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Evaluation of Grain Shape Distribution in Polycrystalline Materials

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We have proposed equations describing the numbers of faces, edges, and corners of a polyhedron-shaped crystal grain in terms of the volume-equivalent grain diameter, and we have developed a method to calculate the grain shape distribution in polycrystalline materials using these equations. We then experimentally confirmed that the proposed equations apply to crystal grains in actual polycrystalline materials and that the grain shape distribution can be calculated accurately using our method.

The present method was used to calculate the grain shape distribution in annealed SUS304 stainless steel. The average numbers of grain faces, edges, and corners were 14, 31, and 19, respectively; and their respective distribution ranges were 4-56, 6-139, and 4-86. The average surface area of the crystal grain per unit volume, the average edge length per unit volume, and the average number of corners per unit volume were 9 mm^{-1}, 61 mm^{-2}, and 175 mm^{-3}, respectively; and their respective ranges were 2-31 mm^{-1}, 6-304 mm^{-2}, and 18-863 mm^{-3}.

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I. Introduction

It is well known that the grain boundary in polycrystalline materials acts as the nucleation site of the phase transformation or recrystallization. The boundary of polyhedron-shaped crystal grains is classified into the following three parts: the face on which two grains make contact, the edge along which three grains make contact, and the corner at which four grains make contact. There are marked differences in the nucleation rates of transformed or recrystallized grains at the respective parts of the grain boundary[1].

Though the grain shape in actual polycrystalline materials varies and is quite complex, grain models based on simple shapes, such as spheres or tetrakaidecahedra, have been used to calculate grain growth[2] or phase transformation[3]. Calculations based on these models may yield incorrect results because the grain shape is extremely simplified.

Matsuura and Itoh recently proposed a new grain model, termed a composed polyhedron model, and applied this model both to the analysis of grain growth by computer simulation[4] and to the evaluation of three-dimensional grain size distribution[5]. In this model, the shape of a crystal grain is described as a type of polyhedron and the type for each grain is determined from the grain size.

Matsuura and Itoh[4] confirmed that the process of grain growth can be calculated more accurately using this model than by using the sphere model[2]. Matsuura[6] also confirmed that the three-dimensional grain size distribution calculated using this model is more accurate than that calculated using either the sphere model[7] or the tetrakaidecahedron model[8]. It is likely that the growth process of transformed or recrystallized grains that are nucleated on the grain boundary of the matrix can also be calculated more accurately using this model than by using the tetrakaidecahedron model[3]. In the latter model, all grains are assumed to be the same shape and therefore to have the same numbers of faces, edges, and corners of the grain boundary. The distribution of grain shape, namely the distribution of nucleation sites having different nucleation rates, should be taken into account when calculating phase transformation or recrystallization.

The purpose of the present study is to apply the composed polyhedron model to the evaluation of grain shape distribution, i.e., the distribution of the numbers of grain faces, edges, and corners.

II. Procedure

The present authors previously proposed a method to calculate the distribution of the volume-equivalent grain diameter in polycrystalline materials from the distribution of the area-equivalent cross-sectional grain diameter measured on the section of the material[5]. Therefore, provided that the numbers of faces, edges, and corners of the grain are described in terms of its volume-equivalent diameter, the grain shape distribution can be calculated from the measured distribution of the area-equivalent cross-sectional grain diameter.

1. Grain face

The number of faces of a crystal grain is calculated from its volume-equivalent diameter, as shown in eq. (1). We previously determined this relationship[4] from the geometrical calculations for the types of polyhedra

shown in Fig. 1, based on the composed polyhedron model.

$$F_i=17D_i/D_{av}-3 \quad (1)$$

where F_i and D_i are the number of faces and the volume-equivalent diameter of a grain named "i", respectively, and D_{av} is the average grain diameter in the polycrystalline material.

The area of the grain face per unit volume, A_i, is described by eq. (2), where one face is shared by two grains:

$$A_i=S_i/2V_i \quad (2)$$

where S_i and V_i are the surface area and the volume of grain "i", respectively. The ratio of S_i/V_i can be calculated from F_i and D_i, as shown in eq. (3). We also obtained this relationship previously[4] from the geometrical calculation based on the composed polyhedron model.

$$S_i/V_i=(9/F_i+6)/D_i. \quad (3)$$

From eqs. (1) through (3), the area of the grain face per unit volume for grain "i" is described in terms of its volume-equivalent diameter and the average grain diameter, as described in eq. (4).

$$A_i=\{9/(17D_i/D_{av}-3)+6\}/2D_i. \quad (4)$$

Consequently, the distributions of the number of faces and of the face area per unit volume are calculated from eq. (1) and eq. (4), respectively. These distributions are given as the correlations of (F_i, f_i) and (A_i, f_i), respectively, where f_i is the frequency of the existence of crystal grains having a volume-equivalent diameter of D_i in the grain size distribution, given as the correlation of (D_i, f_i). The grain size distribution is calculated from the distribution of the area-equivalent grain diameter, using the method proposed previously[5]. The distribution of the area-equivalent grain diameter can be easily measured on the section of the material, using an image analyzer.

2. Grain edge and grain corner

We investigated the relationship between the number of faces, F_i, and the number of edges, E_i, for the various polyhedra shown in Fig. 1. The result is shown in Fig. 2. This relationship is regressed as:

$$E_i=2.6F_i-5.4. \quad (5)$$

From eqs. (1) and (5),

$$E_i=44.2D_i/D_{av}-13.2. \quad (6)$$

Equation (7) is Euler's relationship among the numbers of faces, edges, and corners of a polyhedron.

$$F_i-E_i+C_i=2 \quad (7)$$

where C_i is the number of corners. From eqs. (5) and (7),

$$C_i=1.6F_i-3.4. \quad (8)$$

From eqs. (1) and (8),

$$C_i=27.2D_i/D_{av}-8.2. \quad (9)$$

Consequently, the distributions of the numbers of edges and corners are calculated from eq. (6) and eq. (9), respectively. These distributions are given as (E_i, f_i) and (C_i, f_i), using the frequency f_i in the grain size distribution (D_i, f_i).

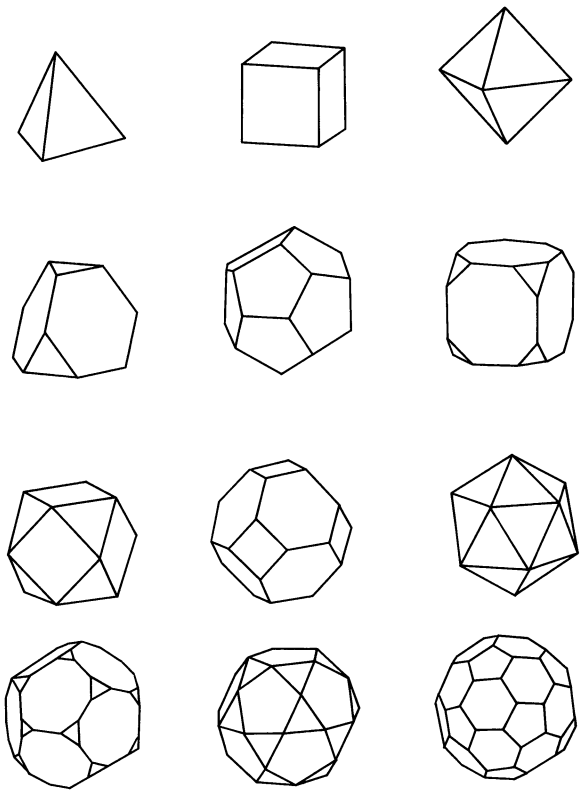

Fig. 1 Various types of polyhedra used in the composed polyhedron model.

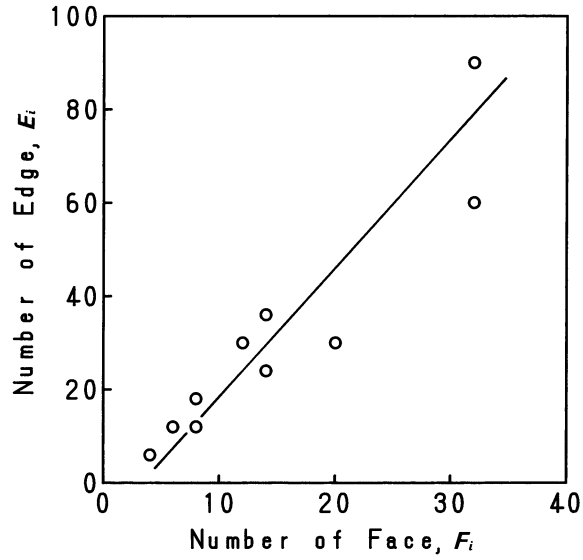

Fig. 2 Relationship between the number of faces and the number of edges of the polyhedra shown in Fig. 1.

Next, we calculate the edge length per unit volume and the corner number per unit volume.

Figure 3 shows the relationship between the number of edges of a polyhedron and its normalized volume. This

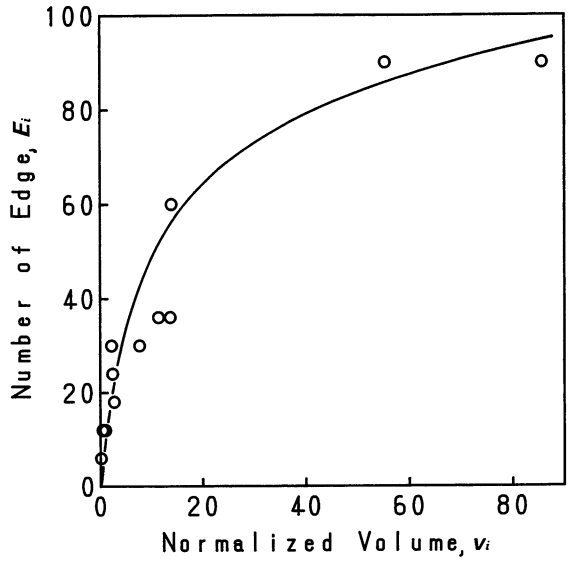

Fig. 3 Relationship between the number of edges of a polyhedron and the volume normalized by the cube of the edge length.

relationship is regressed as:

$$E_i=14.9v_i^{0.42} \tag{10}$$

where v_i is the normalized volume and is defined as:

$$v_i=V_i/e^3 \tag{11}$$

where V_i and e are the volume and edge length of the polyhedron, respectively.

Figure 4 shows the relationship between the number of corners of a polyhedron and the normalized volume. This relationship is regressed as:

$$C_i=8.7v_i^{0.44}. \tag{12}$$

The relationship between the number of faces of a polyhedron and the normalized volume is derived from different equations, as follows. From eqs. (5) and (10),

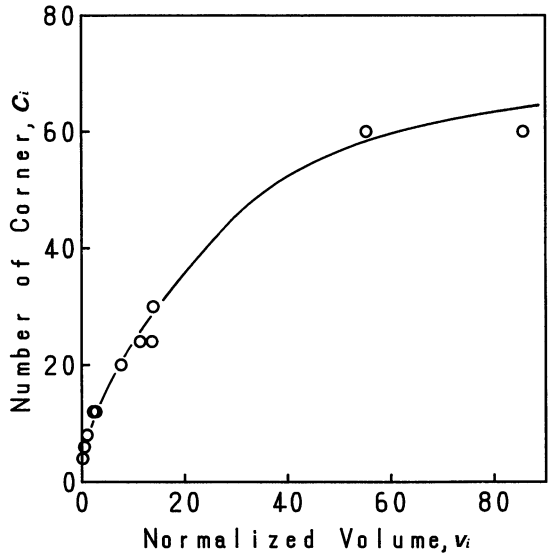

Fig. 4 Relationship between the number of corners of a polyhedron and the volume normalized by the cube of the edge length.

$$F_i=5.73v_i^{0.42}+2.1. \tag{13}$$

By contrast, from eqs. (8) and (12),

$$F_i=5.44v_i^{0.44}+2.1. \tag{14}$$

Since both eqs. (13) and (14) present the relationship between F_i and v_i for the polyhedra shown in Fig. 1, they must be equivalent to each other. However, they are slightly different. This difference may have arisen from the regression errors of eqs. (10) and (12). Therefore, we averaged the values of the coefficients and the exponents in eqs. (13) and (14), and modified these equations as:

$$F_i=5.59v_i^{0.43}+2.1. \tag{15}$$

Therefore, eq. (10) is modified as eq. (16) from eqs. (5) and (15), and eq. (12) is modified as eq. (17) from eqs. (8) and (15).

$$E_i=14.5v_i^{0.43} \tag{16}$$

$$C_i=8.9v_i^{0.43}. \tag{17}$$

The length of the grain edge per unit volume, L_i, is given by eq. (18), because one edge is shared with three grains:

$$L_i=E_ie/3V_i. \tag{18}$$

From eqs. (11), (16) and (18),

$$L_i=14.5(V_i/e^3)^{0.43}e/3V_i. \tag{19}$$

According to Smith's model[9], when all grains have the same size, the grain shape is tetrakaidecahedral. Therefore, it may be assumed that grains of average size are tetrakaidecahedral in shape. The volume of a tetrakaidecahedron described in terms of the edge length is $11.3e^3$, while that described in terms of the volume-equivalent diameter is $\pi D_{av}^3/6$. Therefore,

$$e=0.36D_{av}. \tag{20}$$

Substituting eq. (20) and $V_i=\pi D_i^3/6$ into eq. (19), we obtain the edge length per unit volume as:

$$L_i=3.32(11.2D_i^3/D_{av}^3)^{0.43}D_{av}/D_i^3. \tag{21}$$

The number of grain corners per unit volume, N_i, is given by eq. (22), because one corner is shared with four grains:

$$N_i=C_i/4V_i. \tag{22}$$

From eqs. (11), (17), (20), and (22),

$$N_i=4.25(11.2D_i^3/D_{av}^3)^{0.43}/D_i^3. \tag{23}$$

From eqs. (6) and (9), we can derive different equations which describe the edge length and the corner number per unit volume, as eqs. (24) and (25):

$$L_i=0.23(44.2D_i/D_{av}-13.2)D_{av}/D_i^3 \tag{24}$$

$$N_i=0.48(27.2D_i/D_{av}-8.2)/D_i^3. \tag{25}$$

These equations are essentially similar to eqs. (21) and (23), because the similar values of L_i and N_i can be ob-

tained from eqs. (21) and (24) and from eqs. (23) and (25), respectively. However, a peak value of L_i and N_i appears in eqs. (24) and (25), because they include difference terms in the parentheses. This is not adequate as the relationship between D_i and L_i or between D_i and N_i.

Consequently, the distributions of edge length per unit volume and corner number per unit volume are calculated from eq. (21) and eq. (23), respectively. They are given as (L_i, f_i) and (N_i, f_i), respectively, using the frequency f_i in the grain size distribution, (D_i, f_i).

III. Discussion

In Chapter II, we described a method to calculate the grain shape distribution in polycrystalline materials. In this chapter, we provide experimental verification of the validity of this method.

1. Grain model

The essential equation to calculate the grain shape distribution is eq. (1). This is derived from the geometrical calculations for the various polyhedra shown in Fig. 1, based on the composed polyhedron model. In this equation, the number of faces of a crystal grain is presented in terms of its volume-equivalent diameter normalized by the average grain diameter. We will examine here whether this equation is applicable to actual crystal grains.

We corrected 70 clear-shaped separate crystal grains of a commercial Fe–62 mass%Cr alloy, which is marketed in the form of grains for adding chromium to iron-based alloys. Figure 5 shows the crystal grains of this alloy. We microscopically counted the number of faces, of each crystal grain, and measured its weight. It was assumed that half of the face number is visible when observed from one side. The weight of each grain was converted into its volume-equivalent diameter using the density of this alloy, 7.34 Mg/m^3. The average grain diameter was 2.54 mm.

Figure 6 shows the relationship between the number of grain faces and the volume-equivalent grain diameter normalized by the average diameter. The plotted marks and the straight line in the figure indicate the measured results of the commercial Fe–Cr alloy and eq. (1), respectively.

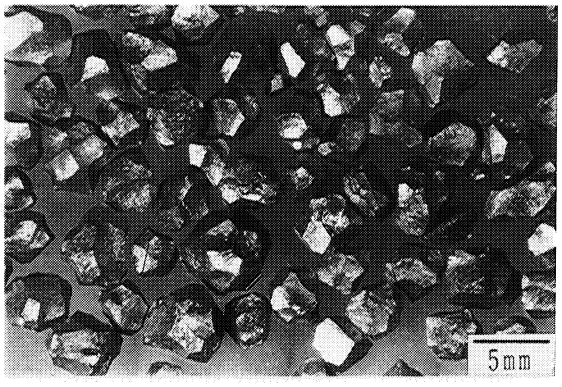

Fig. 5 Crystal grains of a commercial Fe–62 mass%Cr alloy.

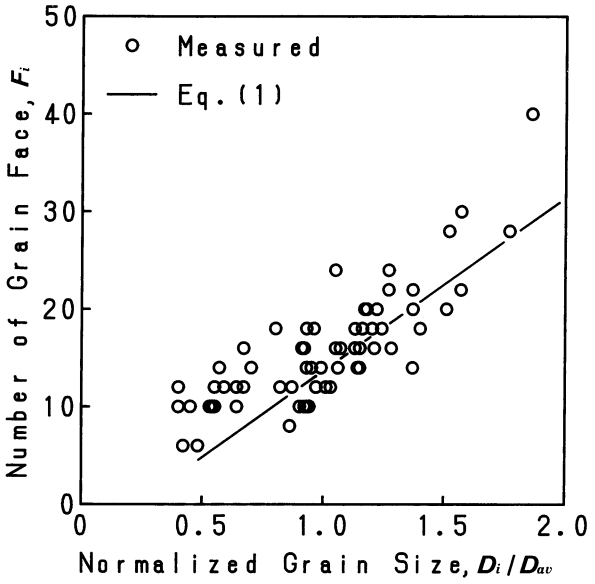

Fig. 6 Relationship between the number of grain faces and the volume-equivalent diameter normalized by the average diameter. Plotted marks indicate the measured results of an Fe–Cr alloy and the straight line indicates eq. (1).

As expected, the measured results scatter along the straight line. This result indicates that eq. (1) is applicable to crystal grains in actual materials.

2. Grain shape distribution

In order to experimentally verify the validity of the method to calculate grain shape distribution, we made a model material using crystal grains of a commercial Fe–Cr alloy, whose grain shape distribution was measured directly. We cast the crystal grains with a molten Al–Cu alloy into a cylinder measuring 80 mm in height and 16 mm in diameter, and calculated the grain shape distribution in the ingot of the model material using the method described in Chapter II. We then compared the calculated distribution with the measured distribution for the separate grains prior to making the ingot.

We sliced the cylindrical ingot transversely at about 5-mm intervals, and used an image analyzer to measure the distribution of the area-equivalent cross-sectional grain diameter for about 140 grain sections on the surface of the slices. The distribution of the volume-equivalent grain diameter was calculated using the distribution of the area-equivalent grain diameter[5]. From the distribution of the volume-equivalent grain diameter, the distributions of the numbers of faces, edges, and corners of the grain were calculated using the method described in Chapter II.

Figure 7 shows the grain size distribution of the Fe–Cr alloy dispersed in the matrix of Al–Cu alloy. The solid circles in the figure indicate the distribution of the volume-equivalent grain diameter, which was converted from the grain weight measured for the separate grains prior to making the model ingot. The open circles indicate the distribution of the volume-equivalent grain diameter calculated using our method[5]. As seen in this

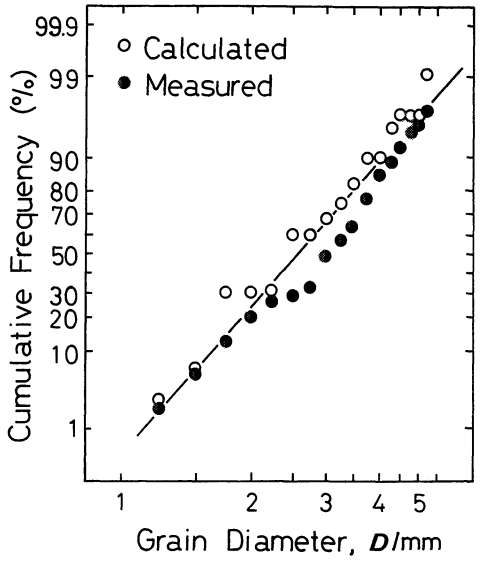

Fig. 7 Calculated and measured distributions of the volume-equivalent grain diameter of an Fe-Cr alloy dispersed in the matrix of an Al-Cu alloy.

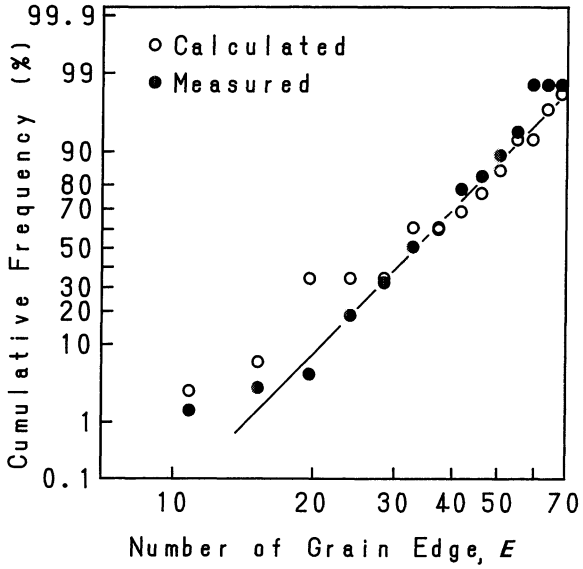

Fig. 9 Calculated and measured distributions of the number of grain edges of an Fe-Cr alloy dispersed in the matrix of an Al-Cu alloy.

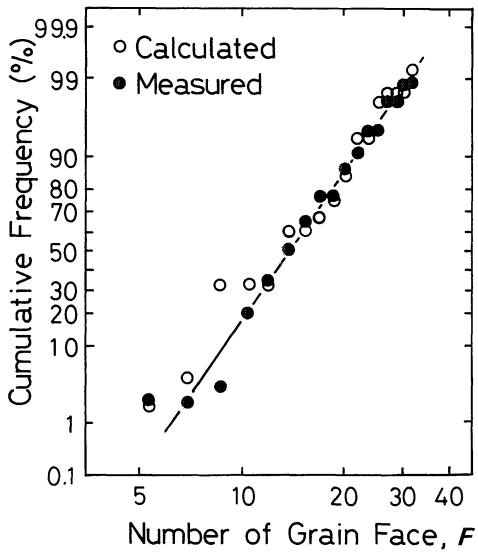

Fig. 8 Calculated and measured distributions of the number of grain faces of an Fe-Cr alloy dispersed in the matrix of an Al-Cu alloy.

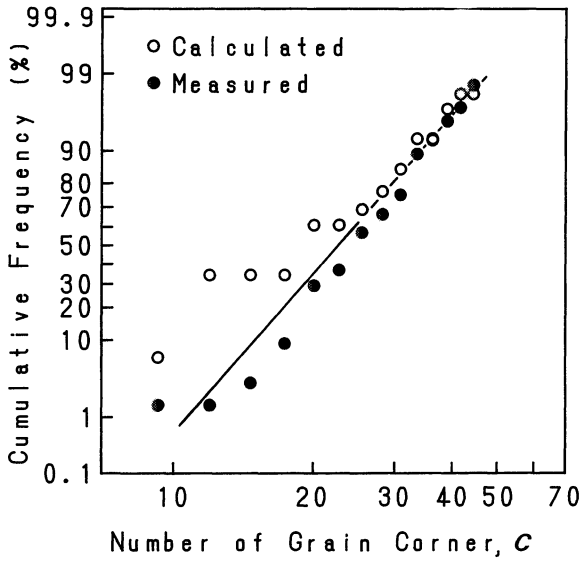

Fig. 10 Calculated and measured distributions of the number of grain corners of an Fe-Cr alloy dispersed in the matrix of an Al-Cu alloy.

figure, the calculated distribution is very similar to the measured distribution. From the calculated distribution of the volume-equivalent grain diameter, we calculated the grain shape distribution using the method described in Chapter II.

The distributions of the numbers of grain faces, edges, and corners are shown in Figs. 8–10, respectively. As seen in these figures, the calculated distributions shown by the open circles are generally similar to the measured distributions shown by the solid circles. This confirms that the grain shape distribution in polycrystalline materials can be calculated using the method described in Chapter II. The difference between the calculated and measured results, however, seems to increase gradually as the figure number increase from 8 to 10. The distribution of the edge number shown in Fig. 9 is calculated from the results of the distribution of the face number shown in Fig. 8, using eqs. (1) and (5). The distribution of the corner number shown in Fig. 10 is calculated from the results of the distribution of the face number shown in Fig. 8 and the distribution of the edge number shown in Fig. 9, using eqs. (1), (5) and (7). These equations hold approximately for various types of regular and semi-regular polyhedra shown in Fig. 1, but they do not hold strictly for individual crystal grains in actual polycrystalline materials. Therefore, the difference between the calculated and measured results increases in the order described above.

3. Application of the present method

Using the method described in Chapter II, we cal-

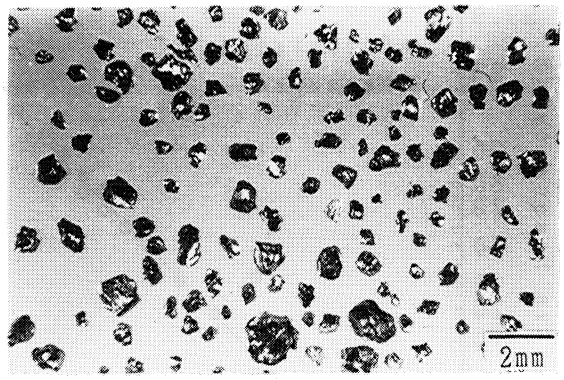

Fig. 11 Disintegrated crystal grains of SUS304 stainless steel.

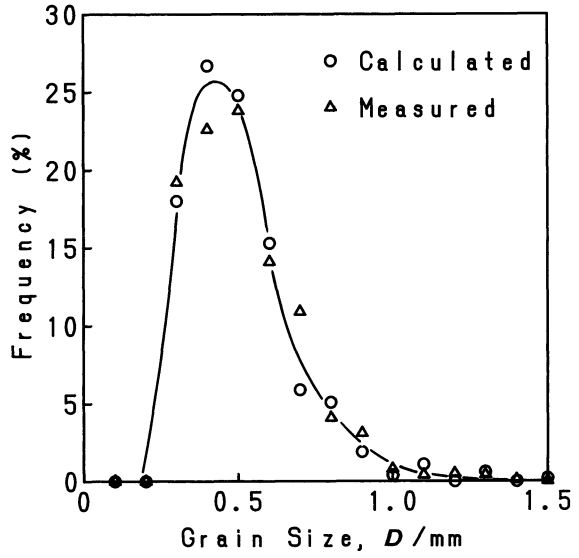

Fig. 12 Calculated and measured distributions of the volume-equivalent grain diameter of SUS304 stainless steel.

culated the grain shape distribution in SUS304 stainless steel annealed at 1623 K for 250 ks. We measured the distribution of the area-equivalent cross-sectional grain diameter on the section of the sample, and converted it into the distribution of the volume-equivalent grain diameter using our method[5]. We also measured the distribution of the grain weight for approximately 1000 disintegrated grains of the same sample, and converted it into the distribution of the volume-equivalent grain diameter using the density of the steel, 7.89 Mg/m^3. The disintegration of the stainless steel was carried out using a method involving selective corrosion of the grain boundary[10]. Figure 11 shows the disintegrated grains.

Figure 12 shows the calculated and the measured distributions of the volume-equivalent grain diameter of the stainless steel. The calculated and the measured average grain diameters are 0.45 mm and 0.46 mm, respectively. This similarity indicates that our method of calculation is quite accurate.

We used the method described in Chapter II to calculate the grain shape distribution from the calculated distribution of the volume-equivalent grain diameter. The result is shown in Fig. 13. The numbers of grain faces, edges, and corners range from 4 to 56, from 6 to 139, and from 4 to 86, respectively; with the average numbers being 14, 31, and 19, respectively.

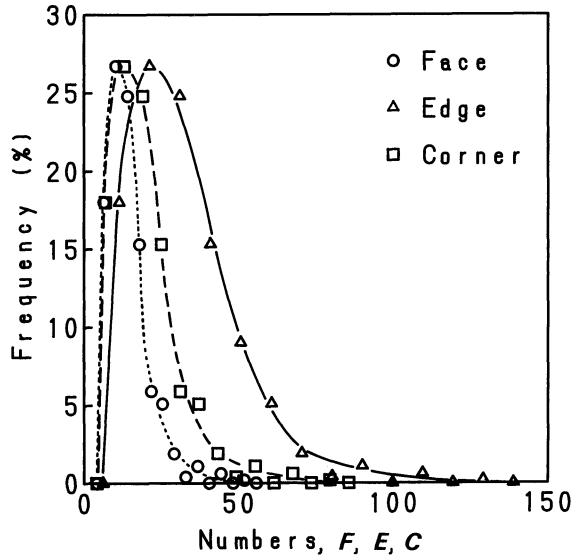

Fig. 13 Calculated distributions of the numbers of grain faces, edges, and corners of SUS304 stainless steel.

According to Rhines and Patterson[11], the number of faces per grain and the number of edges per grain face ranged from 3 to 60 and from 2 to 20, respectively, and the average numbers were 9 and 5, respectively, for aluminum. According to Williams and Smith[12], the numbers ranged from 6 to 23 and from 3 to 8, respectively, and the average numbers were 12 and 5, respectively, for Al–Sn alloy. The former authors counted the numbers for about 100 grains, while the latter authors counted them for 91 grains.

In the present study, the grain size distribution measured for about 1000 disintegrated grains was quite similar to the calculated grain size distribution, as seen in Fig. 12. From this grain size distribution, the distributions of the numbers of faces, edges, and corners were calculated. The distribution range of the number of faces in the present study is similar to that in Ref. (11). The distribution range and the average value of the number of edges per grain face in the present study are calculated to be 3–5 and 4.4, respectively, considering that one edge is shared with two faces. The results are similar to those in Ref. (12). According to Smith's model[9], the average number of faces is 14. Smith's model has been accepted by many researchers, and his model was supported by the present result. But the numbers in Refs. (11) and (12) are smaller than 14.

Figure 14 shows the distribution of the surface area per unit volume of the crystal grains in the stainless steel. The value ranges from 2 to 31 mm^{-1}, and the average value is 8.7 mm^{-1}. We calculated the distributions of the edge length and the corner number per unit volume using the results in Fig. 14, eq. (21), and eq. (23). The results

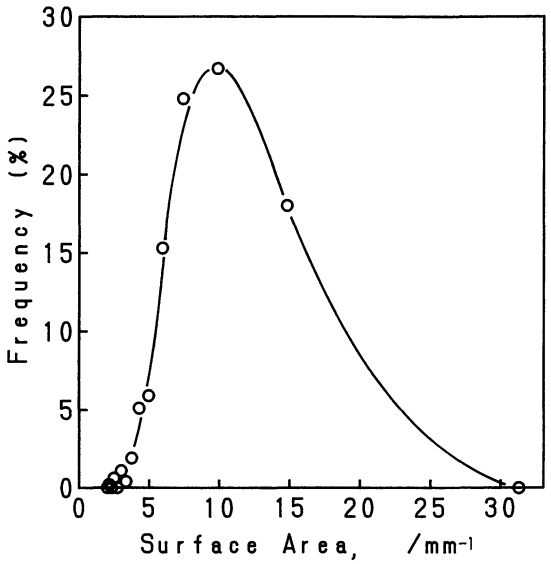

Fig. 14 Calculated distribution of the surface area per unit volume in SUS304 stainless steel.

were: 6–304 mm^{-2} and 18–863 mm^{-3}, respectively; with average values of 61 mm^{-2} and 175 mm^{-3}, respectively.

IV. Conclusions

We have proposed equations describing the numbers of faces, edges, and corners of a polyhedron-shaped crystal grain in terms of its volume-equivalent diameter. We then used these equations to develop a method to calculate the grain shape distribution in polycrystalline materials.

The validity of the grain model used in this method and the accuracy of the calculated grain shape distribution were verified experimentally. The present results confirm that the equations describing the relationship between grain size and grain shape are applicable to crystal grains in actual polycrystalline materials, and that grain shape distribution can be accurately calculated using the present method.

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