Determination of Size Distribution and Probable Maximum Size of Inclusions in AISI304 Stainless Steel

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The probable maximum sizes (PMS) of inclusions in AISI304 stainless steels were predicted by two methods of the statistics of extreme values (SEV) and the particle size distributions (PSD). Firstly, the PMS of inclusions in the molten steel taken from a tundish agreed well with those in the slab sample. The particle size distributions (PSD) of inclusions almost obeyed exponential functions. The results of comparison between the two methods showed that the PMS by the SEV analysis agreed with that by the PSD approximation in the case of the steel with the higher oxygen content. However, in the case of the steel with the lower oxygen content, the PMS by the PSD approximation overestimated the predicted size with the reference to the SEV analysis. In this case, the approximations with elimination of some largest inclusions which were deviated from an exponential distribution were found to be effective to predict the probable largest size in a reference area. This above result suggests that it is necessary to decide if some largest inclusions are employed for adequate predictions. It is considered that necessity of this operation increases with decreasing oxygen content.

KEY WORDS: stainless steel; inclusion; particle size distribution; statistics of extreme values; maximum size.

1. Introduction

In recent years, steels with high cleanliness are being required more and more to prevent surface defects. In addition, highly clean steel can maintain physical properties of products. For producing clean steel, a number of researchers make efforts to decrease number and size of inclusions in steel products.1–4) In addition to the studies to improve steel cleanliness, it is also important to determine the large size and the number of inclusions existing in steel to evaluate the quality of final products. For the measurement of a size distribution of large size inclusions (>22.6 μm), optical microscopic observation was tried as well as ultrasonic testing for the samples taken from liquid steel.5) Statistics of extreme values (SEV) is one of useful methods to evaluate the probable maximum size (PMS) of inclusions in a steel product. The PMS of inclusions can be predicted with sufficient accuracy in a certain area or volume by using an extreme value distribution (EVD) of inclusion sizes.5) In the previous studies,6–8) the authors have applied this EVD method to several kinds of steel samples taken from a tundish, a continuously cast slab and a hot band for prediction of size for the largest inclusion in the final steel products. The three-dimensional (3D) observation of inclusions with different morphologies was tried by electrolytic extraction method for more precise estimation of the maximum size inclusions in steel samples.6,7) In particular, 3D observation was found to be quite effective for the measurements of sulfide inclusions elongated by the hot-rolling process.

The measurement of particle size distribution (PSD) is also very important to understand the characteristic of inclusions. Probability distribution functions have been applied to describe numerically the PSD of inclusions in steel.9,10) Takahashi and Suito9) compared the PSD of spherical particles dispersed in a 3D space with the two-dimensional (2D) PSD appeared on a cross section by means of the numerical simulation. They applied various arithmetical functions (Mono-Dispersed, Exponential, Rayleigh, Log-Normal and Pseudo-Normal) for inclusions to approximate the PSD. It was further found11) that the functions arithmetically determined for expressed PSD would be used for estimation of the largest size inclusions in a unit area or volume in steel products. In addition, peaks over threshold (POT) method12–14) makes it possible to estimate the probable maximum size of an inclusion by using PSD with an adequate inclusion size as threshold for an analysis. It has been shown that assuming a numerically ideal distribution,13) the estimated inclusion size by the POT method provides the value mostly identical to the estimated size by the SEV.

In the previous studies,2,8) the analysis of inclusion sizes by SEV has been applied for AISI304 stainless steel in which silicate inclusions were observed. However, PSD of inclusions in this stainless steel grade has not been studied, yet. Therefore, in this study, the size distributions of inclu-
sions in AISI304 stainless steel were investigated in the samples taken from liquid steel in a tundish before casting and from solidified “as-cast” steel of slab after continuous casting process for two kinds of heats. Moreover, the PMS of inclusions determined by the EVD in steel samples was compared with the corresponding values obtained by the PSD. Based on the comparison, discussion will be made to give adequate guidance on how one should treat size distributions to have accurate predictions. Particularly, treatment of relatively large and less frequent inclusions is focused on for the predictions.

2. Experimental

The chemical compositions of the steels are shown in Table 1. Two different heats of AISI304 stainless steels with the total oxygen contents of 17 mass ppm (Steel A) and 28 mass ppm (Steel B) were selected for evaluation of the PMS of inclusions in steel samples. This steel grade typically contains 0.5% Si, 1.0% Mn, 8.0% Ni and 18% Cr (in mass%). The samples in the both heats were taken from liquid steels in the tundish (molten steel sample) and from solidified “as-cast” steel slabs (slab sample). The specimens of the slab samples were taken from the position of 1/4 thickness of the slabs at the edge part of slab width. It should be noticed that the samples were taken avoiding the positions where the both columnar structures met which grew from the narrow and the broad surfaces. The inclusions were observed aiming at the prediction of the largest inclusions at the surface layers at which the surface defects were in issues. For this aim, the validity to take the samples from the corresponding positions was proved in our previous study. Namely, the largest predicted sizes at the 1/4 thickness positions were in good agreement with those at the surface layers.

At first, the compositions of oxide inclusions in the samples of Steel A and Steel B were analysed by energy dispersive spectrometry (EDS) equipped to scanning electron microscopy (SEM). 10 typical inclusions in each sample were analyzed quantitatively.

The size of largest inclusion on each unit area of polished cross sections of the steel samples was measured in 2D by using a light optical microscopy (LOM) at a magnification of 200 times. The largest inclusions were measured on 40 unit areas to apply SEV. The unit area, \( A_{0} \), for every experiment was chosen as 1 mm\(^2\), and the total observed area for each sample was 40 mm\(^2\).

The SEV analysis is based on the method proposed by Murakami. According to Murakami’s method in conjunction with the ASTM standard, all the measured inclusions were ordered with an increasing of the maximum sizes as following, \( x_{1} \leq x_{2} \leq \ldots \leq x_{n} \) (1 \( \leq i \leq n \)), where \( x_{i} \) is the representing size parameter of the largest inclusions on the \( i \)-th unit area field. Thereafter, the reduced variate of each size data, \( y_{i} \), was determined by Eq. (1) and plotted against a size parameter.

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

A regression line of EVD data can be calculated by the maximum-likelihood (ML) method according to the ASTM E2283-03 standard. The obtained EVD regression line is applied to extrapolate the PMS of the largest inclusion in a reference area of steel sample, \( A_{\text{ref}} \). In this study, the \( A_{\text{ref}} \) value was decided as 1000 mm\(^2\), which is 1000 times larger than the \( A_{0} \) value. The PMS in the reference area in steel sample can be determined from the \( y \) value which was calculated for \( A_{\text{ref}} \) from Eqs. (2) and (3).

\[
y = -\ln\left( \frac{T-1}{T} \right) \quad (2)\]

\[
T = \frac{A_{\text{ref}}}{A_{0}} \quad (3)
\]

where \( y \) is the reduced variate, \( T \) (\( =1 \) 000 in this study) is the return period for the expected area. The standard errors (SE) of the PMS values were also calculated according to the ASTM E2283-03 standard. The detailed calculation methods of the ML method and SE values are also described elsewhere in the previous study.

In all the samples, a square root of an inclusion area, \( \sqrt{\text{area}_{\text{max}}} \), was applied as a size parameter of inclusions. The typical inclusions observed in different samples of Steel A and Steel B are spherical, as shown in Fig. 1. Therefore, for the samples, the size parameter of each inclusion was determined using a measured diameter of an inclusion, \( d \), as follows:

\[
\sqrt{\text{area}_{\text{max}}} = \sqrt{\pi \times \frac{d}{2}} \quad (4)
\]

In addition, the PSD of inclusions in the slab samples were measured in 2D by using LOM for estimation of the PMS. For the inclusions smaller than 5 \( \mu \)m, the total observed area on each slab sample was 5.6 mm\(^2\) which corresponds to 80 photos by the magnification of 200 times. All inclusions larger than 0.5 \( \mu \)m were counted and measured on a cross section. Total number of measured inclusions was in the range from 301 in the Steel A to 302 in the Steel B. On the other hand, the population density of larger inclusions (> 5 \( \mu \)m) in 100 mm\(^2\) was also counted at a magnification of 200. Their diameters were measured in order to obtain enough number of inclusion for statistical analysis. In the Steel A and B, 24 and 146 inclusions were counted in 100 mm\(^2\), respectively. These obtained data were plotted on a same figure by normalizing the inclusion population density per 1

![Fig. 1. Typical inclusions in (a) Steel A and (b) Steel B.](image)
mm². Some researchers⁹,¹⁰,¹⁶,¹⁷) reported that the PSD of inclusion obeyed arithmetical distribution function, such as Log-Normal, Exponential or Rayleigh distribution function. In this study, approximation by using Exponential function was attempted as for the regression lines for the obtained PSD data. The probable maximum inclusion size in a larger area was extrapolated by using the obtained regression line according to Eq. (5):

\[ N_A = a \cdot \exp(-b \cdot d) \]  

where \( N_A \) is the inclusion population density on a cross section, \( a \) and \( b \) are the regression coefficients determined by the size distribution and \( d \) is the inclusion diameter.

In addition, the distributions of the inclusions in the slab samples may be affected by the dendrite structures. Therefore, solidification structures were revealed by an electrical etching method using 10 mass% oxalic acid aqueous solution with a current density of 1 A/cm² for 1 minute at a room temperature.

3. Results and Discussion

3.1. Inclusion Compositions in the Slab Samples

The compositions of typical inclusions in the slab samples were plotted on the ternary diagram, as shown in Fig. 2. As can be seen, the inclusions in the Steel A are mostly a single phase of MgO or composite of dual phases of MgO and CaO–Al₂O₃, while those in the Steel B consists of SiO₂–MgO–CaO–Al₂O₃ complex oxides. A typical element distribution of an inclusion in the Steel A is shown in Fig. 3(a). It can be seen that the core of this inclusion is almost pure MgO, while a surface layer consists of Ca and Al oxides. This inclusion is similar to the ones observed in the previous studies.¹³ Meanwhile, the major components such as SiO₂, CaO, MgO and Al₂O₃ are detected in an inclusion in the Steel B with homogeneous distribution as understood in Fig. 3(b). This inclusion is similar to those reported in the previous study.²³

3.2. Effect of Dendrite Structures of the Slab Samples on Inclusion Distributions

For as-cast steels with high cleanliness, the compositions, distributions and morphology of inclusions are intimately related to the dendrite structures of the steels. Therefore, the dendrite structures for the Steel A and B were observed as shown in Figs. 4(a) and 4(b), respectively. One can clearly see the acicular type ferrite with directional growth of the primary dendrite arms. In the present study, it was assumed that inclusions were homogeneously distributed. This assumption was attributed to the following two reasons:

One is due to the avoidance of the observed positions where the both columnar structures met which grew from the narrow and the broad surfaces. The other is the relatively homogeneous dendrite structures for the both steels and the two structures are quite identical to each other. The secondary dendrite arm spacing was measured as 15 to 20 μm at around 2 mm from the surfaces of the both steels. These facts can be obviously observed from Fig. 4. Importantly, we have confirmed that the inclusions are distributed either at the inter-dendritic regions of the primary or secondary dendrite arms.

As for the morphology of the inclusions, globular shape is assumed for every inclusion as represented by Figs. 3(a) and 3(b). This is strongly related to the inclusion compositions; the steels contain Mg and Ca as realized in Table 1 resulting in the condensation of those elements at the regions between the dendrites. It is known that the condensation at the local terminal solidification positions affect the crystallography of the inclusions.¹⁸ However, in this study, this condensation finally causes the formation or maintenance of the amorphous inclusions containing CaO and MgO. Those are composed of CaO–Al₂O₃ surrounding MgO for the Steel A and SiO₂–MgO–CaO–Al₂O₃ for the Steel B. The CaO–Al₂O₃ and the SiO₂–MgO–CaO–Al₂O₃ oxides are considered to be molten in the molten steels, and solidified as amorphous. It should be noted that this condensation particularly affects the secondary inclusions which

| SEI | O | Mg | Al | Si | Ca |
|-----|---|----|----|----|----|
| (a) Steel A | ![Image](image1.png) | ![Image](image2.png) | ![Image](image3.png) | ![Image](image4.png) | ![Image](image5.png) |
| (b) Steel B | ![Image](image6.png) | ![Image](image7.png) | ![Image](image8.png) | ![Image](image9.png) | ![Image](image10.png) |

Fig. 2. Compositions of typical oxide inclusions in the slab samples.

Fig. 3. Distribution of main elements in inclusions in (a) Steel A and (b) Steel B.
are formed at the inter-dendritic regions during solidification of the steels.

3.3. Extreme Value Distribution (EVD)

Figure 5 shows the EVD of inclusions obtained experimentally in the molten steel and the slab samples from the both heats. The EVD lines for the Steel A samples have steeper slopes than those for the Steel B. It can be seen in Fig. 5(a) that the largest size of inclusions in the observed unit areas of the molten steel sample for the Steel B ranges from 3 to 13 μm, while that for the Steel A is from 1 to 7 μm. Similar tendency is obvious for the slab samples in Fig. 5(b). Though the maximum sizes of inclusions observed in the slab samples (13 and 17 μm for the Steel A and Steel B, respectively) are much larger in comparison with those in the molten steel, it can be seen that the points of maximum size inclusions significantly deviated from the obtained EVD lines. This fact can be explained by the limit of an observed area for the largest inclusions, as shown in the previous article.⁸

The main parameters of the regression lines (such as slope and intercept) calculated from the EVDs are given in Table 2 along with the PMS of inclusions on a 1000 mm² reference area of steel samples (A_ref). It is apparent that the parameters of the EVD lines and values of PMS for inclusions in the molten steel and slab samples are in good agreement with each other for the both heats. For instance, the slope values of the regression lines for the Steel A are 0.52 and 0.51 for the molten steel and slab samples, respectively. The corresponding values for the Steel B are 0.38 and 0.39, respectively. The PMS values of inclusions for samples of the Steel A calculated for 1000 mm² are 8 ± 1.6 and 9 ± 1.7 μm for the molten steel and slab samples, respectively, as given in Table 1. The PMS values for the both samples in the Steel B (19 ± 3.6 μm) are significantly larger than those in the Steel A. This can be explained by the higher total oxygen content in the Steel B (28 mass ppm) than that in the Steel A (17 mass ppm). Another possible reason for this difference of the PMS values is considered to be the difference in the inclusion compositions, as shown in Figs. 2 and 3.

According to the obtained results, it can be assumed that the size of the largest inclusions did not significantly vary

![Fig. 4. Dendrite structures of (a) Steel A and (b) Steel B at 2 mm from the surfaces.](image)

![Fig. 5. EVD lines for inclusions in (a) the molten steel and (b) slab samples.](image)

### Table 2. Slopes and intercepts of regression lines and the probable maximum size of inclusions on a 1000 mm² area.

| Steel  | Sample  | Slope | Intercept | Probable maximum size (μm) | 2SE |
|--------|---------|-------|-----------|----------------------------|-----|
| A      | Molten steel* | 0.52  | –2.92     | 9                          | 1.7 |
|        | Slab    | 0.51  | –2.88     | 8                          | 1.6 |
| B      | Molten steel* | 0.38  | –2.80     | 19                         | 3.6 |
|        | Slab    | 0.39  | –3.03     | 19                         | 3.6 |

*: Molten steel sample taken from a tundish.
during continuous casting process. In principle, inclusions in molten steel could be taken away by floatation during staying in a tundish and a mould. However, in this study, the EVD of inclusions does not vary before and after the casting process. It can be considered that smaller inclusions are more difficult to be floated out from molten steel compared to larger ones.19) Hereby, most of inclusions ranging between 1 and 20 μm in a diameter may be trapped onto a solidified shell during continuous casting. Thus, the size of the largest inclusion in the slab of a given stainless steel grade could be estimated based on the PMS values of inclusions in the samples of molten steel taken from a tundish during casting. It is benefit for time saving to employ molten steel sample for measurement. A small steel sample can be, in general, more easily handled than a large slab to prepare a test specimen for measurements in 2D.

3.4. Particle Size Distribution (PSD)

The PSD in the slab samples of the both heats are shown in Fig. 6. The separately obtained distributions for the smaller and the larger inclusions than 5 μm show good sequentiality for both of the Steel A and B. This fact implies that these combined distributions can be treated as a unified distribution. The highest population is obviously seen in the size range between 0.5 and 2 μm in diameter. It can be seen that the PSD for the Steel B shows a peak at 1 μm, while the PSD for the Steel A shows sequentially decreasing tendency. The number of inclusions per unit area in the size of 0.5 μm for the Steel A is higher than that for the Steel B. In the region larger than 1 μm, the constant values decrease subsequently with an increase in particle size for the both samples. The number of inclusions with the size larger than 1 μm in the Steel B is considerably higher than in the Steel A. The higher population in the region of relatively large inclusions in the PSD of the Steel B is considered to bring the higher PMS of the Steel B than the Steel A by the EVD.

When comparing the arithmetical mean sizes of inclusions in the both steels, the mean value obtained from the Steel A is 0.8 μm. This mean value is smaller than that value of 1.3 μm for the Steel B. Moreover, the standard deviation values of the PSD are 1.6 and 4.0 for the Steel A and B, respectively. The Steel A gives smaller σd value compared to the Steel B, as well. According to these results, it can be concluded that most of inclusions in the Steel A distributed in a narrow band of smaller size ranges.

3.5. Comparison of Probable Maximum Size of Inclusions by EVD and PSD

The experimentally determined PSD in slab samples is plotted in a log-scale in Figs. 7(a) and 7(b) in which the exponential regression lines are shown for the Steel A and B, respectively. The distributions for the Steel A and B show mostly linear relationships. The constants of a and b in Eq. (5) are listed in Table 3. As discussed later in detail, several treatments are tried to have the largest predicted size as accurately as possible.

In the distribution of the Steel A, the largest size is deviated from the linear tendency if accounting for all the measured data. The PSD for the Steel B also contains slightly deviated data in the region of the larger sizes. The first attempt considering all the data provides the solid regression lines denoted as Line 1. Another two attempts have been

![Fig. 6. Particle size distributions of inclusions in slab samples.](image)

![Fig. 7. Regression lines determined by particle size distributions with the probable maximum sizes determined by EVD for (a) Steel A and (b) Steel B.](image)

| Steel | Regression lines* | a ** | b ** | Correlation coefficient | Probable maximum size (μm) |
|-------|-------------------|------|------|-------------------------|---------------------------|
| A     | Line 1            | 5.72 | 0.56 | –0.892                  | 16                        |
|       | Line 2            | 11.74| 0.78 | –0.955                  | 12                        |
|       | Line 3            | 13.34| 0.83 | –0.951                  | 11                        |
| B     | Line 1            | 5.06 | 0.36 | –0.927                  | 23                        |
|       | Line 2            | 5.92 | 0.40 | –0.935                  | 22                        |
|       | Line 3            | 7.31 | 0.45 | –0.946                  | 20                        |

*: Line 1, 2 and 3 were calculated by all the data, the data without one maximum size data and the data without two maximum data, respectively.

**: \( N(d) = a \exp(-bd) \)
The PMS values converted to diameter determined by the SEV should be plotted on Figs. 6(a) and 6(b). The \( A_{\text{ref}} \) value of 1.000 mm\(^2\) for the SEV analysis corresponds to an inclusion population probability of \( N_4 (d) = 0.001 \, \text{mm}^{-2} \). The PMS values for the Steel A and B are calculated from EVD as 10±1.8 and 22±4.1 \( \mu \text{m} \) in diameter \((d)\), respectively, by substituting the \( \text{area}_{\text{max}} \) values of 8 and 19 \( \mu \text{m} \) into Eq. (3). The largest predicted diameters of 10 and 22 \( \mu \text{m} \) are plotted at the \( N_4 (d) = 0.001 \, \text{mm}^{-2} \) on the X axis in Figs. 6(a) and 6(b), respectively. This treatment enables the comparison between the two methods equivalently.

The each regressed result is summarized in Table 3. The following statements explain the respective predicted values at 0.001 mm\(^{-2}\) of PMS values of 12 and 11 \( \mu \text{m} \). The solid lines denoted as Line 1 give 16 and 23 \( \mu \text{m} \). The solid lines denoted as Line 2 give 12 and 22 \( \mu \text{m} \). The solid lines denoted as Line 3 give 11 and 20 \( \mu \text{m} \).

The plots are provided to see how the PMS values are correlated to O content of the specimens as shown in Fig. 8. Apparently, the higher O content gives higher PMS values. Further, elimination of the largest value of the PMS denoted as Line 2 and 3 leads to the values closer to the PMS obtained from the EVD analysis. At this moment, it is difficult to determine which treatment gives us the PMS value closer to the truth. It is considered that further investigation is necessary on the relationship between PSD and EVD.

4. Conclusions

The probable maximum sizes (PMS) of inclusions were determined on a polished surface of AISI304 stainless steel samples with different oxygen contents of 17 and 28 mass ppm. The analysis by statistics of extreme values (SEV) was compared with the particle size distributions (PSD) to have the PMS of inclusions on a 1,000 mm\(^2\) reference area. The following specific conclusions were obtained:

1. The regression lines and PMS values calculated from extreme value distributions (EVD) in the molten steel samples agreed well with that in the slab samples for both heats.

2. In the case of the steel with higher oxygen content, the PMS denoted from the EVD analysis agreed reasonably well with that from the exponential approximation of PSD.

3. In the case of the steel with lower oxygen content, the heterogeneity of distribution of largest size inclusions on a sample surface increase significantly. As a result, the PMS determined from the EVD analysis do not agree with that from the PSD. In this case, the approximations with elimination of one or two largest inclusions show better agreement of the PMS predicted from EVD and PSD.

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