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Author(s)	Matsuura, Kiyotaka; Itoh, Youichi; Kudoh, Masayuki; Ohmi, Tatsuya; Ishii, Kuniyoshi
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# Three-dimensional Grain Size Distribution in SUS304 Stainless Steel

Kiyotaka MATSUURA, Youichi ITOH, Masayuki KUDOH, Tatsuya OHMI and Kuniyoshi ISHII

Faculty of Engineering, Hokkaido University, Kita-ku, Sapporo, Hokkaido, 060 Japan.

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We disintegrated an annealed SUS304 stainless steel bar into separate crystal grains, and measured the distribution of the volume-equivalent grain diameter (the three-dimensional grain size distribution) by weighing the individual grains. We also measured the distribution of the area-equivalent grain diameter (the two-dimensional grain size distribution) on a cross section of the steel bar. From this two-dimensional grain size distribution, the three-dimensional grain size distribution was calculated by the method previously proposed by the authors. The measured and the calculated three-dimensional grain size distributions were then compared. The measured three-dimensional grain size distribution was approximately log-normal. The variation coefficient of the distribution (the standard deviation divided by the average value) was 0.4, and the ratio of the maximum grain size to the average size was about 3.0. The calculated three-dimensional grain size distribution was very similar to the measured one, indicating that the method of calculation previously proposed by the authors yields accurate results.

KEY WORDS: grain size; grain size distribution; grain growth; grain boundary; stainless steel.

## 1. Introduction

The grain size of polycrystalline materials plays an important role in determining the physical properties of materials. These properties are often described in terms of the average grain size, as seen, for example, in the Hall–Petch formula for the yield strength of mild steels.<sup>1)</sup> However, when the variation in the grain size is large, descriptions of the physical properties of the material in terms of only the average grain size are not accurate. In this case, the grain size distribution may be used to predict the properties of the material. The grain size distribution represents the proportions at which the various crystal grain sizes are found within the material.

An image analyzer can measure the cross-sectional area of the grains on a cross section of the material. Because crystal grains are, in general, in equi-axed shape, the grain size can be defined as the volume-equivalent diameter,<sup>2)</sup> which is the diameter of the sphere whose volume is equal to that of the crystal grain. In this paper, the volume-equivalent grain diameter is called the three-dimensional grain size, and the area-equivalent grain diameter is called the two-dimensional grain size.

The authors have previously proposed a method to calculate the three-dimensional grain size distribution using the two-dimensional grain size distribution.<sup>3)</sup> The purpose of the present study is to experimentally verify the accuracy of this calculation.

## 2. Calculation

The calculation used to estimate the three-dimensional

grain size distribution from the two-dimensional grain size distribution has been described in detail in a previous paper.<sup>3)</sup> This calculation is broadly outlined below.

### 2.1. Distribution of Cross-sectional Diameters of the Grains

We used the twelve types of polyhedra shown in Fig. 1 to simulate the various shapes of crystal grains found in actual materials. The distribution of cross-sectional

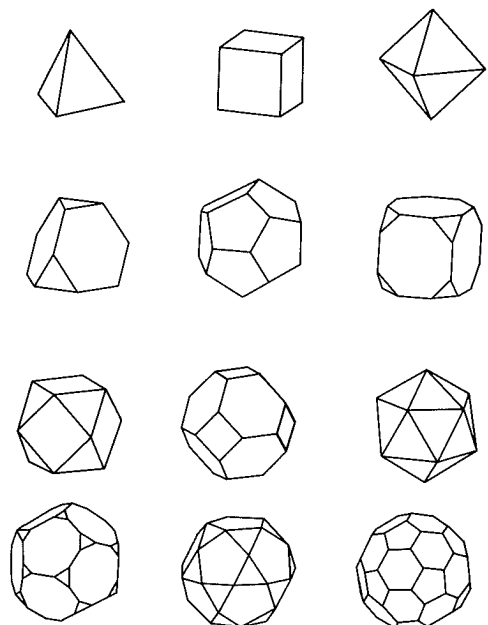


Fig. 1. Twelve types of polyhedra used as the grain model.

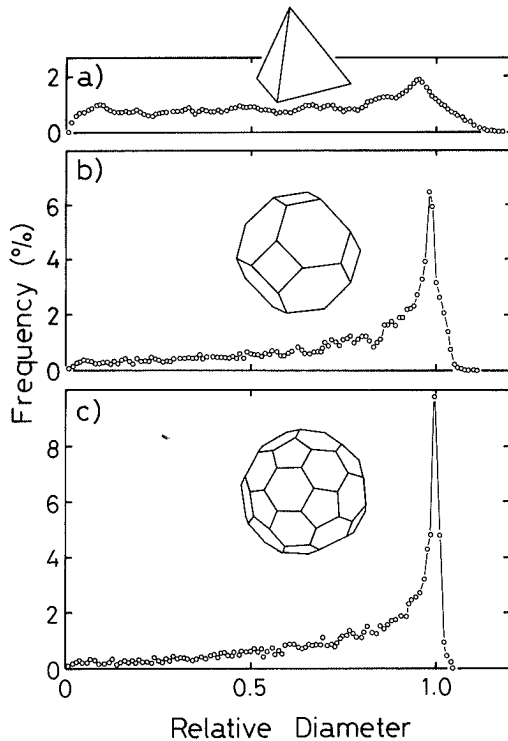


Fig. 2. Distributions of the cross-sectional diameter of polyhedra with (a) 4, (b) 14, and (c) 32 faces.

diameters for each polyhedron was investigated using a computer cutting method.<sup>4)</sup> Some examples of the results are given in Fig. 2.

The distribution of the cross-sectional diameter for any type of polyhedron shown in Fig. 1 can be approximately described by Eq. (1).

$$P = K \cdot \exp \left[ - \left\{ \ln \frac{d_{\text{Max}} - d}{d_{\text{Max}}(1 - d_p)} \right\}^2 \right] \dots\dots\dots(1)$$

where,  $d$ : relative cross-sectional diameter, which is the ratio of the area-equivalent diameter of the polygon-shaped cross section to the volume-equivalent diameter of the polyhedron

$P$ : probability density with which the relative cross-sectional diameter with the value  $d$  appears

$d_{\text{Max}}$ : maximum relative cross-sectional diameter

$d_p$ :  $d$ -coordinate of the peak of the probability density function

$K$ :  $P$ -coordinate of the peak of the probability density function.

Equations (2) through (4) show how the values of  $d_{\text{Max}}$ ,  $d_p$  and,  $K$  are related to the number of faces of the polyhedron.

$$d_{\text{Max}} = 0.31m^{-0.53} + 1.0 \dots\dots\dots(2)$$

$$d_p = -0.47m^{0.63} + 1.0 \dots\dots\dots(3)$$

$$K = 1.22m^{0.55} \dots\dots\dots(4)$$

where  $m$  is the number of faces of the polyhedron or the number of the grains adjacent to grain "i" in a polycrystalline material. The value of  $m$  is calculated from the size of grain "i" using Eq. (5).<sup>5)</sup>

$$m = 17D_i/D_{\text{Av}} - 3 \dots\dots\dots(5)$$

where  $D_i$  is the three-dimensional diameter of grain "i", and  $D_{\text{Av}}$  is the average grain diameter in the material. Equations (2) through (5) are derived from the results of the geometrical calculations for the polyhedra shown in Fig. 1.

**2.2. Composition of Cross-sectional Diameter Distributions**

Equations (1) through (5) are used to calculate the probability density functions  $P_1$  to  $P_n$  for grains with diameters  $D_1$  to  $D_n$ , respectively. The function  $P_i$  can be regarded as the two-dimensional grain size distribution measured on a cross section of material that is perfectly uniform with grains of diameter  $D_i$ , because  $P_i$  is the distribution of the cross-sectional diameter of grains with diameter  $D_i$ . If a material is composed of grains having diameters  $D_1$  to  $D_n$ , in the ratios  $F_1$  to  $F_n$ , respectively, then the two-dimensional grain size distribution measured on the section of this material will be the composite function given as Eq. (6).

$$P_c = \sum_{i=1}^n (f_i P_i) \dots\dots\dots(6)$$

where  $f_i$  is the composing ratio for the function  $P_i$ . Because the values of  $f_1$  to  $f_n$  are unknown, the function  $P_c$  in Eq. (6) can not be calculated. The function  $P_c$ , however, can be measured on a section of the material as the two-dimensional grain size distribution. Therefore, the composing ratio  $f_i$  can be calculated from  $P_c$  and  $P_i$ .

**2.3. Three-dimensional Grain Size Distribution**

The existing frequency  $F_i$  for the three-dimensional grain diameter  $D_i$  must be calculated, because the three-dimensional grain size distribution is presented as the relationship between  $D_i$  and  $F_i$ .

Because the composing ratio  $f_i$  in Eq. (6) describes the frequency at which a cross section of grain "i" appears on the section of the material,  $f_i$  is proportional to both  $D_i$  and  $F_i$ , as given in Eq. (7).

$$f_i = aD_i F_i \dots\dots\dots(7)$$

where  $a$  is the constant of proportion. From Eq. (7), the existing frequency of grain "i" is calculated such that:

$$F_i = f_i / (aD_i) \dots\dots\dots(8)$$

Because the summation of  $F_1$  to  $F_n$  is unity, the value of the constant in Eq. (7) is:

$$a = \sum_{j=1}^n (f_j / D_j) \dots\dots\dots(9)$$

Thus, the existing frequency of the grain with the diameter  $D_i$  is calculated as:

$$F_i = (f_i / D_i) / \sum_{j=1}^n (f_j / D_j) \dots\dots\dots(10)$$

**3. Experiment**

We machined a cylindrical sample with a diameter of 15 mm and a length of 60 mm from a commercial SUS304

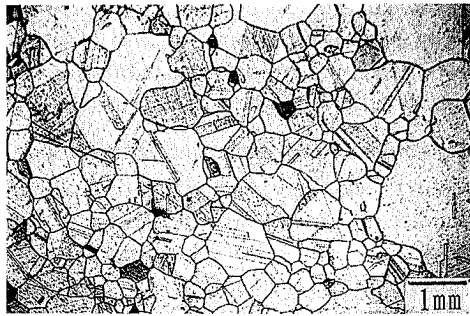
stainless steel bar. The chemical composition of this steel is given in **Table 1**. The crystal grains in the sample were coarsened by annealing the sample at 1 623 K for 250 ks in an argon atmosphere. The sample was then divided into two cylindrical samples, each 30 mm in length.

We sliced one of the sample transversely, at about 5 mm intervals. On the surface of the slices, the cross-sectional area of each grain was measured using an image analyzer. We then converted this measurement to the area-equivalent diameter and calculated the two-dimensional grain size distribution. As shown in **Fig. 3**, the annealing created twin crystals in the coarsened grains. The twin boundaries were carefully erased during the image analyzing process. We measured approximately 1600 crystal grains. We then calculated the three-dimensional grain size distribution from the two-dimensional grain size distribution using the method described above.

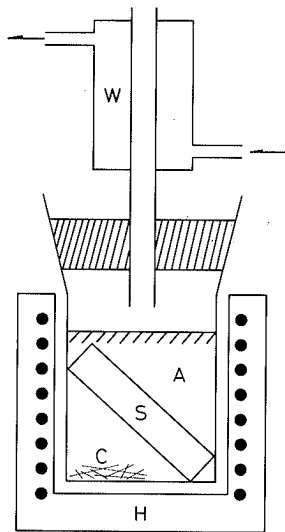
We disintegrated the other cylindrical sample into separate crystal grains in order to measure the three-

**Table 1.** Chemical composition of SUS304 stainless steel.

C	Si	Mn	P	S	Cr	Ni
0.07	0.48	1.20	0.024	0.025	18.46	8.22



**Fig. 3.** Optical micrograph of SUS304 stainless steel annealed at 1 623 K for 250 ks.



**Fig. 4.** Apparatus for the selective corrosion of grain boundaries of stainless steel.  
W: water, A: solution, S: sample, C: copper wire, H: heater.

dimensional grain size distribution by weighing the individual grains. We precipitated chromium carbide on the grain boundary by heat-treating the sample at 923 K for 7.2 ks in an argon atmosphere, prior to disintegration. The precipitation of the chromium carbide reduces the concentration of chromium near the grain boundary, thereby reducing the corrosion resistance of the grain boundary causing selective corrosion of the grain boundary. Preliminary examination confirmed that the grain growth at 923 K for 7.2 ks was negligible.

**Figure 4** shows the apparatus for the selective corrosion of the grain boundary of stainless steel. The sample was boiled for 173 ks in an aqueous solution of 10% copper sulfate with 10% sulfuric acid. According to Nagayama and Ishikawa,<sup>6)</sup> at the electric potential where  $\text{Cu}^+$  and  $\text{Cu}^{2+}$  are in equilibrium in a  $\text{Cu}-\text{Cu}^+-\text{Cu}^{2+}$  system, the grain boundary with a low chromium concentration is electro-chemically active and is resolved in the acid solution, while the matrix in the grain with a high chromium concentration is passive. A small amount of very thin copper wire was immersed into the acid solution to achieve equilibrium more rapidly.

After the selective corrosion of the grain boundary, we stressed by bending the sample to disintegrate it into separate crystal grains. We collected and weighed approximately 1000 grains. The volume-equivalent diameter of each grain was calculated from its weight and density. The density of the sample was determined to be  $7.89 \text{ g/cm}^3$ , according to the method of Archimedes. The measured three-dimensional grain size distribution was then compared to the calculated three-dimensional grain size distribution.

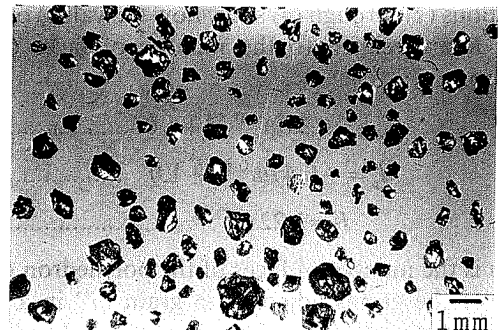
## 4. Results and Discussion

### 4.1. Disintegrated Crystal Grains

**Figure 5** shows the disintegrated crystal grains of SUS304 stainless steel annealed at 1 623 K for 250 ks. The corners and the edges of the polyhedron-shaped grains are very sharp. This findings implies that only the vicinity of the grain boundary was selectively corroded. Though the type of the polyhedron for the individual grains varies, it seems that the bigger the grain is, the more faces it has. This tendency supports Eq. (5).

### 4.2. The Relationship between Grain Size and Grain Shape

We investigated the quantitative relationship between grain size and grain shape in the disintegrated grains of



**Fig. 5.** Disintegrated crystal grains of SUS304 stainless steel.

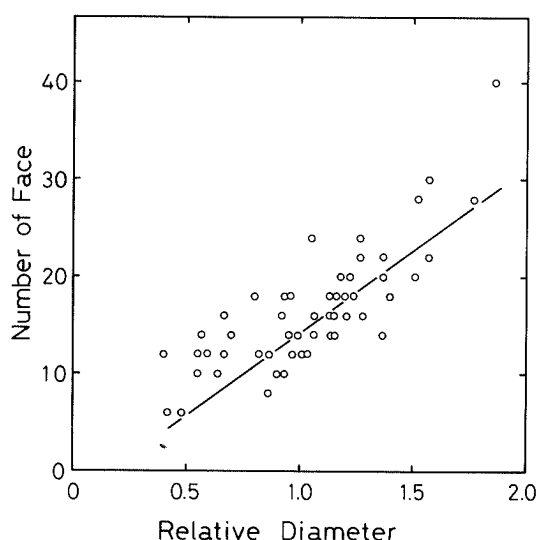


Fig. 6. Relationship between the grain size and the grain shape for a commercial Fe-62mass%Cr alloy.

SUS304 stainless steel. However, as shown in Fig. 5, the grain size was too small to count the number of faces on the grain. Instead, we measured the weight and number of faces of the large and clear-shaped crystal grains found in a commercial Fe-62mass%Cr alloy, which is marketed in the form of grains for adding chromium to iron-based alloys. This alloy contains 61.60% Cr, 0.060% C, 0.76% Si, 0.020% P, and 0.011% S. The average grain diameter of this alloy is 2.54 mm. Figure 6 shows the relationship between the grain size and the grain shape for this alloy. The straight line in the figure shows the relationship given by Eq. (5). The measured results are scattered along this straight line. Consequently, the relationship presented by Eq. (5), which is derived from the results of the geometrical calculations for the polyhedra shown in Fig. 1, is applicable to the crystal grains in practical materials.

#### 4.3. Comparison between Calculated and Measured Three-dimensional Grain Size Distributions

Figure 7(a) shows the two-dimensional grain size distribution measured on the cross section of the annealed SUS304 stainless steel. Figure 7(b) shows the three-dimensional grain size distribution calculated from the two-dimensional grain size distribution shown in Fig. 7(a), using the method proposed by the authors. Figure 7(c) shows the three-dimensional grain size distribution measured for the disintegrated grains of the same sample. The calculated and the measured three-dimensional grain size distributions are in fairly good agreement. The two-dimensional grain size distribution shown in Fig. 7(a) is different from the three-dimensional grain size distributions in Figs. 7(b) and 7(c). The distribution in Fig. 7(a) is wider than those in Figs. 7(b) and 7(c).

The average grain size, the standard deviation, and the variation coefficient are given in Table 2 for the three grain size distributions shown in Fig. 7. Each statistical value of the calculated three-dimensional grain size distribution is very similar to each value of the measured three-dimensional grain size distribution. The average grain size of the two-dimensional grain size distribution

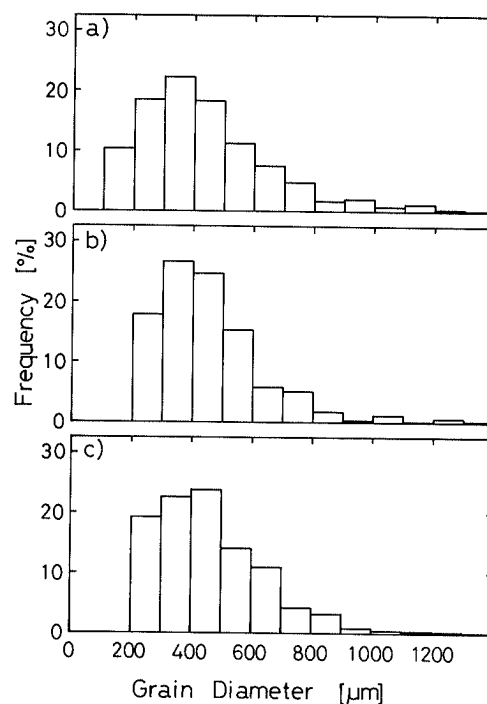


Fig. 7. Grain size distribution of SUS304 stainless steel annealed at 1623 K for 250 ks.  
(a) Distribution measured on the section.  
(b) Distribution calculated by the present method.  
(c) Distribution measured of the disintegrated grains.

Table 2. The average grain size  $D_{Av}$ , the standard deviation  $\sigma$  and the variation coefficient  $\sigma/D_{Av}$  of the grain size distributions shown in Fig. 7.

	$D_{Av}$ ( $\mu\text{m}$ )	$\sigma$ ( $\mu\text{m}$ )	$\sigma/D_{Av}$
Measured 2-D	446	245	0.55
Calculated 3-D	464	183	0.39
Measured 3-D	460	182	0.40

is smaller than those of the three-dimensional ones, while the standard deviation of the two-dimensional grain size distribution is larger than those of the three-dimensional ones. As seen in Fig. 2, the cross-sectional diameter ranges for 0 to  $d_{\text{Max}}$  when the three-dimensional diameter is unity, and in most cases, the cross-sectional diameter is smaller than the three-dimensional diameter. This may explain why the average grain size of the two-dimensional grain size distribution is smaller, and the standard deviation is larger (Table 2).

#### 4.4. Ratio of $D_{\text{Max}}/D_{Av}$

The maximum grain size was about 1400  $\mu\text{m}$  in all grain size distributions shown in Fig. 7. The ratio of the maximum grain size to the average grain size ( $D_{\text{Max}}/D_{Av}$ ) was 3.1, 3.0, and 3.0 for the two-dimensional, the calculated three-dimensional, and the measured three-dimensional grain size distributions, respectively.

Watanabe and Masuda<sup>7)</sup> calculated the effect of the residual pores on the grain size distribution in sintered metal compacts, and found that the ratio of  $D_{\text{Max}}/D_{Av}$  increases with the value  $pR_c/r$ , where  $p$  and  $r$  are the volume fraction and the average radius of the pores, respectively, and  $R_c$  is the critical radius of the grain (as

proposed by Hillert<sup>8)</sup>). Hillert regards  $R_c$  to be related to the average grain radius. According to the methods proposed by Watanabe and Masuda, the ratio of  $D_{Max}/D_{Av}$  is about 2.0 when no pore is included, and it increases from 2.5 to 3.3 when the value  $pR_c/r$  increases from 0.2 to 0.4. Hunderi and Ryum<sup>9)</sup> studied the stagnation in grain growth by computer simulation and found that the ratio of  $D_{Max}/D_{Av}$  was 3.0 for the stagnation condition due to the secondary phase particles.

It is unlikely that pores were included in the sample used in the present study, because the commercial stainless steel bar from which the sample was machined was not produced by a powder metallurgy process. On the other hand, the effect of the secondary phase particles, such as nitrides or oxides, should be considered. Nevertheless, the effect of such particles would be negligible, because they are almost entirely dissolved in the austenitic matrix at an annealing temperature of 1623 K. Therefore, according to the calculations of either Watanabe and Masuda or Hunderi and Ryum, the ratio of  $D_{Max}/D_{Av}$  in the present study should be about 2.0. The actual value, however, was about 3.

We previously studied the grain growth of SUS310 stainless steel and found that the variation coefficient of the grain size distribution increases during grain growth.<sup>10)</sup> The increase in the variation coefficient leads to the increase in the ratio of  $D_{Max}/D_{Av}$ . Watanabe<sup>11)</sup> reported that random high-angle boundaries tend to surround large grains, while low-angle boundaries and low- $\Sigma$  coincidence boundaries tend to surround small grains. Watanabe explained that low-angle boundaries and low- $\Sigma$  coincidence boundaries, which are generated at the early stage of recrystallization, are replaced by or modified to random high-angle boundaries during growth. Because the mobility of the high-angle boundaries is high, the difference in the grain size increases during grain growth resulting in the high ratio of  $D_{Max}/D_{Av}$ . The distribution of this grain boundary character<sup>11)</sup> is not taken into account in the calculation models proposed by either Watanabe and Masuda or Hunderi and Ryum. This may account for the higher ratio of  $D_{Max}/D_{Av}$  in the present study, than was predicted from the calculation.

#### 4.5. Type of Distribution

The grain size distributions shown in Fig. 7 are replotted in Fig. 8, which shows the normal probability plot of the cumulative frequency vs. the natural logarithmic grain size. All distributions are log-normal, because they are approximately linear in this figure. The calculated three-dimensional grain size distribution is in very good agreement with the measured three-dimensional grain size distribution. This result demonstrates that the calculation method proposed by Matsuura and Itoh<sup>3)</sup> is correct. The measured two-dimensional grain size distribution is obviously different from the three-dimensional grain size distributions. Consequently, the true grain size distribution cannot be

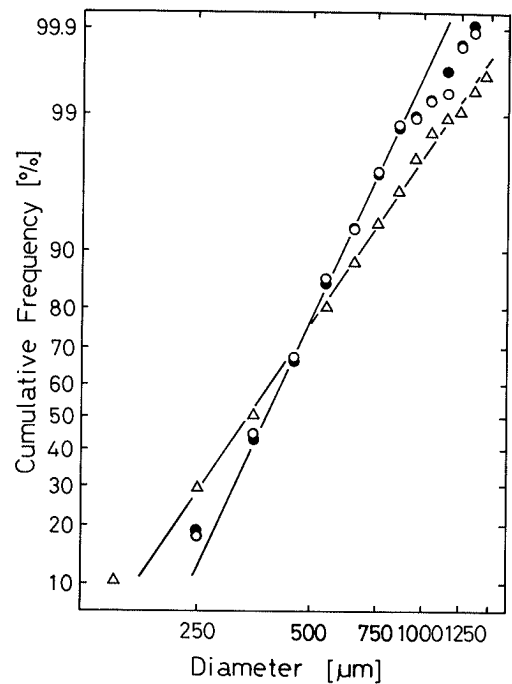


Fig. 8. Normal probability plot of cumulative frequency vs. natural logarithmic grain size.  
 $\Delta$  Distribution measured on the section.  
 $\circ$  Distribution calculated by the present method.  
 $\bullet$  Distribution measured of the disintegrated grains.

measured on the section of a material, but can be calculated accurately from the apparent grain size distribution measured on the section, using the method proposed by the authors.

#### 5. Conclusions

We disintegrated an annealed SUS304 stainless steel bar into separate crystal grains, and measured the three-dimensional grain size distribution by weighing the individual grains. We calculated the three-dimensional grain size distribution from the two-dimensional grain size distribution measured on the section of the steel, using the method previously proposed by the authors. The present results suggest that a fairly accurate three-dimensional grain size distribution can be calculated using this method.

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