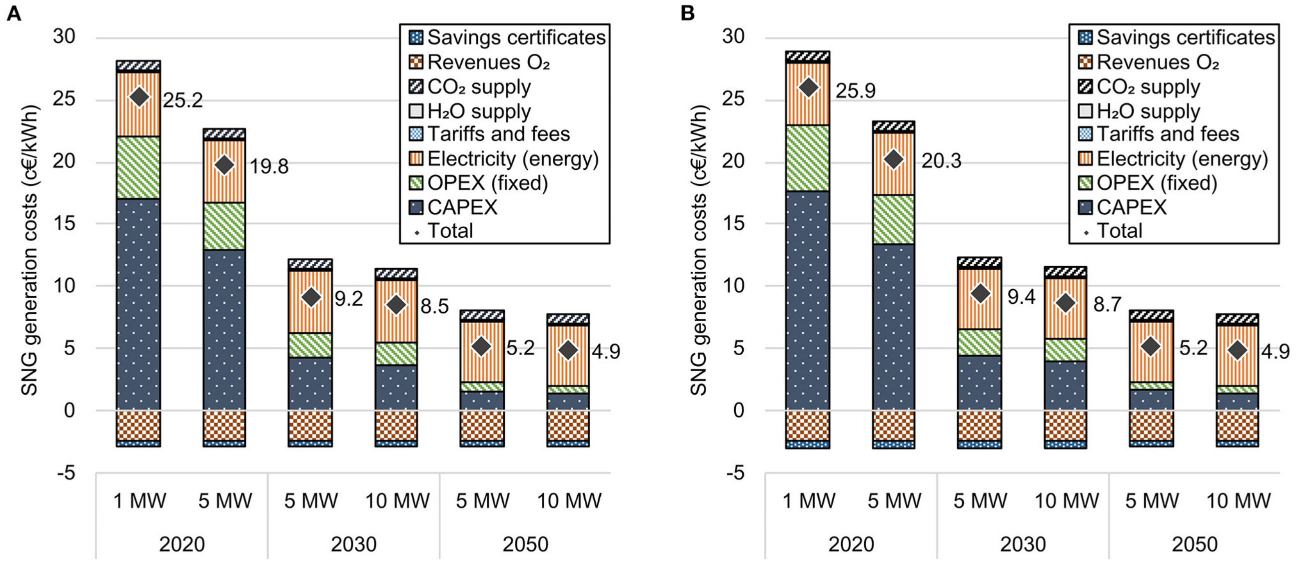

FIGURE 7 | SNG generation costs for different plant capacities and years of implementation. **(A)** steel industry scenario, **(B)** cement industry scenario.

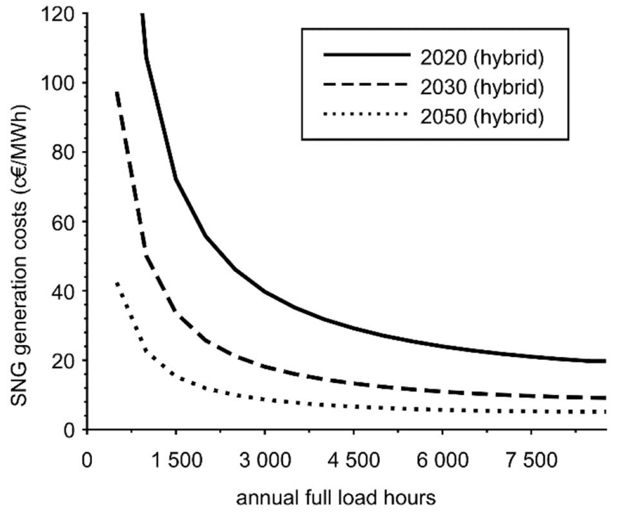

FIGURE 8 | SNG generation costs in dependence of annual FLH of operation for the 5 MW reference plant.

1 MW plant today, this cost share is expected to be reduced to only 16% when assuming a 10 MW plant in 2050. These cost reductions are primarily induced by technological learning according to the electrolysis uptake projected in a previous report (Böhm et al., 2020). Additionally, high-temperature electrolysis systems are expected to exhibit a stronger cost dependency on the system scale than are low-temperature technologies. Hence, the implementation of large-scale systems up to 100 MW electrical capacity involves additional cost reduction potential for industrial applications (Böhm et al., 2020). Though, even without these additional scaling effects the higher efficiency of the thermally integrated co-electrolysis approach reveals significant economic benefits over projected large-scale implementations (50–100 MW) using conventional electrolysis and methanation in combination with PV or wind power (Böhm et al., 2020). Also, in comparison with assessments for PtM in single MW scales, as analyzed by Gorre et al. (2019) the economic advantage of higher system efficiencies beyond yet higher CAPEX is apparent as long as electricity prices are not significantly below current levels.

Based on the current certificate cost for CO_2 emissions of 25 €/t_{CO2}, the impact of avoiding the buying or selling of excess allowances is relatively low. Even for the cement industry scenario where the emission factors of the substituted fuels are ~19% higher than those of the natural gas substituted for the steel industry, the savings only compensate for <3% of the overall generation costs in the 2020 scenario. With decreasing CAPEX, this becomes more relevant in the long term and can account for up to 6% of the SNG generation costs. However, to generate a significant impact, certificate costs would need to increase significantly (see section SNG Generation Costs).

Although the implementation in an industrial context generally suggests an operation of the power-to-gas plant at a high annual load, the lower electricity supply costs at a lower number of annual FLH may appear beneficial from a cost perspective. However, as presented in **Figure 8**, the impact of high fixed costs (CAPEX) for the 5 MW reference implementation out-values this supply cost advantage for both the hybrid and grid-only supply. Considering the presumption of constant electricity costs for the entire period until 2050, the cost curve flattens at >7,000 FLH with potential cost advantages in that range for large-scale implementations (> 100 MW). However, in relation to the decreasing cost share of CAPEX (cf. **Figure 7**), this can be considered insignificant.

Impact of By-Product Oxygen Use

As shown in **Figure 7**, the savings due to the utilization of by-product oxygen exert a significant impact on the overall SNG generation costs and can substitute for 9–30% of the given expenditures. Based on gross reactions (cf. Equation 1 and 2) and the LHV of the product gas, ∼282 kg of O_2 per MWh of SNG were produced by the co-electrolysis process. This relates to 223–233 kg_{O2}/MWh of input power depending on the process efficiencies.

In relation to the average oxygen demand of an oxyfuel cement plant, the required electrolysis capacity would relate to 91–128 kW/kt_{cement} (for 8,000 FLH per year) or ∼434–610 MW for a complete adaption of the Austrian cement production (4.8 Mt in 2016 acc. to VÖZ, 2017). In contrast, fuel substitution requires a power-to-gas plant capacity that ranges from ∼25 kW/kt_{cement} (fossil fuels only) to 119 kW/kt_{cement} (including alternative fuels). Therefore, depending on the substituted fuel input and the oxygen requirement of the oxyfuel process, the savings generated from by-product oxygen utilization are related to external sales instead of direct use (up to 0.56 c€/kWh_{SNG}).

The oxygen demand in the investigated steel plant scenario depends upon the underlying steel production process and ranges from 40 kg_{O2}/t_{steel} for DRI to 78 kg_{O2}/t_{steel} for conventional BOF routes. Considering the primary use of SNG over hydrogen production to extend the lifetime of the BOF route, an electrolyzer capacity of ∼39–41 kW/t_{steel} would be required to meet that demand (for 8,500 FLH per year). In contrast, the required power-to-gas capacity to substitute the natural gas demand of the steel plant is 75–79 kW/t_{steel}. Therefore, for a complete substitution of the natural gas input, the by-product oxygen would exceed the actual demands of the investigated steel plant. Consequently, the requirement of external oxygen production becomes obsolete, and external sales of the by-product oxygen are necessary to completely enable the considered revenues.

CO_2 Mitigation and Resulting Certificate Savings

As discussed previously, the impact of CO_2 certificate savings on the overall SNG generation costs is insignificantly low under the current conditions. Under these conditions, economic competitiveness with fossil fuels cannot be achieved. Compared to the EU-28 average price of natural gas for non-household consumers that was ∼3.27 c€/kWh in 2019 (Eurostat, 2020), in future evaluations the calculated product costs are 1.5–3 times as high, even if all revenues and savings are included. However, with increasing carbon prices, the integrated generation of SNG as a carbon neutral fuel in the context of a closed carbon circle may provide significant benefits over the consumption of taxed fossil fuels. Presuming a CO_2 price of 150 €/t, the effective generation costs of SNG for the given applications would fall below the current price for natural gas for the 2050 (5 MW) scenario. For a carbon price of 330 €/t that was suggested to be necessary by the mid-century to achieve rapid decarbonization (Rockström et al., 2017, 400$/t), a similar level of competitiveness could already be reached at the 2030 CAPEX levels. From a long-term perspective (2050 scenario), this level of carbon pricing could even lead to negative effective generation costs, as the appropriate savings (6.6 c€/kWh) and revenues from by-product sales exceed the actual production costs.

Sensitivity Analysis

To evaluate the impact of the individual calculation parameters used in the techno-economic assessment, a sensitivity analysis was performed. In **Figure 9** the variation of the resulting generation costs is presented for a parameter variation of ±25% from the appropriate reference value for the implementation scenarios 2020 and 2050. The most significant relative impact is electric efficiency, particularly if the projected values cannot be provided. Thus, a reduction of 25%, which represents an absolute value of 59 and 62% respectively according to the scenario, the SNG generation costs are calculated to increase by up to 45%. Depending on the dominance in the overall cost structure, the impacts of CAPEX and electricity costs are also significant in the sensitivity analysis. Therefore, the impact of potentially increasing electricity costs can be decisive for the economic viability of future implementations. The data presented in **Figure 9** reveal that the impact of supply costs on SNG generation costs typically increases with a decrease in the overall costs. In contrast, the sensitivity to durability and replacement intervals decrease according to the expected development of component and system lifetimes that can result in longer operation periods.

Although most calculation parameters exert a linear impact on the resulting generation costs, lifetimes and observation period, as well as the system efficiency exhibit a non-linear dependency. As illustrated in **Figure 10**, the impact of the system and component lifetimes increases with decreasing values as the number of replacement investments increases. The same scenario applies to the observation period that defines the annuity of the CAPEX according to Equation (10). However, the impact of these parameters decreases at higher values. **Figure 10** again illustrates the high impact of the actual efficiency on the resulting costs that further increase at lower values. For positive variation, the absolute value was limited to $100\%_{el,LHV}$.

Another relevant impact on future installations can occur from tariffs and fees that must be paid for the operation of power-to-gas plants and their integration into public energy networks (power and gas grids). For the given calculations, Austrian conditions according to the regulatory framework in place by the end of 2020 were considered. Although this currently results in negligible cost factors (approx. 0.2 c€/kWh), individual charges, taxes, and levies are still legally unclear and can change in future regulations. Additionally, they differ between countries. Thus, this cost factor should be monitored in future studies.

CONCLUSION

The investigations performed in this study evaluate the current and future economic competitiveness of high-performance power-to-gas systems in energy-intensive industries. Therefore, highest efficiencies of power-to-gas systems are required. This requirement is met by combining high-temperature co-electrolysis with catalytic methanation in a thermally integrated complete system. The proposed implementation scenarios aim

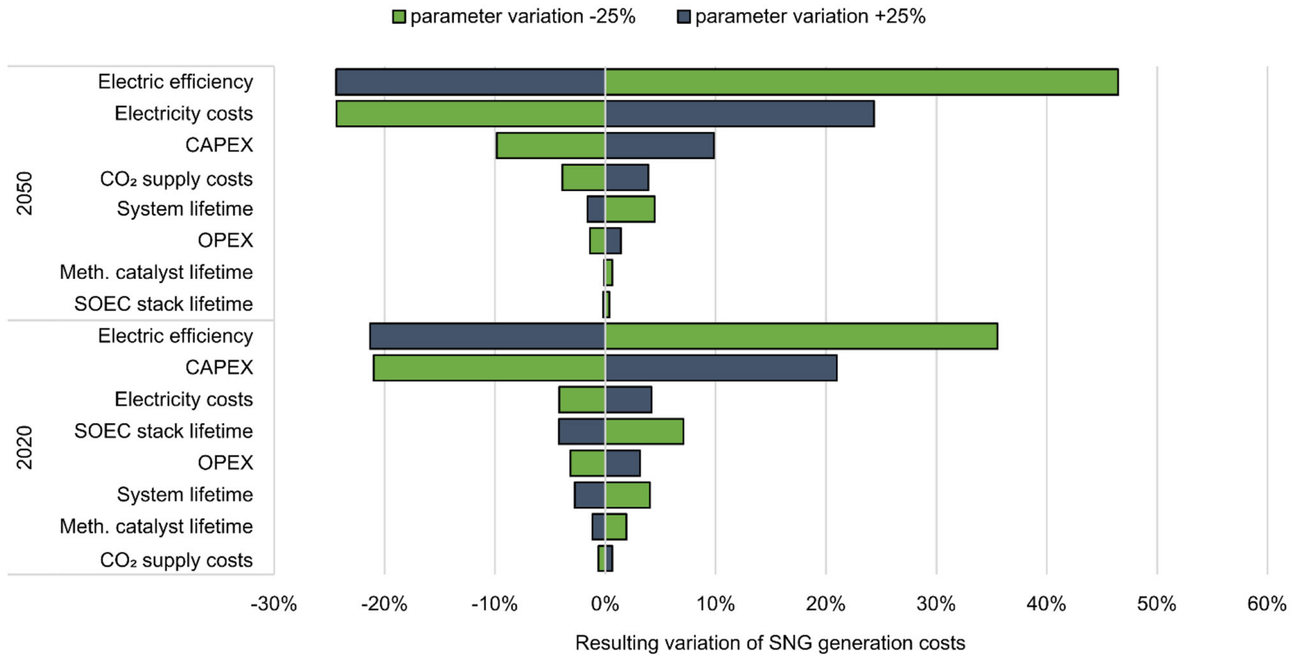

FIGURE 9 | Impact of calculation parameter variation on the resulting SNG generation costs.

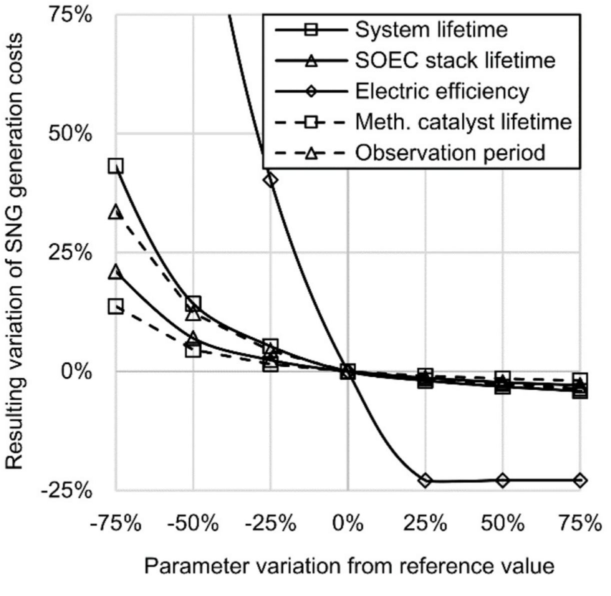

FIGURE 10 | Sensitivity analysis of non-linear impact parameters for the 2030 reference case.

to allow for the creation of closed carbon cycles for industrial sectors and plants that are expected to still rely on non-avoidable process-related carbon emissions in their mid- and long-term transition to renewable processes. In this context, our analyses have revealed that the production costs for the given system performances will reach values of 15 c€/kWh and below when appropriate learning curve and scaling effects occur for this low TRL technology. The effective generation costs of the primary product can be further reduced by the utilization of by-product oxygen, either by direct use in related process streams substituting dedicated production capacities or by external sales. However, despite accounting for these side effects, effective generation costs are expected to be two to three times as high as the current industrial costs for natural gas, even in long-term scenarios. Therefore, effective pricing of carbon emissions must be established to allow these synthetic fuel substitutes to be competitive with fossil fuels. It has been shown that a carbon price of 150 €/t_{CO2} can enable beneficial business cases for large-scale implementations from a long-term perspective. Similarly, if carbon pricing is implemented as proposed with appropriate costs of 330 €/t_{CO2} and higher, cost parity with fossil fuels could be reached even earlier and prior to exhausting long-term scaling effects. Thus, even short- and mid-term implementations (e.g., 2025–2030) could benefit from long-term savings.

As another aspect of the integration of power-to-gas technology in future energy systems, the applicability of renewable electricity supply was investigated. As co-electrolysis, or solid-oxide electrolysis in general, is often considered to be inflexible to load changes, more dynamic electrolysis technologies (e.g., PEM) are preferred to provide grid services. However, simulations of direct coupling with wind power have shown that a grid-supportive operation is possible. Additionally, with direct supply costs of ∼39 €/MWh, the power-to-gas plant can be operated in a manner that is cost-effective compared to spot-market prices while providing economic alternatives for wind park operators to curtailment or shutdown at production peaks.

Although this study demonstrates the general potential of co-electrolysis-based power-to-gas plants in industrial applications, these effects are still dependent on the positive development of the technology beyond the current TRL. In this context, the technology has to compete with more mature and thus less cost-intense technologies. Thus, in addition to the achievement of set efficiency goals of the heat integration approach, this requires appropriate efforts in regard to upscaling both capacities and numbers over the next years in combination with the according R&D tasks. Furthermore, for the considered industrial applications implementations at single MW-scale will not suffice to meet actual demands for a fully decarbonized production. Hence, additional investigations will have to be performed for plant scales beyond 100 MW where a legitimate upscaling from current technology status cannot be done.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

HB conceived and designed the study, was responsible for data acquisition, calculations, and analysis, and wrote the first draft of the manuscript. ML and TK reviewed and edited the manuscript prior to submission. All authors contributed to manuscript revisions and read and approved the submitted version.

FUNDING

The contents included in this chapter are part of the national Austrian flagship project HYDROMETHA (contract # 864578) that has received funding from the Austrian Climate and Energy fund under the framework of the Energy Research Program.

ACKNOWLEDGMENTS

The support of the Energieinstitut an der Johannes Kepler Universität Linz and the Austrian Research Promotion Agency (FFG) is gratefully acknowledged.

SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/frsus.2021.726332/full#supplementary-material

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.