CRITICAL EVALUATION OF PROSPECTS AND LIMITATIONS OF STEEL CLEANNESS CHARACTERISATION USING AUTOMATED SEM/EDS ANALYSIS

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ABSTRACT

Automated SEM/EDS analysis has become state of the art for the characterisation of non-metallic inclusions. The present study focuses on the representativeness of this method, especially evaluating the influence of the analysed sample area on the obtained particle diameters and size distributions of non-metallic inclusions. Next to an experimental analysis comparing the results of different area sizes and intersection planes on a metallographic specimen, a geometric-statistic model was formulated estimating the error of area ratio as a function of inclusion content. Secondly, the significance of the method regarding the determination of the maximum inclusion diameter in the analysed sample is assessed. For this purpose, the theoretically defined values from the model are compared with the results from automated SEM/EDS analyses. Additionally, chemical extraction is used in order to get a three-dimensional view of the non-metallic inclusions distributed in the analysed sample and hence also evaluating the influence of intersection probability. The results showed that for the defined conditions the analysis of 100-200 mm^2 is sufficient in order to get a representative impression concerning the global cleanness in the sample. Regarding the maximum inclusion diameter the significance of automated SEM/EDS analyses is limited.

INTRODUCTION

The cleanness level has emerged to an important quality criterion for a wide field of steel applications. In order to ensure the reliable characterisation of non-metallic inclusions (NMIs), the constant optimisation of current analysing methods is essential. Out of the varied characterisation methods, the automated SEM/EDS analysis became state of the art [1-4] regarding the evaluation of micro cleanness primarily for research purposes, due to the extensive obtained information including data on inclusion size, their area fraction as well as the distribution and chemical composition of inclusions. Despite these apparent advantages, the significance of this method has to be considered critically concerning certain aspects. Next to the evident influence of measurement settings [5], questions dealing with the correct classification of multiphase inclusions or matrix correction [6] have to be kept in mind.

However, the present paper only focuses on the morphological parameters gained out of automated SEM/EDS analysis. The method is based on a two-dimensional view of inclusions distributed in space. Even if this reduction of dimensions brings along a substantial facilitation for the measuring process – considering the fact that a particle is not cut at its maximum diameter necessarily – a metallographic specimen yields only an apparent distribution of the inclusion diameters [7]. Thus, in combination with the usually rather small measuring area the aspects of representativeness and reproducibility have to be considered. In this context, two decisive questions for two-dimensional inclusion analysis arise:

- Firstly, if the analysis of a defined sample area is adequate to give an impression of the mean inclusion size and the global cleanness (also involving the influence of smaller inclusions) of a whole volume.

- Secondly, whether the sample area is sufficient to determine large, however stochastically rare inclusions.

The present study focuses on the influence of the measured area size on the mean and the maximum detected inclusion diameter and also gives an indication for the representative measuring area for the defined conditions using a geometric-statistical model in combination with experimental measurements. Additionally, the significance of intersection probability on detected inclusion diameters is discussed.

STATISTICAL CONSIDERATIONS

One of the most questionable aspects dealing with automated SEM/EDS analysis is the comparable small measuring sample area. Based on the results of several analyses using a measuring area between 100 and 200 mm^2, often conclusions on the inclusion landscape in a whole steel ladle, cast product or finished product are drawn. Next to the classical question of a representative sample out of the entirety, also the transferability of results from to two-dimensional analysis to the whole volume has to be kept in mind; especially due to the fact that a planar metallographic specimen is necessary for automated SEM/EDS analysis. In order to get a better understanding of the latter aspect, a geometric-statistical model was formulated. This model allows the description of the representative sample area in dependence of sample volume and inclusion content:

A defined number of inclusions is distributed in a cube. Although inclusions can show very complex geometries in reality, the model is currently based on the assumption of spherical shapes only. As illustrated in **Figure 1a**, the individual spheres with the centre coordinates $S_i=(s_{x,i},s_{y,i},s_{z,i})$ are randomly distributed in the cube, with the diameters of the spheres d_i according to a lognormal distribution. The required input parameters are gained from experimental analyses and are dependent on the inclusion content. Consequently the input parameters are varying for every steel grade. For a detailed mathematical description of the whole model and the used input parameters it is referred to a previous publication [8].

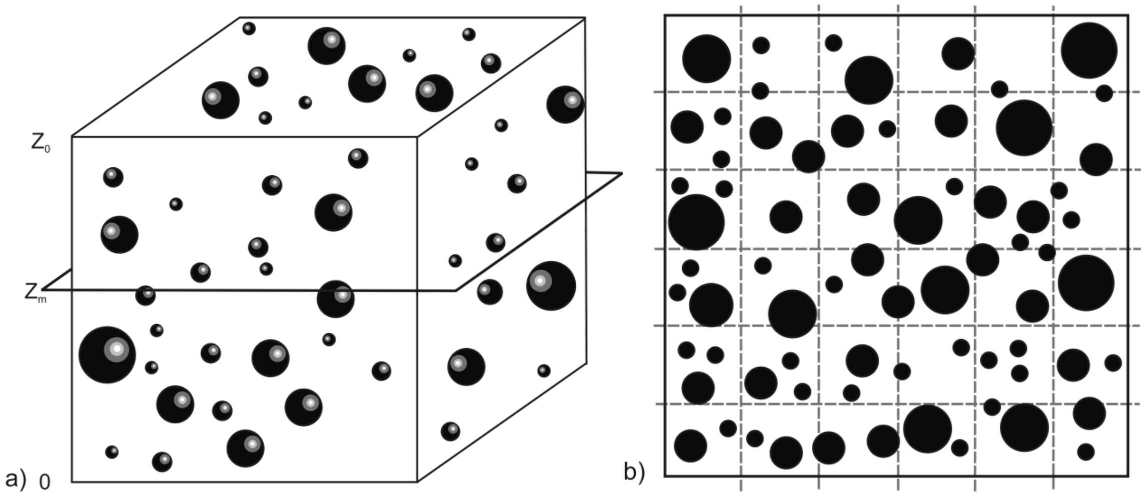

Figure 1: a) Schematic illustration of a cube with distributed spheres. b) Schematic cut through the cube plane parallel to the cube base.

In a next step, the cube is numerically intersected m times with the intersection plane parallel to the cube base and the size of the obtained circles is determined. The position of the plane is termed z_m with $0<z_m<z_0$ where $x_0=y_0=z_0$ is the cube side length. The set of spheres cut by the plane m is schematically illustrated in **Figure 1b**. For the present case m = 5, therefore the maximum virtual analysed sample area is 2'000 mm² since each intersection plane has an area of 400 mm². Evidently this maximum sample area exceeds any practically employed experimental analysing area (typically < 400 mm²). Thus, the high ratio between analysed sample area and total sample volume ensures an unbiased, stochastic distribution of the particles.

Finally, by numerically limiting the single intersection plane, the influence of the minimisation of analysed sample area could be studied. The error in analysed sample area is finally given by

$$Error = \frac{\overline{A} - A_T}{A_T} 100\%$$

where A_T is the area of all intersected circles at the maximum intersection area of 2'000 mm².

Moreover, in difference to the results of the automated SEM/EDS analyses, there is no truncation limit for the circle diameter in the model. Hence, the sum of all intersected spheres equals entirety. In order to analyse the influence of this truncation, the truncation limit from the practical observations (1.1 µm) is also introduced numerically.

EXPERIMENTAL PROCEDURE

In automated SEM/EDS analysis, non-metallic inclusions are detected due to material contrast differences in the backscattered electron (BSE) image. As a result, information about the exact position of the inclusion on the analysed sample area, as well as the morphological data and its chemical composition is gained. The present work only focuses on the morphological parameters of inclusions and does not go into detail as far as the chemical composition is concerned. All investigated particles in the practical part of this study are Al_2O_3 inclusions. Consequently, the theoretical results are also only valid for the defined inclusion content.

As far as the morphological information is concerned, the most essential parameter is the so called Equivalent Circle Diameter (ECD). This is a calculated value resulting of the measured area of each particle. The relationship is defined as follows:

$$ECD = \sqrt{\frac{4A}{\pi}}$$

All measurements were carried out with a stainless steel sample which has been melted in an induction furnace at the Chair of Metallurgy. Its composition is shown in **Table 1**. Out of the cast steel ingot a cuboid with the dimensions 49x49x65 mm³ was formed at 1200 °C with a deformation degree of $\varphi = 0.7$.

Table 1: Chemical composition of the steel used in the experimental part.

%C	**%Cr**	**%Si**	**%Mn**	**%Mo**	**%V**
0.34	5.02	1.57	0.53	1.28	0.49

At the Chair of Metallurgy a Scanning Electron Microscope from FEI (Quanta 200 MK2) in combination with an EDS system from Oxford Instruments is used. As already mentioned beforehand, the correct and reasonable adjustment of experimental settings is indispensable for automated SEM/EDS analysis. **Table 2** summarises the standard settings which are used for measurements at the Chair of Metallurgy.

Table 2: Experimental settings.

Beam energy	15 keV
Working distance	10 mm
Resolution	1'024 px × 960 px
Magnification	600x
Minimum particle size	4 px
EDS evaluation time for one particle	3 s

For every analysis, the minimum number of pixels which is needed to identify a particle has to be defined. Usually a limit of 1.1 µm is used, achieving an acceptable compromise between measuring time and obtained results. Thus, in contrast to the calculations, where the whole size spectrum can be displayed, the truncation of data has to be considered when interpreting the measurement results. In order to analyse the representativeness of the measuring area for the used steel grade, two different experimental approaches were applied:

Case 1: The measuring area on the metallographic specimen was enlarged continuously starting with 50 mm^2 as shown in **Figure 2**, always using the identical SEM/EDS settings and a minimum particle diameter of 1.1 µm. In sum, three different measurements were performed, each including the exactly same measuring area of the previous analysis. No further preparation step was done between the single measurements. Out of this variation, the following aspects should be examined:

- Influence of measured area size on the mean and maximum ECD of detected particles;
- Reproducibility of results when measuring exactly the same sample area.

The latter has only been investigated for reasons of completeness, since an accordance is necessarily required if identical measurement settings are applied. A possible reason for differences in the number of detected particles is often a shift in the defined grey scale value during measurement. A chronological record of the grey scale value over the whole measuring time is therefore recommended in order to exclude this source of error. In the present case, superimposing the three reiterations for the results of 50 mm^2, leads to an accordance of 99 %. Consequently, variations resulting out of measurement influences are seen as negligible.

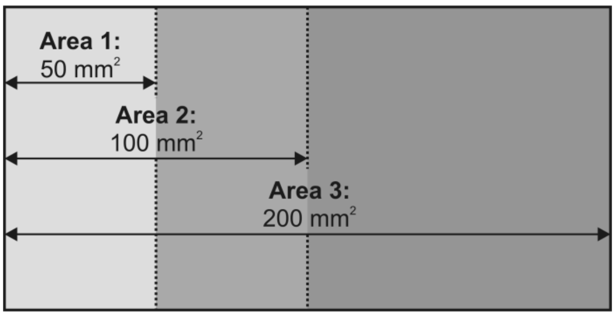

Figure 2: Schematic illustration of the arrangement of the different used measuring areas on one section of the specimen.

Case 2: Five superimposed sections of the metallographic specimen were analysed. For this purpose, at first an area of 200 mm^2 was measured on the polished metallographic specimen. Beforehand, the area was exactly defined with 6 Vickers marks (as shown in **Figure 3**). In a next step the specimen was polished again. In order to obtain a constant and comparable abrasion for all sections, new Vickers marks were set for every section. The diagonals of every hardness mark are measured and out of it the difference in depth of the Vickers mark is calculated. The Vickers marks were evaluated before and after polishing and hence the abrasion of each step could be defined. This procedure was repeated four times. For all sections the abrasion was determined with 12 µm ±0.71 µm. Out of this variation, the following aspects should be examined:

- Influence of the analysed cross section on the mean and the maximum ECD of detected particles.
- Effect of the only two-dimensional view on the detected morphology of large particles.

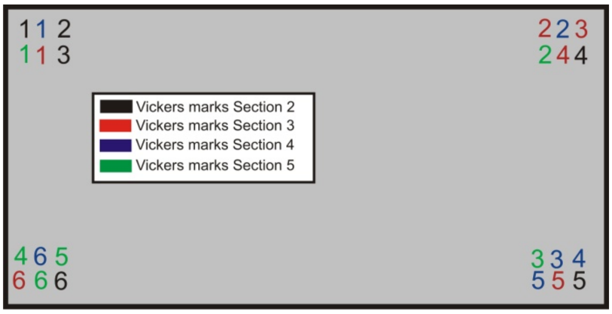

Figure 3: Schematic illustration of the position of Vickers marks for the different analysed sections on the specimen.

In addition to the performed automated SEM/EDS analyses as well as the statistical considerations, a part of the analysed metallographic specimen was finally used for electrolytic extraction experiments. This method is based on the dissolution of the steel matrix in a galvanic cell, non-metallic inclusions rest in the residue an can subsequently be analysed in the SEM. So, a three-dimensional view on the non-metallic inclusions and a detailed insight concerning inclusion morphology can be gained. For details concerning the experimental set-up as well as the used electrolytes and test parameters it is referred to [9].

RESULTS

Representative Sample Area as a Function of the Inclusion Content

Based on the formulated geometric-statistical model (details given in [8]), the representative sample area for the investigated steel grade was defined. **Figure 4** illustrates the mean value and the standard deviation of the error of the area ratio of the NMIs in dependence of the measuring area. Since in practice usually a lower number of particles – resulting in a smaller area ratio – is detected during the measurement, the indicated error is displayed with a negative algebraic sign.

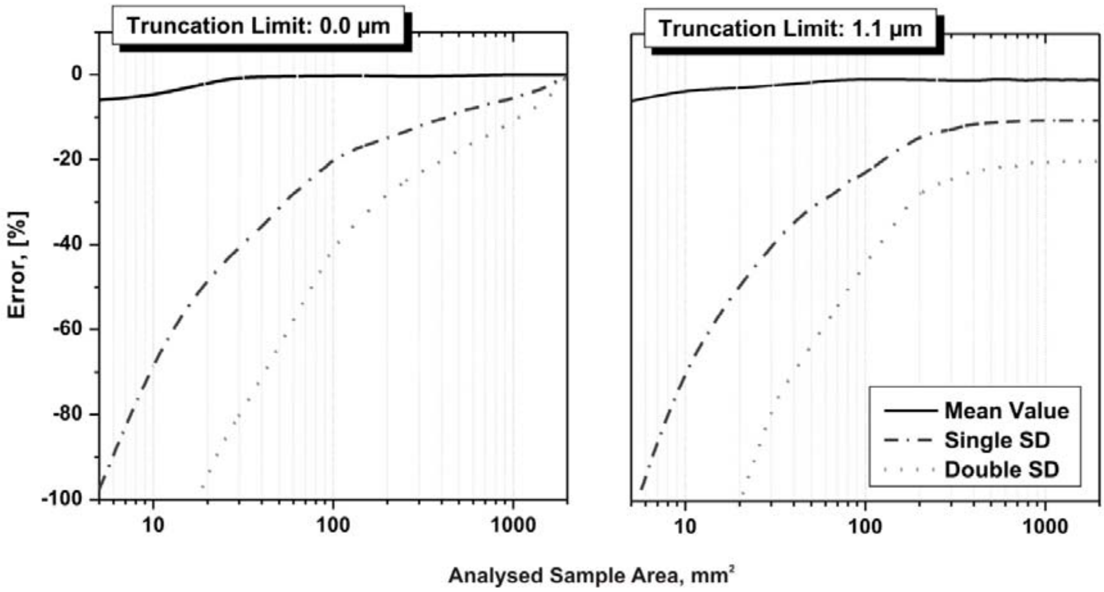

Figure 4: Error of area ratio in dependence of the analysed sample area for different truncation limits (SD: Standard Deviation).

Out of the results shown in **Figure 4** the following conclusions can be drawn for the application of automated SEM/EDS analyses for the defined conditions:

- If an area of 200 mm^2 – a defined number of inclusions per mm^2 provided – is measured by presetting a minimum particle diameter of 1.1 µm, the mean error of the measured area ratio lies approximately at 2 %. Looking at the standard deviation, in the worst case the error can amount to nearly 30 %.

- Regarding the data truncated at 1.1 µm, it can be concluded that an increase of the analysed sample area above 200 mm² would not effectively ameliorate the results, as there is no noticeable influence on the resulting error due to the truncation of the data. The truncation of the experimental data implies a certain systematic error, independent of the analysed area.

- In contrast to this, a reduction of the measured area causes a rapid and significant increase of the error. An area of 100-200 mm^2 seems to be a reasonable compromise under the given parameters.

- Since in the present study only one inclusion type is considered the assumed inclusion content in the steel matrix is very low (appr. 2 NMI per mm^2). Consequently, 200 mm^2 should be a sufficient area size in either case.

Further results and explanations of the statistical results as well as comparison with experimental data can be found in [8].

Mean Equivalent Circle Diameter and Size Distributions

Figure 5 shows a comparison of the detected mean ECDs in the two different experimental cases as well as in the performed calculations. It can be seen that the observed fluctuation range over all plotted values is remarkably tight, all measured and calculated ECDs lie between 3.2 and 3.6 µm ECD with a standard deviation of 0.123 µm. Comparing the three examined areas the most noticeable differences are observed in case 2, meaning the experimental investigation of five superimposed sections. For the latter case the standard deviation was determined with 0.201 µm.

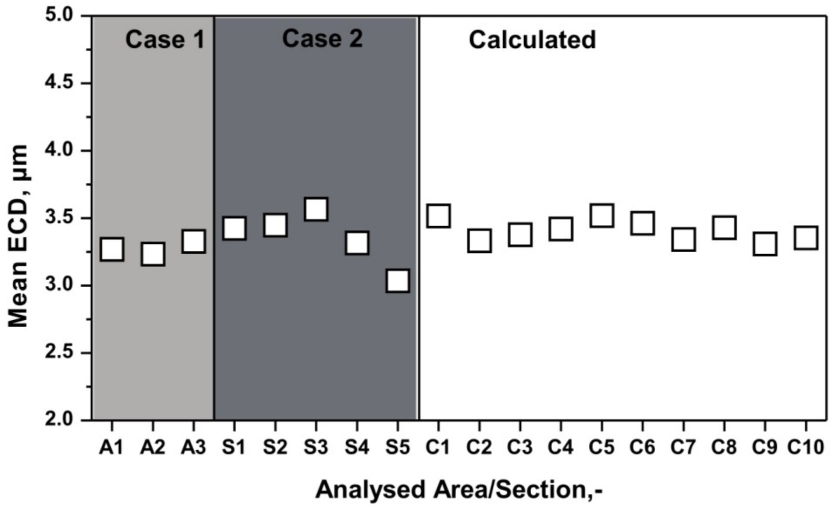

Figure 5: Comparison of mean ECD measured on the different analysed areas and sections with calculated values.

A comparison between the size distributions of Al_2O_3 inclusions between 1 and 10 µm for the experimentally analysed cases 1 and 2 is given in **Figure 6**. Case 1 demonstrates the influence of different measured area sizes: Principally, no significant difference can be observed, especially since this is a comparison of absolute values. The larger the ECD, the higher the observed differences. In contrast to this, the five analysed sections in case 2 show a remarkable larger deviation. Moreover, in case two generally a higher number of Al_2O_3 inclusions per mm^2 was detected.

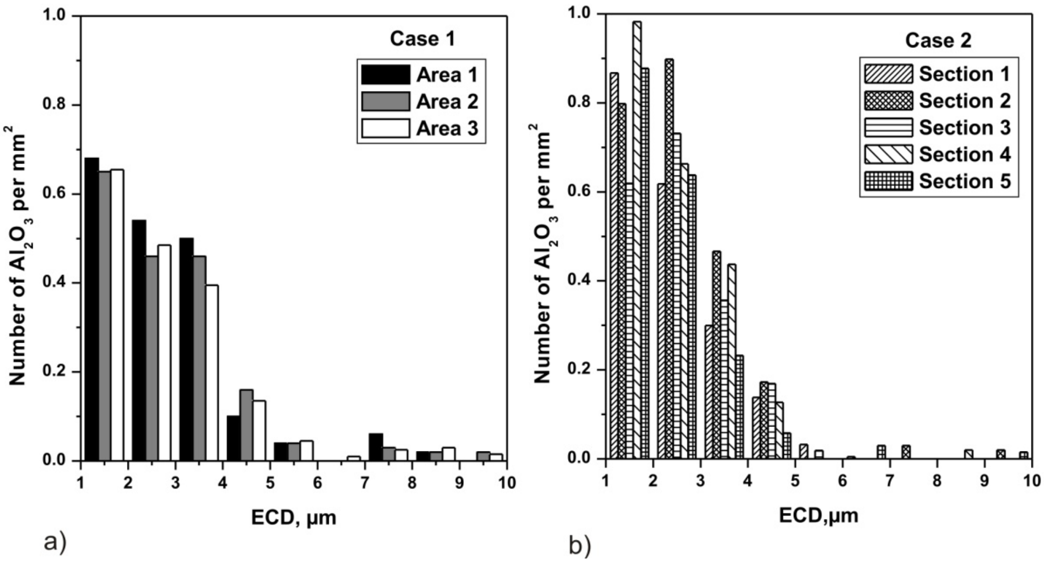

Figure 6: Comparison of size distributions measured on the different analysed areas and investigated sections.

Figure 7 summarises the size distributions of the 10 calculated sections as well as a comparison with experimental values from case 1 (Area 3). The calculated and measured values lie in comparable ranges regarding the overall number of particles per mm^2 as well as the distribution between 1 and 10 µm ECD. Nonetheless the observed fluctuations indicate the effect of the probability of intersection mentioned beforehand. In the performed investigations, a decrease of the analysed sample area seems to have a minor influence compared to the analysis of different superimposed sample sections.

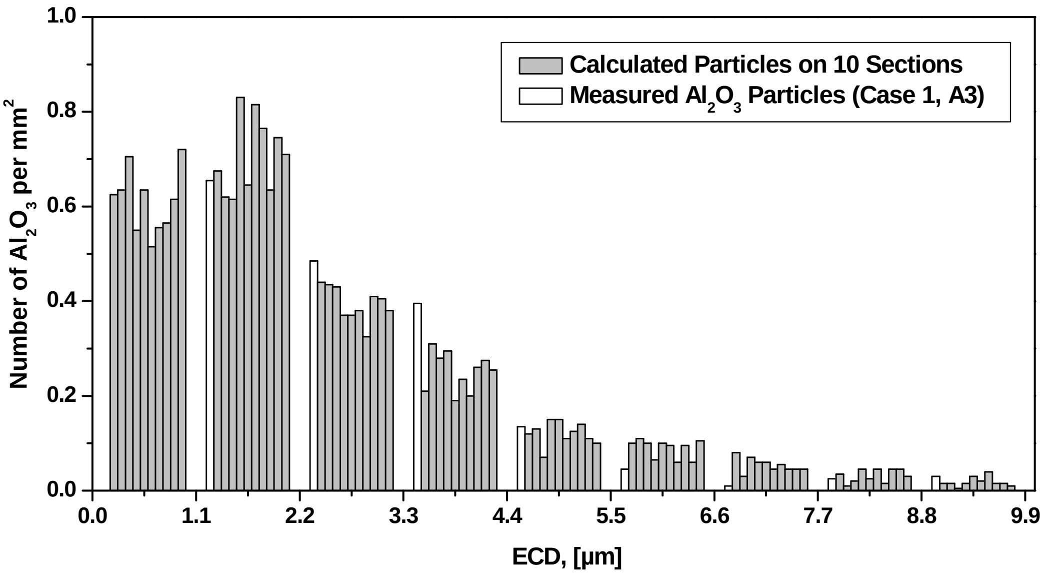

Figure 7: Size distribution of particles for the 10 sections of the calculations compared to the measured values of Al_2O_3 resulting from the analysis of Area 3 in case 1.

Maximum Detected Inclusion Diameter

An overview on the maximum detected inclusion diameters in the different cases is given in **Figure 8**. Compared to the detected mean ECDs (see **Figure 5**) a significantly enlarged scatter is found. In principle, apart from the intersection probability as a general problem, the probability of detecting inclusions larger than 50 µm ECD is low for the examined conditions because of the following reasons:

- The investigated steel grade already has reached a high cleanness level. Thus, larger inclusions are rare and randomly distributed in the analysed sample area.
- The analysed sample area is limited to 200 mm^2 in experimental analyses.

The most important conclusions from **Figure 8** can be summarised as follows:

- A variation in measuring area between the applied ranges (case 1 in **Figure 8**) does not increase the probability for the detection of a large inclusion. For all three areas the maximum detected inclusion is smaller than 20 µm ECD.

- Examining different sections (see case 2 in **Figure 8)** reflects the influence of intersection probability very clearly: Here the detected maximum ECDs are much higher compared to case 1 and also the scatter between the five sections is increased noticeably. The largest particle detected in case 2 has an ECD of 37.56 µm.

- The largest scatter was observed for the calculated values of the different sections. It can be seen that although most values are situated near the experimentally determined maximum ECDs, one outlier is observed. The largest detected particle in section C6 has an ECD of 103 µm. In contrast to the experimental analyses, where the real maximum inclusion is not known, the calculations enable a direct comparison: The largest distributed sphere in the defined volume had a diameter of 110 µm. Only in one of the calculated sections a maximum diameter larger than 50 µm was observed.

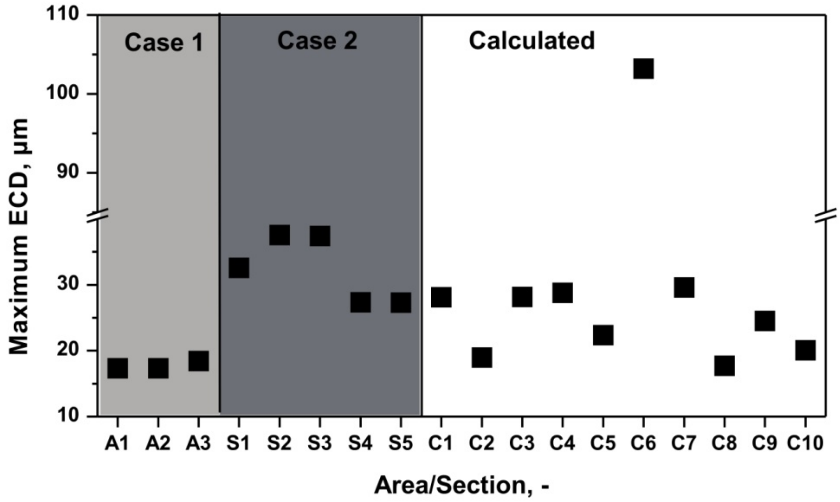

Figure 8: Comparison of maximum ECD measured on the different analysed areas and sections with calculated values.

After the automated analyses, the inclusion with the maximum ECD of each section of case 2 was analysed manually in detail. It was found that the largest particle in every examined section is situated at exactly the same position. Consequently, the largest particle in automated analyses in case 2 was always the same particle intersected at different distances from its centre. **Figure 9** schematically shows the development of the particle diameter from analysed section 1 to 5. A difference of more than 10 µm is observed between the different sections. A SEM-image of this inclusion in section 1 and 5 is also given in **Figure 9**.

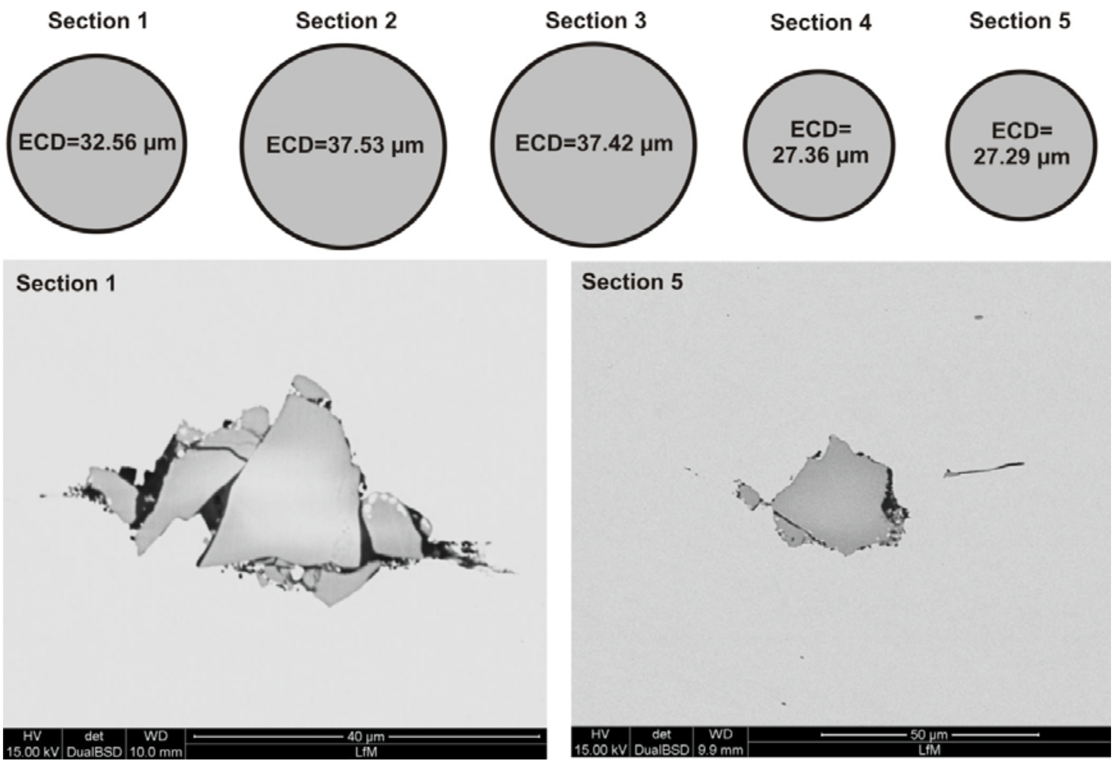

Figure 9: ECD-Development of the largest detected particle from section 1 to 5 in case 2 and corresponding SEM-images (The images show exactly the same particle cut at different distances from the centre).

These results underline the randomness regarding the detection of the maximum ECD of particles using automated SEM/EDS analysis and foremost the influence of the intersection plane on the appearance and size of the particle. Therefore, a concrete indication regarding the maximum inclusion in a defined steel volume has to be reviewed critically. Furthermore, it must be noticed that also a large measuring area is not a warranty for detecting the largest particle in the analysed volume.

In addition to the SEM/EDS analyses a part of the sample was finally used for electrolytic extraction experiments. **Figure 10** demonstrates two SEM-images of typical particles found on the filter residue. The majority of extracted inclusions features a diameter smaller than 5 µm. The largest particle that was found using this inclusion characterisation method had an ECD of approximately 30 µm (see **Figure 10).** The apparent advantage of this method is the three-dimensional view on the inclusion. Thus, the effective inclusion size can be determined. Comparing the largest detected particles from automated SEM/EDS analysis and electrolytic extraction a rather good accordance is achieved. Since the dissolved sample volume was rather small (a few grams), also the probability of detecting a larger particle was limited.

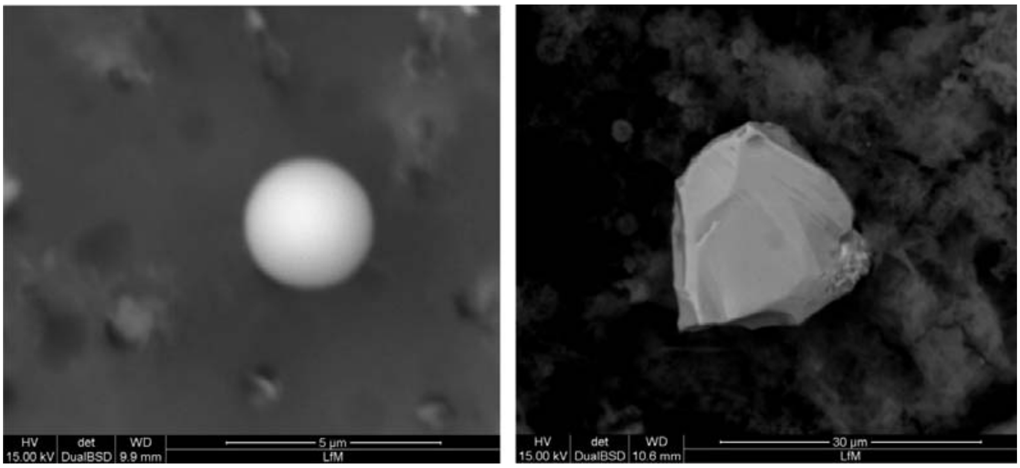

Figure 10: SEM-images of typical inclusions on the filter residue after electrolytic extraction experiments.

SUMMARY AND CONCLUSION

Within the present paper the influence measuring area size as well as the analysis of different intersection planes on the mean and maximum inclusion diameter has been examined experimentally. Additionally, a geometric-statistical model was applied in order to compare the experimental results with theoretic calculations. Furthermore, the established model enables the definition of a representative sample area as a function of inclusion content. Out of the performed investigations the following conclusions can be drawn:

- The analysis of a sample area between 100 and 200 mm^2 offers a valuable basis for automated SEM/EDS measurements under the assumed conditions. A significant increase of the analysing area, also resulting in a considerable increase of measuring time, would not result in a more representative output in this case. In contrast to this, a decrease of the measuring area can provoke a substantial increase of the error especially as far as inclusions larger than 5 µm in ECD are concerned.

- Concerning the mean ECD a satisfying consistency was observed not only between the two experimental cases but also in comparison with the calculations. Moreover, the size distributions showed a good agreement. Thus, a defined number of inclusions per mm^2 provided, a representative insight regarding the global inclusion landscape between 1 and 10 µm ECD is gained by investigating a single intersection plane of a metallographic specimen.

- Substantial differences regarding the maximum detected ECD were found not only between the two experimental cases, but also compared to the calculations. While the maximum sphere diameter distributed in the model was defined with 110 µm, only in one calculated section a particle larger than 100 µm was found. In all experimental analyses no inclusion larger than 40 µm was observed. This fact underlines the influence of intersection probability on the obtained result. Thus, the significance of automated SEM/EDS analyses regarding the detection of the maximum inclusion diameter is limited.

- Comparing the experimental results of case 1 and 2 the investigated intersection plane seems to have a higher influence on the results than the variation of measuring area on a single intersection plane, especially as far as the detection of the maximum particle is concerned.

- Although the results of electrolytic extraction and SEM/EDS are comparable concerning the detected inclusion sizes, the extraction method offers a three-dimensional view on the inclusion and therefore also the determination of the effective inclusion size.

Since the analysed inclusion content in the present study was rather small due to the fact that only one single inclusion type has been considered the drawn conclusions should be applicable for a broad range of steel grades.

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