Application of Extreme Value Analysis for Two- and Three-Dimensional Determinations of the Largest Inclusion in Metal Samples

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The maximum size of single inclusion particles and clusters in an Fe–10 mass% Ni alloy deoxidized with Al or Ti/Al were examined using extreme value analysis. The results obtained from conventional two-dimensional observations of inclusions on a polished cross section of metal sample (the CS-method) were compared to those from three-dimensional investigations of inclusions on a film filter after electrolytic extraction (the EE-method). It was found that the EE-method can successfully be used as a reference method for estimation of the probable maximum size of single inclusion particles and clusters by using an extreme value distribution (EVD). The EVD results for single inclusion particles obtained from the EE-method agreed satisfactorily well with those from a conventional CS-method. However, this required identification as well as neglect of pores on an investigated cross section of a metal sample. The predicted maximum size of single inclusion particles in a 1 mm³ volume was confirmed by results from the EE-method.

KEY WORDS: extreme value analysis; inclusion size; cross section; electrolytic extraction; single inclusion; cluster.

1. Introduction

In general, it is well known that the surface and internal qualities as well as the mechanical properties of steel are deteriorated by the presence of large inclusions. In order to assess the cleanliness of steel, the statistical extreme value analysis of the largest inclusion particle is one useful method to detect large inclusions.1–5) The probable maximum size of inclusion particles in a particular area or volume of metal can statistically be predicted by determining the extreme value distribution (EVD) of inclusion sizes. In addition, the advantage of this method is that data can easily and quickly be obtained by using light optical microscopy (LOM). However, other methods such as the ultrasound sonic test6) or the electric sensing zone method7) require special equipments for investigation of inclusions. Also, the extreme value analysis of inclusion size has been applied by many researchers8–11) for precise estimation of the maximum size of inclusion particles in various steel grades.

In the previous studies1–5,9–11) focusing on extreme value analysis, an apparent maximum size of inclusion particles was measured on a polished cross section of metal samples. However, the apparent maximum size of an inclusion section on a cross section determined by two-dimensional (2D) observations did usually not equal to the actual maximum size of inclusion particles. This difference was particularly true for the inclusion particles and clusters whose shapes were irregular. Moreover, it is difficult to directly predict the probable maximum size of an inclusion particle in a particular volume using 2D observations. For the determination of the relation between the apparent and actual diameter of inclusions in extreme value analysis and for estimation of the probable largest inclusion particle in a volume from 2D observations, several studies have been performed.12–14) More specifically, for recalculation of EVD data obtained from 2D observations to those data in a volume, Uemura and Murakami12) have assumed that the thickness of a metal layer analyzed by 2D measurement equals to the average value of the maximum inclusion sizes obtained from the extreme value analysis. Also, Takahashi13) has studied the relationship between EVD of particle sizes obtained by 2D measurements and those by 3D measurements using numerical simulations. In his study, it has been pointed out that the conversion of a dimension from 2D to 3D by using an assumed thickness may introduce over estimation of a unit volume. In addition, Zhou et al.14) has investigated the maximum inclusion size in 3D by the depth-profile method. Although a relationship between the 2D and 3D measurements and results from an extreme value analysis of inclusions has been clear from these studies, the direct observation and verification of the actual largest inclusion in a metal for extreme value analysis has not been presented yet.

When investigating large inclusions, clusters should also
be included since they are very harmful in the steel making process as well as for the material properties. Here, several studies used numerical simulation to clarify that clusters are formed due to collision and agglomeration of particles in liquid steel during ladle treatment and casting. Meanwhile, it is known that the growth mechanism of single inclusion particles is generally due to Ostwald ripening in the liquid metal. Hereby, it can be assumed that single inclusion particles are generally due to Ostwald ripening in the liquid metal. It has been measured using optical microscopy. The detection of pores was carefully carried out by separating them from non-metallic inclusions based on the difference of the focusing depth compared to the matrix. In addition, a scanning electron microscope (SEM) was used to confirm the objects as non-metallic inclusions or pores by using backscattered electron composition image analysis.

On the other hand, in order to measure the actual maximum inclusion size for a 3D investigation, inclusions on a film filter were observed after electrolytic extraction. More specifically, a potentiostatic electrolytic extraction of inclusions from metal samples was carried out using a 10% AA non-aqueous solution (10 v/v% acetylacetone – 1 w/v% tetramethylammonium chloride – methanol). The following parameters were used during the extraction: a 150 mV voltage, a 40–50 mA/cm² electric current and a 500 Coulomb extraction rate, followed by water quenching. The metal specimens for extraction were cut to pieces of almost the same size to establish constant dissolution conditions. The total surface area of each specimen was adjusted to approximately 240 mm² (7×10×3 mm).

After electrolytic extraction, the electrolyte containing non-metallic inclusions was filtered through a membrane polycarbonate (PC) film filter having a 0.05 μm open-pore diameter. The weight of the dissolved metal, Wdis, and the calculated thickness of the dissolved metal layer during electrolytic extraction varied between 0.1380 and 0.1434 g, and between 73 and 75 μm, respectively (Table 2). The inclusions on a film filter were observed using an SEM. This method is denoted as the EE-method in the following part of this study. In addition, the largest inclusion particle in a metal volume of 1 mm³ was determined for the extracted inclusions. Thereafter, the largest inclusion particle was compared to the predicted maximum size of inclusion particles using extreme value analysis in order to confirm the accuracy of the prediction.

| Exp. No | Wdis (g) | Vdis (mm³) | Thickness of dissolved metal (μm) |
|--------|---------|-----------|----------------------------------|
| 1      | 0.1380  | 17.69     | 74                               |
| 2      | 0.1426  | 18.28     | 73                               |
| 3      | 0.1380  | 17.69     | 74                               |
| 4      | 0.1434  | 18.38     | 75                               |
| 5      | 0.1433  | 18.37     | 73                               |

3. Analytical Methods for Determination of the Extreme Value Distribution

The statistical analysis in the present work was performed...
using Murakami’s method. The basic procedure of this analysis is to first rank the size of inclusions in an increasing order, like $x_1 \leq x_2 \leq ... \leq x_n$, where $x_i$ is representing size parameter of inclusion. Thereafter, in order to obtain EVD, the reduced variate of each size data, $y_i$, determined by Eq. (1) is plotted against a size parameter.

$$y = -\ln \left( -\ln \left( \frac{i}{n+1} \right) \right) \quad (1)$$

According to Murakami’s method, the square root of an area of the largest single inclusion particle and cluster in a unit area, $\sqrt{\text{area}_{\text{max}}}$, was used as a parameter to represent the inclusion size for both the CS- and EE-methods. In addition, non-metallic inclusions in a metal sample were classified into two groups, such as a “single inclusion” and a “cluster”. In this study, the “single inclusions” on cross sections and on film filters is defined as an isolated single inclusion particle without any connections with other particles, as illustrated in the upper photograph of Fig. 1. Also, a group of several single inclusions closely spaced on a cross section were also assumed to be one cluster in the CS-method. This is due to the possibility for the particles to be connected under the surface. In such case, it was judged to be a “cluster” if the distance between neighboring particles was closer than 5 $\mu$m.

For spherical and round shaped inclusions in the CS- and EE-methods, the $\sqrt{\text{area}_{\text{max}}}$ value is calculated as a square root of a circular area from a particle diameter, $d$, by using Eq. (2):

$$\sqrt{\text{area}_{\text{max}}} = \sqrt{\pi \times \frac{d}{2}} \quad \text{(for spherical particles)} \quad (2)$$

The $\sqrt{\text{area}_{\text{max}}}$ values of clusters and non-spherical single inclusions were determined as a rectangular shape based on the longest length with the maximum width as shown in Fig. 1. In this case, the $\sqrt{\text{area}_{\text{max}}}$ values are calculated from Eq. (3):

$$\sqrt{\text{area}_{\text{max}}} = \sqrt{a \times b} \quad \text{(for non-spherical particles and clusters)} \quad (3)$$

where $a$ is the longest length and $b$ is the maximum width of inclusions. It is easily considered that the $\sqrt{\text{area}_{\text{max}}}$ value calculated by Eq. (3) is excessively estimated from the lower right hand side of Fig. 1. This will be explained in Section 4.2. In order to compare the results obtained by 2D measurements using the CS-method to those by using the 3D measurements, the experimental results of the cross sectional measurements need to be recalculated to 3D data. According to Uemura and Murakami, the thickness of a metal layer analyzed by 2D observation, $h_0$, corresponds to the mean $\sqrt{\text{area}_{\text{max}}}$ value given by the following equation:

$$h_0 = \frac{1}{n} \sum \sqrt{\text{area}_{\text{max}}} \quad (4)$$

where $n$ is the number of measured inclusions for the CS-method. From this assumption, the unit volume of a metal sample analyzed by the CS-method, $V_{CS}$, was estimated according to the following equation:

$$V_{CS} = h_0 \times A_{CS} \quad (5)$$

where $A_{CS}$ is a unit observed area of a metal sample analyzed by CS-method.

In the EE-method, representing the 3D analysis, the unit investigated or analyzed volume of a metal sample, $V_{EE}$, for extreme value analysis was calculated from the following equation:

$$V_{EE} = \left( A_{EE} / A_{fil} \right) \times (W_{dis} / \rho_m) \quad (6)$$

where $A_{EE}$ and $A_{fil}$ are an observed area and the whole filtration area (1017 mm$^2$) of a film filter, respectively. Furthermore, $\rho_m$ is the iron density (0.0078 g/mm$^3$). The $W_{dis}$ value is the dissolved metal weight, determined by the difference of sample weight before and after electrolytic extraction.

According to Eq. (6), it should be pointed out that the $V_{EE}$ value depends on the dissolved metal weight as well as on the area of one SEM photograph. Therefore, the values of $W_{dis}$ should be kept almost constant in all electrolytic extraction experiments in order to obtain a similar $V_{EE}$ value. This study aims at evaluating the inclusion size between the CS- and EE-methods equivalently. Therefore, the unit volumes evaluated by the both methods have to be kept equal:

| Inclusion | CS-method | EE-method | $\sqrt{\text{area}_{\text{max}}}$ |
|-----------|------------|-----------|----------------------------------|
| Single inclusion | $\sqrt{\pi \times \frac{d}{2}}$ | $\sqrt{a \times b}$ |
| Cluster | $\sqrt{\pi \times \frac{d}{2}}$ | $\sqrt{a \times b}$ |

Fig. 1. Estimation of size for single inclusions and clusters by 2D (CS-method) and 3D (EE-method) investigation.
$V_{EE} = V_{CS}$ \hspace{1cm} (7)

By introducing Eqs. (5) and (6) into Eq. (7), the relationship between the observed unit areas from both methods can be given as follow:

\[ \frac{A_{EE}}{A_{CS}} = \frac{h_0 \cdot A_{cl} \cdot \rho_{m}}{W_{dis}} \] \hspace{1cm} (8)

The average values of $\sqrt{\text{area}_{\text{max}}}$ in each experiment are shown as $\bar{T}_0$ for single inclusion particles and as $\bar{T}_d$ for clusters in Table 3. The $\bar{T}_0$ and $\bar{T}_d$ values were introduced using the following equations, respectively:

\[ \bar{T}_0 = \frac{1}{n} \sum \sqrt{\text{area}_{\text{max}}} \hspace{1cm} (9) \]
\[ \bar{T}_d = \frac{1}{n_{cl}} \sum \sqrt{\text{area}_{\text{max}}} \hspace{1cm} (10) \]

where $n$ and $n_{cl}$ is the number of measured single inclusion particles and clusters, respectively. Table 3 also shows the revised $T_0$ values given as Exps. 1a, 2a and 4a by the CS-method to verify the influence of pores on a cross section on EVDs. The detail about the effect of pores on the results will be described later in Section 4.1. In this study, the $h_0$ values were determined as the $T_0$ values obtained by the CS-method under the specific assumption of a unit volume. Thereafter, the $A_{EE}/A_{CS}$ ratio was calculated to be 0.23 from Eq. (8), by using the mean values of $h_0$ ($h_0 = 4.0 \mu m$) for all CS experiments and $W_{dis}$ ($W_{dis} = 0.1411$ g) for all EE experiments. The $h_0$ and $W_{dis}$ values in different experiments were scattered between 3.5 and 4.7 $\mu m$ and between 0.1380 and 0.1434 g, respectively. Thus, the mean values of both parameters ($\bar{T}_0$ and $\bar{T}_d$) were applied for the determination of the constant unit area which was observed. Based on the given $A_{EE}/A_{CS}$ ratio value, the $A_{EE}$ value can be determined to be 0.042 mm$^2$. Thus, $V_{EE}$ can be calculated as (0.042 mm$^2$/1017 mm$^3$)$\times$(0.1411 g/0.0078 g/mm$^3$) = 7.47$\times$10$^{-4}$ mm$^3$ by Eq. (6). Furthermore, $V_{CS}$ can be given as 4$\times$10$^{-3}$ mm$^3$/0.186 mm$^3$ = 7.44$\times$10$^{-4}$ mm$^3$ by Eq. (5). The both unit volumes are in accordance with each other to enable an equivalent comparison. Thereby, 0.045 mm$^2$ (0.18$\times$0.25 mm) was decided as $A_{EE}$ on an SEM photograph taken at a magnification of 500 times. In fact, this procedure is so important to adopt to enable a comparison between the CS- and EE-methods. Figure 2 shows how well the both unit volumes of each experiment agree with each other. Here, the values of $V_{CS}$ and $V_{EE}$ for different experiments have been determined from the individual $h_0$ and $W_{dis}$ values obtained from each experiment. It can be seen that the mean $V_{EE}$ value ($7.82\times$10$^{-4}$ mm$^3$) agrees well with the mean $V_{CS}$ value ($7.53\times$10$^{-4}$ mm$^3$).

The regression lines of every EVD data were calculated as a linear function from the maximum-likelihood method, according to the ASTM E2283-03 standard.\(^{20}\) By using a regression line, it is possible to extrapolate the probable maximum size of an inclusion in a reference volume, $V_{ref}$. In this study, the $V_{ref}$ value was determined as 1 mm$^3$ for all experiments. The probable maximum size of the inclusion in a reference volume can be estimated by introducing the $y$ value, which corresponds to a 1 mm$^3$ volume in the $V_{ref}$ value according to Eqs. (11) and (12), to the linear regression formula given in Table 4.

\[ y = -\ln \left( \frac{-\ln \left( \frac{T-1}{T} \right)}{T} \right) \hspace{1cm} (11) \]
\[ T = \frac{V_{ref}}{V_0} \hspace{1cm} (12) \]

where $y$ is the reduced variate, $T$ is the return period for the expected volume and $V_0$ is a unit volume. In this study, the individual $V_{CS}$ and $V_{EE}$ values obtained from each experiment were used as a unit volume, $V_0$. The details regarding the regression formulas and the $y$ values for each experiment will be mentioned later in Section 4.3.

The difference in unit volumes between the CS- and EE-methods in each experiment is caused by the difference in the individual $h_0$ and $W_{dis}$ values obtained from each experiment. In order to estimate the influence of such variability of a unit area and volume on the EVD in each experiment, the extreme value analysis has been examined using two different values of the observed unit areas. Figure 3 shows the comparison of the EVD obtained by using a single ($A_{CS}$ and $A_{EE}$) and a double (2$\times$A$A_{CS}$ and 2$\times$A$A_{EE}$) unit area. More spe-

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**Table 3.** Measurement number, average size for the largest inclusion and unit volume for each experiment.

| Exp. No | Single inclusion | Cluster | $V_{CS}$ | $V_{EE}$ |
|---------|------------------|---------|----------|----------|
|         | $n$ | $\bar{T}_0$ ($\mu m$) | $n_{cl}$ | $\bar{T}_d$ ($\mu m$) | ($\times$10$^{-4}$ mm$^3$) |
| 1       | 40  | 4.7  | 23  | 8.5  | 8.78  | – |
| 2       | 40  | 3.5  | 30  | 6.9  | 6.59  | – |
| 3       | 40  | 3.8  | 30  | 6.1  | 7.14  | – |
| 4       | 40  | 4.7  | 23  | 12.6 | 8.69  | – |
| 5       | 40  | 3.5  | 30  | 8.3  | 6.43  | – |

CS-method

| Exp. No | Single inclusion | Cluster | $V_{CS}$ | $V_{EE}$ |
|---------|------------------|---------|----------|----------|
| 1a      | 40  | 3.0  | –   | –   | 8.78  | – |
| 2a      | 40  | 3.8  | –   | –   | 6.59  | – |
| 4a      | 40  | 3.4  | –   | –   | 8.69  | – |

EE-method

| Exp. No | Single inclusion | Cluster | $V_{CS}$ | $V_{EE}$ |
|---------|------------------|---------|----------|----------|
| 1       | 40  | 2.9  | 29  | 5.3  | 7.65  | – |
| 2       | 40  | 2.9  | 34  | 6.9  | 7.90  | – |
| 3       | 40  | 2.8  | 29  | 4.4  | 7.65  | – |
| 4       | 40  | 2.5  | 36  | 3.8  | 7.95  | – |
| 5       | 40  | 3.1  | 34  | 5.1  | 7.94  | – |
Specifically, this figure includes the data for Exp. 5 obtained from (a) the CS-method and (b) the EE-method. For the CS-method, the regression lines for both single inclusions and clusters are determined from \(2 \times A_{CS}\). This corresponds to the dashed line. It is slightly shifted to the right side of the data obtained from \(A_{CS}\) with a similar slope, as shown in Fig. 3(a). Also, it can be seen in Fig. 3(b) that the regression lines from both unit areas in the EE-method have a similar slope. Thus, the slopes of the regression lines for the EVD determined from the single and double unit areas were almost the same. In these cases, the double unit areas for the CS- and EE-methods yield mostly two times of a unit volume compared to the single unit area. The difference of unit volume between the CS- and EE-methods in all experiments is smaller than 20%, as shown in Fig. 2. Hereby, the experimental result shows that the difference of a unit volume does not have a significant effect on the slopes of the regression lines determined from each EVD. The influence of the variability of a unit volume on the EVD determined in this study can be considered to be negligibly small.

### 4. Results and Discussion

#### 4.1. Extreme Value Distribution of Single Inclusions

Figure 4 shows the typical EVD values for single inclusions obtained from the CS- and EE-methods. Two typical tendencies could be observed by comparison of the distributions. One of them shows mostly a coincidence of distributions obtained from the CS- and EE-methods, as shown in Fig. 4(a). Another tendency shows a significant discrepancy between the regressed lines of EVD data obtained from each method, as shown in Fig. 4(b). In this case, the slope of the regression line from the EE-method is considerably larger compared to that from the CS-method. Figure 5 shows the comparison of the regression lines determined from EVD data for single inclusions in each experiment obtained by (a)
the EE-method and (b) the CS-method. The equations of regression lines for each experiment are given in Table 4. It is obvious that the regression lines for single inclusions by the EE-method agree satisfactorily well with each other for most experiments, as illustrated in Fig. 5(a). However, the regression lines obtained by using the CS-method for Exps. 1 and 4 are significantly different from those for Exps. 2, 3 and 5, as shown in Fig. 5(b).

To clarify the factor causing such a discrepancy in the EVD values between both methods in Exps. 1 and 4, a careful observation of the metal surface was carried out by using SEM. **Figure 6** shows the optical microscopic image (Fig. 6(a)) and the backscattered electron composition image (Fig. 6(b)) of the same object on a cross section of a metal sample obtained from Exp. 4. As shown in Fig. 6(a), this object determined by optical microscopic observation looks like typical non-metallic inclusion, as shown in Fig. 1 for the CS-method. However, as illustrated in Fig. 6(b), the same object has been identified as a pore on a cross section due to the same composition with the metal matrix by using backscattered electron compositional image analysis. Several objects on a cross section were also detected as pores. **Figure 7** shows that the number of pores larger than 5 μm on a 50 mm² area of a cross section in the metal sample of Exps. 1 and 4 is significantly more than the number of pores in other experiments. More specifically, the total number of pores larger than 6 μm in size has been counted as 8 and 10 for Exps. 1 and 4, respectively. Meanwhile, there were 1–3 pores in the samples from Exps. 2, 3 and 5. Such large size pores may introduce a large discrepancy between the EVD obtained from the CS-method by optical microscopic observation and the EE-method.

In order to exclude the detrimental effect of pores, the metal samples of Exps. 1, 2 and 4 were examined with the CS-method again after carefully polishing their cross sections again. **Figure 8** shows the comparison of the revised EVD plots shown as Exps. 1a and 4a with the original ones (Exps. 1 and 4). The tendency of the distributions in the range smaller than 4 μm agrees well between the original and revised data. However, when the $\sqrt{\text{area}_{\max}}$ value is larger than 5 μm, the distributions firstly measured show significant discrepancies with the revised ones. The comparison of the regression lines obtained from the original and revised results is shown in **Fig. 9**. Consequently, the revised regression lines in Exps. 1a and 4a have lower slopes than the respective original lines in Exps. 1 and 4 based on the larger sized data. Thus, it can be suggested that the frequent existence of large-size pores significantly affects the slope of the regression lines representing the EVD data. On the other hand, the regression line from the sample in Exp. 2, which has fewer pores, shows a satisfactory agreement between two measurements (2 and 2a).

As one can realize in Table 3, the $T_0$ values obtained by

![Fig. 6. Optical microscopic image (a) and backscattered electron compositional image (b) of typical pore on a cross section of metal sample (Exp. 4).](image)

![Fig. 7. Size distributions of pores on a cross section of metal samples in different deoxidation experiments. The analyzed area is 50 mm².](image)

![Fig. 8. Comparison of the revised EVD plots with the original one in (a) Exp. 1 and (b) Exp. 4 by CS-method.](image)

![Fig. 9. Comparison of regression lines obtained by revision of metal samples in CS-method.](image)
the CS-method are originally larger than those obtained by the EE-method although these have to be agreed each other by definition. This tendency is enhanced especially for Exps. 1 and 4 which have more pores as mentioned above. When focusing on Exp. 1, the difference between the two measurements is originally 1.8 μm. However, if neglecting the pores, the corresponding difference is only 0.1 μm realized by a comparison of Exp. 1a of the CS-method with Exp 1 of the EE-method. The mean \( \bar{\mu}_l \) values by the CS and EE-methods are originally 4.0 and 2.8 μm, respectively. This brings the difference of 1.2 μm caused mainly by pores. However, the mean \( \bar{\mu}_l \) value by the CS-method accounting for the identified pores is 3.4 μm that is the average of the values of Exps. 1a, 2, 3, 4a, and 5. This correction gives the minimization in difference. Even though the correction has been made by this manner, the CS-method still has 0.6 μm larger value in average. This reason is not fully understood at this moment but the measurement error on photographs is considered to be one of the reasons. This will be discussed in a separate paper.

According to these results, it may be safely suggested that the pores on the surface of a metal cross section have a very negative effect on the accuracy of the EVD of single inclusion’s size when determined by LOM. Meanwhile, the EE-method enables us to cancel the influence of pores in the metal sample on EVD of single inclusions, due to the extraction of non-metallic inclusions and dissolution of the metal matrix.

4.2. Extreme Value Distribution of Clusters

The comparison of the typical EVD relations for single inclusions and clusters in one metal sample (Exp. 5) is shown in Fig. 10. In this figure, a 95% confidence interval (95%CI) of every regression line is also shown according to the ASTM E2283 standard. It can be seen that the EVDs obtained from the EE-method for single inclusions and clusters have almost linear relationships. In the case of the CS-method, the plots for clusters are slightly divergent against the regression line. Moreover, it is apparent that the clusters have the smaller slope of the EVD regression lines compared to single inclusions in both methods. A similar tendency was obtained for the other experiments. This is due to the difference of growing mechanisms for single inclusions grown by Ostwald ripening and for clusters grown by collision and agglomeration of inclusions in a molten metal. Thus, clusters in a metal should be investigated separately from single inclusions for a precise prediction of the probable maximum size of them using extreme value analysis.

The typical result of the comparison of EVD for clusters determined by the CS- and EE-methods is shown in Fig. 11. Figure 11(a) shows the relatively close relationships obtained from both methods for the metal sample of Exp. 3. The data of Exps. 1 and 2 show same tendency for clusters as was found for Exp. 3. Meanwhile, Fig. 11(b) shows a considerable difference between the two EVD regression lines in Exp. 4. The data for Exp. 5 shows a significant difference between regression lines obtained from the CS-method and the EE-method, as well.

The results of experimental measurements and statistical analysis for clusters as well as single inclusions in all experiments are given in Tables 3 and 4. The measured number of clusters for extreme value analysis, \( n_{cl} \), was less than 40 which is required by the Murakami method. This is caused by the absence of clusters in some observed fields. More specifically, the \( n_{cl} \) values obtained from the EE-method varied from 29 to 36. The corresponding number for the CS-method varied between 23 and 30. As given in Table 3, the \( \bar{\mu}_l \) values obtained from the EE-method are significantly larger than those from the EE-method for most experiments. The \( \bar{\mu}_l \) values for different samples obtained by using the EE-method range from 3.8 to 6.9 μm, while the corresponding values from the CS-method varied between 6.1 and 12.6 μm. Based on these comparisons, it may be concluded that the 2D investigations correlate with smaller \( n_{cl} \) and larger \( \bar{\mu}_l \) values than the 3D investigations. Moreover, Fig. 12 shows the comparison of the size distribution for the clusters between the CS- and EE-methods in the sample of Exp. 4, which has largest discrepancy between the CS and EE results. It can be seen that most of the clusters observed in 2D by using the CS-method have a larger size (from 7 to 17 μm) compared to the clusters observed by using the EE-method (from 2 to 9 μm).

According to the geometrical study reported by Söder, the probability of the appearance of a cluster on a cross section for small clusters, such as two particles, is very low.

![Fig. 10. Comparison of EVD for single inclusions and clusters in metal samples of Exp. 5 obtained from EE-method (a) and CS-method (b).](image)

![Fig. 11. Comparison of EVD of clusters obtained from CS- and EE-methods. Typical example of closing distribution (a) for Exp. 3 and different distribution (b) for Exp. 4.](image)
Hereby, these experimental results, such as less clusters detection as well as a lack of smaller ones, may show the difficulty of detecting smaller size clusters (<7 μm) by 2D observations using the CS-method. The other factors causing the difference in EVD data between the CS- and EE-methods would be the difficulty of recognition of the actually clustering inclusions as a cluster on a cross section.

Inoue and Suito\textsuperscript{22} have clarified that the 10%AA electrolyte can be applied successfully for electrolytic extraction of Al\textsubscript{2}O\textsubscript{3} inclusions from metal samples deoxidized by Al without them being dissolved. Moreover, the physical destruction of extracted clusters during filtration is also unlikely, since broken parts of clusters were not detected in the SEM observations of clusters on a film filter after electrolytic extraction. As a result, it may be safely suggested that the Al\textsubscript{2}O\textsubscript{3} clusters could be separated successfully without any dissolution or destruction of them during electrolytic extraction when using a 10%AA electrolyte.

Hereby, some overestimation of the actual size of clusters by the CS-method was caused by the detection and measurement of unattached inclusions, clusters or some pores (see Section 4.1) as one cluster. In this study, a group of several particles located within a distance of 5 μm was determined as one cluster, as mentioned in experimental part. Figure 13 shows schematically the pattern of apparent clusters under a cross section of a sample, which consists of a group of several single inclusions (Fig. 13(a)), a single inclusion and a cluster (Fig. 13(b)) or several clusters (Fig. 13(c)). In the case of a group of several clusters (Fig. 13(c)), the measurement number of clusters would increase after electrolytic extraction. This is due to the separation of them from one apparent cluster in the metal matrix. Hereby, it can be concluded that the actual size of coalesced particles as one cluster can be detected and measured precisely by using the EE-method.

### 4.3. Comparison of the Probable Largest Inclusion from the CS- and EE-methods

It is of interest to compare the probable maximum size for single inclusions and clusters in a particular volume by using the 2D and the 3D investigations. To satisfy an equivalent comparison between both ways, the probable maximum sizes of them in a 1 mm\textsuperscript{3} reference volume (\(V_{\text{ref}}\)) have been calculated from the regression lines obtained from the CS- and EE-methods (Table 4). By introducing the individual unit volume for each experiment, \(V_0\), to Eq. (12), the reduced variate, \(y\), for 1 mm\textsuperscript{3} volume can eventually be determined from Eq. (11). This \(y\) value varied between 7.038 and 7.350 for the CS-method and between 7.137 and 7.176 for the EE-method. It should be mentioned that the probable maximum size is calculated from the linear regression lines for each experiment and those given \(y\) values which are out of the range (-2–6 in \(y\) value) of the vertical axis of Figs. 3–5 and 8–10. Some scattering in \(y\) values are caused by the small variability of unit volumes for each experiment.

The estimated values for both single inclusions and clusters are plotted in Fig. 14. It is obvious that the probable maximum sizes of clusters in a 1 mm\textsuperscript{3} volume are significantly larger than those of single inclusions in all experiments. The standard error, which is shown as both horizontal and vertical bars (\(\sqrt{\text{area}_{\text{max}} + 2\text{SE}}\) for each data point in Fig. 14, was cal-

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**Fig. 12.** Comparison of size distribution for maximum size clusters obtained from CS- and EE-methods in Exp. 4.

**Fig. 13.** Schematic illustration of 2D measurement for apparent size of cluster on metal cross section and real size of this cluster in metal volume in 3D.

**Fig. 14.** Comparison of the probable maximum size of single inclusions and clusters in 1 mm\textsuperscript{3} obtained from CS- and EE-methods.
calculated as a 95% CI for the calculated \( \sqrt{area_{\text{max}}} \) values according to ASTM E2283 standard. These values have been calculated from the following equations:

\[
95\% \text{CI} = \pm 2 \cdot SE \quad \cdots \cdots \cdots \cdots \cdots \cdots (13)
\]

\[
SE = \frac{1}{C} \sqrt{\frac{(1.109 + 0.514 \cdot y + 0.608 \cdot y^2)}{n}} \quad \cdots \cdots \cdots \cdots \cdots \cdots (14)
\]

where \( C \) is the slope value of linear regression line. The error bars for both single inclusions and clusters obtained from the CS-method (horizontal bars) tend to be significantly larger than the corresponding data from the EE-method (vertical bars). Hereby, the deviation of the estimated size by the CS-method is considered to become larger compared to the EE-method for the similar investigating conditions. More specifically, the number of measurement and the unit volume which is studied is same in both methods.

It can be seen that the probable maximum sizes of single inclusions and clusters in a 1 mm\(^3\) volume calculated from the CS-method are considerable larger in most experiments than those from the EE-method. Especially, large discrepancies between the probable maximum sizes are observed for single inclusions in Exps. 1 and 4 as well as for clusters in Exps. 4 and 5. For single inclusions, such a difference would be caused by the measurements of pores, as mentioned in Section 4.1. Figure 15 shows the difference between the estimated maximum sizes of single inclusions obtained from the CS- and EE-methods for a 1 mm\(^3\) volume, \( \Delta \sqrt{area_{\text{max}}} \), as a function of the pore population on a cross section of the metal sample. It can be seen that, the \( \Delta \sqrt{area_{\text{max}}} \) values tends to increase with the number of pores on a metal cross section. This is particularly true when the investigation has been done including pores. Thus, a frequent population of pores on a surface of metal sample introduces some overestimation error by estimation of the probable size of the largest single inclusions using the CS-method. Therefore, the extreme value analysis for inclusions using a cross sectional observation should be carried out thoroughly without pores.

The comparison of the maximum size for single inclusions in a 1 mm\(^3\) volume estimated from both methods and the actually observed ones from the EE-method is shown in Fig. 16. It can be seen that the maximum size of single inclusions in a 1 mm\(^3\) volume predicted from the EE-method agrees satisfactorily well with the observed ones. In the CS-method, the predicted sizes excluding pore measurements are also close to the actual size. However, the estimated sizes obtained from the CS-method data without identification and neglect of pores in Exps. 1 and 4 show a large discrepancy with the observed sizes.

In steelmaking production practice, it is difficult to investigate all products by the EE-method due to the high time consumption for electrolytic extraction of steel samples and investigation of inclusions and clusters on a film filter by SEM. However, this 3D investigation of inclusions can be suggested as a reference method for the correction of the results obtained from the faster conventional 2D investigation on a cross section of metal samples for different steel grades.

5. Conclusions

The size measurements of the largest single inclusions and clusters in an Fe–10 mass% Ni alloy have been examined using an extreme value analysis based on both three-dimensional (3D) and two-dimensional (2D) investigations. The extreme value distribution (EVD) and the probable maximum sizes of single inclusions and clusters obtained from a 3D investigation on a film filter after electrolytic extraction (EE-method) were compared with those obtained from a 2D investigation on a cross section of metal samples (CS-method). The most important conclusions may be summarized as follows:

1. The EVD data for single inclusions obtained from the EE-method almost agrees well with data excluding pores obtained from the CS-method.

2. The regression lines for EVD obtained from the CS- and EE-methods for clusters in all experiments have a smaller slope compared to the data for single inclusions. This is due to the difference of the growth mechanisms for single inclusions and clusters.

3. The probable maximum size of single inclusions obtained from the EE-method shows a good agreement with the observed maximum size for a given volume. However, the potential error with respect to the probable maximum size of single inclusions by using the CS-method increases with an increased porosity on the studied cross section of the metal sample.
(4) The probable maximum size of clusters obtained from the CS-method tends to be significantly larger than that from the EE-method. This overestimation of cluster size can be explained by the difficulty of detecting small clusters on a metal cross section as well as the measurement of the apparent cluster size in 2D. The apparent size of clusters in 2D could be larger than the real size in 3D, due to non-clustering. However, it could be closely located inclusions and pores.

(5) A 3D investigation, such as the EE-method, can be used as a reference method for prediction of the maximum size of single inclusions and clusters in the metal samples by using the extreme value analysis. It can be used to correct the result obtained from a conventional 2D investigation.

Finally, it is suggested that each investigating method, the CS- and EE-methods, should be applied for steelmaking production according to individual advantages and specific aims.

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