Microstructures of Al-Al_3Ti functionally graded materials fabricated by centrifugal solid-particle method and centrifugal *in situ* method

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Methods of fabrication by centrifugal casting for functionally graded materials (FGMs) can be classified into two categories on the basis of the relationship between the process temperature and the liquidus temperature of a master alloy. They are the centrifugal solid-particle method and centrifugal *in situ* method, which could be carried out at process temperatures lower and higher than the liquidus temperature of the master alloy, respectively. In a previous study, it was found that the microstructures of Al-Al_3Ti FGMs fabricated by the centrifugal *in situ* method processed at 1600 °C were different from those fabricated by the centrifugal solid-particle method processed at 800 °C. Although it is expected that the FGMs fabricated by the centrifugal *in situ* method processed at approximately the liquidus temperature should show extraordinary microstructures, those microstructures have not been observed. In this study, the microstructures of Al-Al_3Ti FGMs fabricated at 1000 °C (centrifugal solid-particle method) and 1200 °C (centrifugal *in situ* method) were investigated.

1. Introduction

Functionally graded materials (FGMs) are inhomogeneous composite materials that are tailored to have spatially changed properties to take advantage of the different behaviors of their constituents [1-5]. The centrifugal casting (centrifugal method) has been proven to be effective for the fabrication of ring- or pipe-shaped FGMs [6-9]. The compositional gradient formed by the centrifugal casting is determined by the difference in density between particles and a molten metal, the applied G number (the ratio of centrifugal force to gravity), the particle size, the viscosity of the melt, the mean volume fraction of particles, the thickness of the manufactured ring, and the solidification time.

On the other hand, the dispersion of intermetallic compounds in Al alloys is of significant commercial importance owing to the fact that it leads to the improvement of the mechanical properties of Al alloys [10]. The methods of fabrication by centrifugal casting for intermetallic compound-particle-dispersed FGMs can be classified into two categories on the basis of the relationship between the processing temperature and the liquidus temperature of a master alloy [9, 11]. If the liquidus temperature of the master alloy is significantly higher than the processing temperature, as shown in **Fig. 1(a)**, the intermetallic compound particles remain in the solid state in a liquid matrix during centrifugal casting, which is called the centrifugal solid-particle method. Alternatively, if the liquidus temperature of the master alloy is lower than the processing temperature, as shown in **Fig. 1(b)**, centrifugal force can be applied during the solidification both to the dispersed phase and the matrix, which is referred to as the centrifugal *in situ* method.

Since the mechanisms of formation of a compositional gradient during the centrifugal solid-particle method and the centrifugal *in situ* method are different, it is expected that the FGMs fabricated by the centrifugal *in situ* method processed at approximately the liquidus temperature should show extraordinary microstructures. In this study, Al-Al_3Ti FGMs are fabricated by the centrifugal *in situ* method processed at a temperature around the liquidus temperature from a master alloy of an Al-5 mass% Ti ingot containing 11 vol% Al_3Ti platelets in the Al matrix. A detailed study of the microstructures of the fabricated Al-Al_3Ti FGMs is carried out.

2. Microstructures of FGMs fabricated by centrifugal solid-particle

and centrifugal *in situ* methods

The mechanisms of formation of a compositional gradient during the centrifugal solid-particle and the centrifugal *in situ* methods differ. In the case of the centrifugal solid-particle method, a centrifugal force applied to a molten metal with dispersed solid particles drives the formation of the desired gradient. The compositional gradient is then achieved primarily by the difference in the centrifugal force produced owing to the difference in density between the molten metal and the solid particles [6,8]. It is known that the motion of particles in a viscous liquid under a centrifugal force obeys Stokes' law [8,12,13)]

$$\frac{dx}{dt} = \frac{\left|\rho_{\mathrm{p}} - \rho_{\mathrm{m}}\right| G g D_{\mathrm{p}}^{2}}{18\eta} , \quad (1)$$

where dx/dt, ρ, G, g, D, and η are the velocity, density, G number, gravitational acceleration, particle diameter, and viscosity of the molten metal, respectively. The subscripts 'p' and 'm' denote particle and matrix, respectively. Therefore, in the FGMs fabricated by the centrifugal solid-particle method, the mean particle size is distributed gradually along the applied centrifugal force [14]. The average particle size in the outer region is larger than that in the inner region, and the average particle size is gradually distributed in the FGMs, since the migration distance is greater for larger particles.

On the other hand, the smaller and larger particles are found in the outer and inner regions of the ring, respectively, in the FGMs fabricated by the centrifugal *in situ* method [15,16]. Therefore, the mechanisms of formation of the graded composition by the centrifugal solid-particle and centrifugal *in situ* methods differ. It is also found that as G-number increases, the particle size in the ring's outer region decreases. [15] It has been accepted in general that crystallized particle size varies depending on the solidification process. In the case of the centrifugal casting, it is reported that the cooling rate is higher in the outer region of the ring than in the inner region. [12,17] Moreover, the higher cooling rate for larger-G-number specimens is found. Therefore, the difference in the particle size distribution of the FGMs fabricated by the centrifugal *in situ* method should mainly originate from the gradation of a cooling rate.

To clarify the formation mechanism of a graded composition during the centrifugal *in* *situ* method, an Al-33 mass%Cu eutectic alloy without any primary crystals has been prepared by the centrifugal *in situ* method.[16] Since the graded composition forms in the Al-33 mass%Cu eutectic alloy sample, the origin of the graded structure could not be explained by the migration of primary crystals under the centrifugal force. The mechanism of the graded composition formation in the A-B alloy by the centrifugal *in situ* method can be summarized as follows.[16] First, owing to the density difference, the partial separation of A and B elements in the liquid state occurs. Then, a chemical compositional gradient is formed before the crystallization of the primary crystal. The primary crystal in the matrix appears to depend on the local chemical composition. The primary crystal migrates according to the density difference, and a further compositional gradient is formed

Then, one can expect to obtain unique graded distribution patterns by simultaneously using the centrifugal solid-particle method for an Al-Al_3Ti system and the centrifugal *in situ* method for an Al-Al_3Ni system. Thus, hybrid Al-(Al_3Ti+Al_3Ni) FGMs have been fabricated by simultaneously using the centrifugal solid-particle and centrifugal *in situ* methods with a mixture of both Al-Ti and Al-Ni master alloys.[18] It is found that the volume fraction of Al_3Ti particles increases toward the outer region of the ring, and a steeper graded distribution of Al_3Ti particles is formed for larger-*G*-number specimens. However, the volume fraction of Al_3Ni primary crystals gradually decreases towards the outer region. These results cannot be explained by the difference in density, because the densities of Al_3Ti and Al_3Ni intermetallic compounds are 3.4 and 4.0 Mg/m^3, respectively. In the early stage of the process, Al_3Ti particles preferentially migrate because no other solid particles are present at this stage. Then, the Al_3Ni particles crystallize directly from the molten matrix. However, since the viscosity of the melt increases owing to the increase in the number of particles in the suspension,[19] the viscosity of the melt is significantly higher in the outer region than in the other regions where the Al_3Ti particles condense. The increase in viscosity inhibits the migration of Al_3Ni particles.

Figure 2 shows a partial phase diagram of the Al-rich side of the Al-Ti binary phase diagram.[20,21] The dotted line in the figure shows the composition of an Al-5 mass% Ti alloy ingot. As can be seen, the liquidus temperature of the Al-5 mass% Ti alloy is about 1040 ^{o}C. When the temperature of the melting furnace was set below the liquidus temperature, Al_3Ti platelets remained in the solid state in a liquid Al matrix. In our previous studies, detailed experiments have been carried out to evaluate the microstructure and mechanical properties of the Al-Al_3Ti FGMs fabricated by the centrifugal solid-particle method. [22-28] On the other hand, when the temperature of the melting furnace was set at a temperature much higher than the liquidus temperature, the Al_3Ti platelets dissolved into molten Al and did not remain in the solid state. It has been found that the microstructure of Al-Al_3Ti FGMs fabricated by the centrifugal *in situ* method processed at 1600 °C was different from that fabricated by the centrifugal solid-particle method. [29)]

The above findings indicate that the mechanisms of formation of a compositional gradient during the centrifugal solid-particle and the centrifugal *in situ* methods differ. Therefore, it is expected that the FGMs fabricated by the centrifugal *in situ* method processed at a temperature around the liquidus temperature should show extraordinary microstructures. However, no study concerning the FGMs fabricated under such conditions has been carried out. In this study, Al-Al_3Ti FGMs are fabricated by the centrifugal *in situ* method processed at 1200 °C from a master alloy of an Al-5 mass% Ti ingot containing 11 vol% Al_3Ti platelets in the Al matrix.

3. Experimental methods

An Al-5 mass% Ti commercial alloy ingot with 11 vol% Al_3Ti platelets was used as the initial material. **Figure 3** shows the vacuum centrifugal casting system for fabricating the Al-Al_3Ti FGMs. The ingot was heated to 1200 ^{o}C, a temperature just above the liquidus line, ensuring the complete melting of all phases before processing, or 1000 ^{o}C, a temperature just below the liquidus line (centrifugal *in situ* method), at which the Al_3Ti platelets could remain in the solid state in a liquid Al matrix (centrifugal solid-particle method). The alloy was directly poured into a spinning mold through an inlet, for which the applied centrifugal forces were 300 and 600 *G*. Therefore, when the temperature of the melting furnace was set at 1200 ^{o}C, the centrifugal force could be applied during the solidification of both the Al_3Ti compound and the Al matrix. However, during melt pouring, the temperature of the molten metal drops to a certain degree, which results in the crystallization of the primary Al_3Ti crystal phase in the melt before the application of centrifugal force. The fabricated FGMs are ring-shaped with an outer diameter of 80 mm, a height of 50 mm, and a thickness of about 7 mm. The casting conditions and notation of samples are given in **Table I**. A detailed description of the centrifugal casting to fabricate FGMs is available in previous reports.[7,9,11,15-18,22,24)]

The microstructure of the FGMs was observed by scanning electron microscopy (SEM) on the plane perpendicular to the spinning direction of the ring-shaped specimens. The thickness direction (centrifugal force direction) was divided into ten regions of equal width, and the microstructures in each region were observed. The volume fraction and the orientation angles of the Al_3Ti platelets in each region were measured directly from the micrographs.

The texture of the Al matrix, as well as the Al_3Ti phase, in Specimens 2 and 4 was also analyzed by field emission (FE) -SEM with electron backscattered diffraction (EBSD) system. The dimensions of EBSD specimens were approximately 4 x 3 mm^2 in a cross section. EBSD measurement was carried out in a step size of 0.8 or 1.0 μm. The acceleration voltage of FE-SEM in the EBSD measurement was 15 kV. The data collected by EBSD were analyzed using TSL-OIM 6.1.

The hardness distribution in the fabricated samples was evaluated along the normalized thickness in 0.7 mm steps. A Vickers microhardness tester was used for the measurements at 490 mN for 15 s. The indentation was made at intermediate positions between particles. The hardness at each position was measured 5 times and the average was obtained.

4. Results

4.1 Microstructures of FGMs

The microstructures of Specimen 1 (temperature of crucible furnace, 1000 ^{o}C; temperature of mold preheating furnace, 300 ^{o}C; and G = 300), Specimen 2 (1200 ^{o}C, 478 ^{o}C, and G = 300), Specimen 3 (1200 ^{o}C, 300 ^{o}C, and G = 300), Specimen 4 (1200 ^{o}C, RT, and G = 300) and Specimen 5 (1200 ^{o}C, 300 ^{o}C, and G = 600) observed by SEM are shown in **Figs. 4(a) to 4(e)**, respectively. The upper, middle, and lower microstructures are observed at the normalized thickness of 1.0, 0.8, and 0.6 regions, respectively, where the normalized thickness shows the region along the centrifugal force direction normalized by the thickness of the ring; i.e., 0.0 is the inner surface and 1.0 is the outer surface. The centrifugal force in these figures is in the vertical direction. As can be seen from Fig. 4, there are three types of particles, namely, thin platelet-shaped particles with a thickness of less than 5 μm (thin platelets), thick platelet-shaped particles with a thickness of more than 10 μm (thick platelets), and small particles. Thick platelets are observed only in Specimen 1, especially at the normalized thickness of the 1.0 region (ring outer region). Although the results of energy dispersive X-ray spectroscopy (EDS) analysis are not presented here, all of the observable particles in Fig. 4 are identified to be the stoichiometric composition of the Al_3Ti phase. As can be seen, the microstructure changes from region to region. Moreover, the planes of both the thin and thick platelets tend to be oriented perpendicular to the centrifugal force direction. Note that the size of platelets in Specimens 2 to 5 decreases with increasing normalized thickness. These extraordinary microstructures can be formed by not the centrifugal solid-particle method but the centrifugal *in situ* method, as described in Sect. 2.

The volume fraction of Al_3Ti platelets was determined from the micrographs. The concentration profiles of Al_3Ti platelets along the centrifugal force direction of Specimens 1 to 5 are shown in **Figs. 5(a) to 5(e)**, respectively, as a function of normalized thickness. The volume fraction of Al_3Ti platelets increases from the inner to the outer surface of the ring. This is because the density of Al_3Ti is higher than that of molten Al. Figures 5(b) -5(d) show the effect of the temperature of the mold preheating furnace on the distribution of Al_3Ti particles. By comparing between Specimens 3 and 5, it is seen that a steeper compositional gradient formed in larger-G-number specimens. Note that negative gradients are found in the outer regions of the ring for Specimens 3 and 4. These findings indicate that mold temperature plays an important role in the solidification behavior in centrifugal casting.[30] Moreover, a steeper compositional gradient is found for Specimen 1 fabricated by the centrifugal solid-particle method processed at 1000 °C compared with Specimen 3 fabricated by the centrifugal *in situ* method processed at 1200 °C. Since a higher casting temperature causes a lower viscosity of the melt and a longer sedimentation period, a steeper gradient must appear for the specimen processed by the centrifugal solid-particle method at a higher temperature. These findings indicated that the microstructures of FGMs are different from those fabricated by the centrifugal solid-particle method.

4.2 Orientation of Al_3Ti platelets

As mentioned above, it was found that most of the platelets are oriented with their planes nearly perpendicular to the radial direction. Let us define the orientation angle θ using the microstructure of Specimen 1, as shown in **Fig. 6(a)**, to describe the degree of orientation. Orientation histograms observed at the normalized thickness of 1.0 and 0.8 regions in Specimen 1 are shown in **Fig. 6(b)**. The peak distributions indicate a tendency for platelets to align their planes perpendicular to the centrifugal force direction for both thin and thick platelets. It is recognized from this figure that a stronger orientation effect is found for thick platelets. As will be discussed latter, thick platelets remain as solid particles in the melt during the process, whereas thin platelets formed by crystallization as primary crystals. These differences cause the difference in orientation.

To express the orientation effect quantitatively, the following Herman's orientation parameter *fp* was calculated. [24,25,28,31-34)]

$$fp = (2\langle\cos^2\phi\rangle - 1), \tag{2}$$

where the trigonometric average is

$$\langle\cos^2\phi\rangle = \int_{-\pi/2}^{\pi/2} \cos^2\phi\, n(\phi)\, d\phi. \tag{3}$$

The term $n(\phi)$ is the orientation distribution function, which specifies the fraction of platelets within the angular element $d\phi$. The parameter *fp* values range between 0 (for a random distribution of planes) and 1 (for a perfect alignment). The intermediate values of this parameter correspond to partial states of orientation. The evaluated Herman's orientation parameter *fp* for Specimen 1 is also shown in **Fig. 6(b)**. Since thick platelets show larger values of Herman's orientation parameter, they show a stronger orientation than thin platelets.

The orientation histograms of thin platelets in Specimens 2 to 5 observed at the normalized thickness of 1.0, 0.8, 0.6 and 0.4 regions are shown in **Figs. 7(a) to 7(d)**, respectively. The evaluated Herman's orientation parameter *fp* is also shown in this figure. By comparing the data of Specimens 3 and 5, a larger orientation effect is found for the larger-*G*-number specimen, which is in agreement with previous studies on the centrifugal solid-particle method. Moreover, it is found that the orientation parameter also changes from place to place, and smaller orientation effects are found at larger-normalized-thickness regions, i.e., the outer region of the ring. In our previous studies,[24,25] it was found that the orientation parameter in the Al-Al_3Ti platelet FGMs fabricated by the centrifugal solid-particle method increases with normalized thickness. These findings indicate unexpected orientation effects in the FGMs fabricated by the centrifugal *in situ* method. By comparing among Specimens 2 to 4, smaller orientation effects are found for the FGMs fabricated at a lower temperature of the mold preheating furnace. Again, mold temperature plays an important role in the solidification behavior in centrifugal casting.

4.3 Crystal orientation analysis of Al matrix

In this study, Al matrix FGMs with oriented Al_3Ti platelets can be fabricated by the centrifugal *in situ* method, as well as by the centrifugal solid-particle method. It was reported that the plane normal direction of the Al_3Ti platelets corresponds to the $[001]_{Al3Ti}$ direction.[23,27,28] Moreover, there are some orientation relationships (ORs) between the Al and Al_3Ti phases. Therefore, the Al matrix in the FGMs also forms a crystallographic texture. The crystal orientation analysis of the Al matrix, as well as Al_3Ti platelets, is carried out by EBSD.

Figures 8(a) - 8(c) show the inverse pole figure (IPF) maps and pole figures of the Al_3Ti and Al phases in Specimen 2 observed at the normalized thickness of 0.9, 0.7, and 0.4 regions, respectively. Similarly to Fig. 4, these IPF maps show a crystal plane orientation on the plane perpendicular to the spinning direction of the ring-shaped specimens; however, the IPF maps in these figures present a crystal plane orientation on the plane perpendicular to the centrifugal force direction. Figures 8(a) - 8(c) show that the orientation of the Al_3Ti platelet face is $(001)_{Al_3Ti}$, which is in agreement with previous studies.[23,27,28] Thus, the $[001]_{Al_3Ti}$ direction of the Al_3Ti phase is aligned perpendicular to the Al_3Ti platelets. Moreover, the crystallographic texture of the Al matrix is also recognized, especially in the outer region.

IPF maps and pole figures of the Al_3Ti and Al phases in Specimen 4 observed at normalized thickness of 0.9, 0.7, and 0.4 regions are shown in **Figs. 9(a) - 9(c)**, respectively. The IPF maps in these figures also show the crystal orientation on the plane perpendicular to the centrifugal force direction. It can be seen from Fig. 9 that not only the Al_3Ti phase but also the Al matrix has texture at the normalized thickness of 0.9 and 0.7. In the comparison of the texture of the Al phase with that of the Al_3Ti phase, however, one can notice that the former is somewhat limited. Why does the Al matrix have a weaker texture than the Al_3Ti phase? This will be discussed later.

4.4 Hardness distribution in the FGMs

Figure 10 shows the micro-Vickers hardness distributions of Specimens 1 to 5 as a function of specimen region. This figure indicates that the hardness along the centrifugal force direction gradually increases from the inner surface to the outer surface of the ring. As the temperature of the mold increases, the hardness in the outer region increases, which can be confirmed by comparing the data of Specimens 2 to 4. Moreover, Specimen 5 fabricated at G=600 has a steeper gradient in hardness than Specimen 3 fabricated at G=300. Therefore, as G increases, the steeper the gradient in hardness becomes. From these results, it is found that the hardness distribution in FGMs is closely related to the Al_3Ti particle distribution.

5. Discussion

5.1 Extraordinary microstructures in fabricated FGMs

In this study, smaller platelets are found in the ring outer region in Specimens 2 to 5. If the graded microstructure is formed by Stokes' migration, the obtained results are conflicting. Since the crystallized particle size varies depending on the solidification process and the cooling rate is higher in the outer region of the ring than in the inner region, [12,17] the particle size of the FGMs fabricated by the centrifugal *in situ* method is mainly affected by the cooling rate.

Therefore, not only the temperature of the crucible furnace but also the temperature of the mold preheating furnace should affect the microstructural evolution in the FGMs fabricated by the centrifugal *in situ* method. As the specimen cools, the Al_3Ti phase produced from the melt will float to the outer surface of the ring under centrifugal force. This is due to the density difference between the Al_3Ti phase and the remaining melt. At the same time, platelets become oriented by centrifugal force. However, a lower mold temperature inhibits such microstructural evolution. As a result, negative gradients in volume fraction and orientation parameter, as shown in Figs. 5 and 7, could appear.

Three types of particles, namely, thin platelets, thick platelets, and small particles, are observed in Specimen 1. The thick platelets are mainly located in the outer region of the ring. Moreover, it is shown in Fig. 6(b) that a stronger orientation effect is found for thick platelets. To discuss these phenomena, gravity casting at 1000 °C was carried out and the result is shown in **Fig. 11**. It is seen that both thin platelets and thick platelets can also be observed. Therefore, the difference in the morphology of platelets observed in Specimen 1 is not due to the applied centrifugal force. It is reported that the morphologies of Al_3Ti particles is related to the temperature of molten Al. **Figure 12** shows the relationship of the morphology of Al_3Ti with the melting temperature and cooling rate. [35,36] Blocky Al_3Ti could be obtained at low temperatures (< 850 °C), and needlelike plate/strip Al_3Ti could be obtained at high melting temperatures (higher than 1000 °C). Judging from this figure, thick platelets observed in Specimen 1 are considered to be formed by crystallization from molten metal during the centrifugal casting, while the thick platelets may be due to Al_3Ti retained in the melt. This is the reason why the stronger orientation effect and compositional gradation are found for thick platelets. Moreover, the small particles observed in Specimens 1 to 5 may be formed at lower temperatures under centrifugal force, since the liquidus line in the Al-Ti system is not vertical, as shown in Fig. 3, and newly formed particles can be crystallized at lower temperatures.

5.2 Origin of texture and prediction of preferred crystal orientation relationship between Al and Al_3Ti

In this study, it is found that the orientation of Al_3Ti platelets and the texture of the Al matrix are formed in the FGMs fabricated by the centrifugal *in situ* method. Although strong orientation effects of Al_3Ti platelets were observed for the FGMs, the texture of the Al matrix of the FGMs is somewhat limited. Since an Al_3Ti particle in an Al melt functions as the heterogeneous nucleation site of pure Al grains, [37-39] the solidification of the Al matrix around Al_3Ti particles would proceed under epitaxial phenomena. However, there are the multiple orientation relationships between the Al and Al_3Ti phases.[40-42] Therefore, prediction of the prefered orientation relationship between Al and Al_3Ti is important.

In previous studies [42-44], the disregistry model of two-dimensional lattices proposed by Bramfitt [45] was adopted to discuss the favorable heterogeneous nucleation sites. The plane disregistry δ can be evaluated as

$$\delta_{(hkl)_n}^{(hkl)_s} = \frac{1}{3}\sum_{i=1}^{3}\left\{ \frac{\left| d[uvw]_s^i \cos\theta - d[uvw]_n^i \right|}{d[uvw]_n^i} \right\} \times 100\% \quad , \tag{4}$$

where $(hkl)_s$ and $(hkl)_n$ are the low-index planes of the substrate and nucleated solid, $[uvw]_s$ and $[uvw]_n$ are the low-index orientations on $(hkl)_s$ and $(hkl)_n$, respectively, $d[uvw]_s$ and $d[uvw]_n$ are the interatomic spacing distances along $[uvw]$, and θ is the angle between $[uvw]_s$ and $[uvw]_n$. However, the physical meaning of the plane disregistry is unclear; for example, it is unclear why three in-plane directions are considered when evaluating the plane disregistry. Therefore, we alternatively adopt the parameter M, which is approximately proportional to the specific misfit strain energy, to discuss the effective nucleant materials. Kato *et al*. have investigated the preferred epitaxial relationship (crystallographic orientation relationship) between the deposit and the substrate of metals using M.[46-48] According to his study, the epitaxial relationship with small M values becomes the preferred one. The parameter M is defined as

$$M = \varepsilon_1^2 + \varepsilon_2^2 + (2/3)\ \varepsilon_1\ \varepsilon_2, \tag{5}$$

where ε_1 and ε_2 are the principal misfit strains calculated from the principal distortions. Although this consideration has often been applied to epitaxial phenomena, it is potentially applicable to predicting the preferred orientation relationship between two solidified phases. On the basis of the unit cell structures of Al and Al_3Ti shown in **Fig. 13**, and using the lattice parameters a = 0.4049 nm of Al [49], and a = 0.3851 nm and c = 0.8608 nm of Al_3Ti [50], and also applying **Eqs. (4) and (5)**, the plane disregistry and M values for six possible ORs shown in **Fig. 14** were calculated. The results are listed in **Table II**. Since OR-E has the smallest *M* among all the possible ORs, it is predicted that OR-E is the most preferred relationship between Al and Al_3Ti. In reality, as reported by Marcantonio and Mondolfo [40], OR-E was the preferred relationship between Al and Al_3Ti. The second candidates of ORs predicted using *M* are OR-D and OR-F. These two ORs have also been observed by Marcantonio and Mondolfo [40] and Kobayashi *et al.* [52] In contrast, although OR-B with a much larger *M* was experimentally found, the amount of the interface was much smaller with OR-B than with OR-E, OR-D, and OR-F. [40),51),52] Therefore, we conclude that the parameter *M* is readily applicable to predicting preferred orientation relationships between solidified Al and Al_3Ti. That is, favorable heterogeneous nucleation sites for casting could be predicted using the parameter *M*.

To describe these orientation relationships on a stereographic projection of Al_3Ti, the composite inverse figure of 001_{Al3Ti} poles in the Al coordinate is shown in **Fig. 15**. The 001_{Al3Ti} poles with OR-A to OR-F are superimposed. As shown in the figure, there is no one-to-one correspondence between the $[001]_{Al_3Ti}$ and $\langle 001 \rangle_{Al}$ directions because of multiple orientation relationships between the Al_3Ti and Al phases. Therefore, a strong texture does not appear for the Al matrix, even in the FGMs with the perfect alignment of Al_3Ti platelets, *i.e.*, Herman's orientation parameter *fp* is 1.

6. Conclusions

In this study, the microstructures of Al-Al_3Ti FGMs fabricated by the centrifugal solid-particle method at 1000 °C and the centrifugal *in situ* method at 1200 °C from a master alloy of an Al-5 mass% Ti ingot containing 11 vol% Al_3Ti platelets in the Al matrix are investigated. The main results are as follows.

1) Compositional gradient and orientation effects are found in the FGMs fabricated by the centrifugal *in situ* method, as well as centrifugal solid-particle method. However, the microstructures of FGMs fabricated by the centrifugal *in situ* method are found to be different from those fabricated by the centrifugal solid-particle method.
2) Although it is found that not only the Al_3Ti phase but also the Al matrix has texture in the outer regions of the ring, the texture of the Al phase is somewhat limited compared with that of the Al_3Ti phase.

3) The parameter *M*, which is approximately proportional to specific misfit strain energy, is readily applicable to predicting preferred orientation relationships between solidified Al and Al_3Ti.
4) Because of multiple orientation relationships between the Al_3Ti and Al phases, a strong texture does not appear for the Al matrix.

Acknowledgments

We gratefully acknowledge The Light Metal Educational Foundation Inc. of Japan and Grants-in-Aid for Scientific Research (S: 26220907, B: 15H04143) from the Japan Society for the Promotion of Science for supporting this resurch.

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Figure Captions

Fig. 1. (Color online) Two types of phase diagrams applicable to (a) centrifugal solid-particle method and (b) centrifugal *in situ* method. Schematic illustrations of the development of microstructures during centrifugal solid-particle method and centrifugal *in situ* method are also shown in (a) and (b), respectively.

Fig. 2. (Color online) Al end of the Al–Ti equilibrium phase diagram. [20,21]

Fig. 3. (Color online) Schematic illustration of the vacuum centrifugal caster used in this study.

Fig. 4. (Color online) Typical microstructures observed at normalized thickness of 1.0, 0.8, and 0.6 regions. (a) Specimen 1 (1000 ^{o}C, 300 ^{o}C, and G = 300), (b) Specimen 2 (1200 ^{o}C, 478 ^{o}C, and G = 300), (c) Specimen 3 (1200 ^{o}C, 300 ^{o}C, and G = 300), (d) Specimen 4 (1200 ^{o}C, RT, and G = 300), and (e) Specimen 5 (1200 ^{o}C, 300 ^{o}C, and G = 600). The vertical direction in these images is the centrifugal force direction.

Fig. 5. (Color online) Histograms of volume fractions of the Al_3Ti platelets in Specimens (a) 1, (b) 2, (c) 3, (d) 4, and (e) 5. The abscissa represents the region in the thickness direction of the ring, normalized by thickness, *i.e.*, 0.0 and 1.0 correspond to the inner and outer peripheries, respectively.

Fig. 6. (Color online) (a) Microstructures of Specimen 1 and the definitions of thick plates, thin plates, and the orientation angle θ. (b) Orientation histograms observed at normalized thickness of 1.0 and 0.8 regions in Specimen 1. The evaluated Herman's orientation parameter fp is also shown in this figure.

Fig. 7. (Color online) Orientation histograms observed at normalized thickness of 1.0, 0.8, 0.6, and 0.4 regions in Specimens (a) 2, (b) 3, (c) 4, and (d) 5. The evaluated

Herman's orientation parameter *fp* is also shown in this figure.

Fig. 8. (Color) Inverse pole figure maps and pole figures of Al_3Ti and Al phases in Specimen 2 (1200 °C, 478 °C, and 300*G*) observed at normalized thickness of (a) 0.9, (b) 0.7, and (c) 0.4 regions. CD and AD are the centrifugal force and axis directions, respectively. Note that the IPF maps show a crystal plane orientation on the plane perpendicular to the centrifugal force direction.

Fig. 9. (Color) Inverse pole figure maps and pole figures of Al_3Ti and Al phases in Specimen 4 (1200 °C, RT, and 300*G*) observed at normalized thickness of (a) 0.9, (b) 0.7, and (c) 0.4 regions. CD and AD are the centrifugal force and axis directions, respectively. Note that the IPF maps show a crystal plane orientation on the plane perpendicular to the centrifugal force direction.

Fig. 10. (Color online) Micro-Vickers hardness distributions in Specimens 1 to 5 as a function of normalized thickness.

Fig. 11. (Black and White) SEM image showing microstructure of Specimen 6 fabricated by gravity casting (1000 °C, 300 °C, and $G = 1$).

Fig. 12. (Color online) Relationship of the morphology of Al_3Ti with melting temperature and cooling rate.[35,36] (A) needlelike Al_3Ti area, (B) mixture of blocky and needlelike Al_3Ti area, (C) blocky Al_3Ti area, (D) metastable area, and (E) Ti supersaturated solid solution area.

Fig. 13. (Color online) Schematic illustrations and lattice constants of (a) Al with fcc structure[49] and (b) Al_3Ti with $D0_{22}$ structure.[50]

Fig. 14. (Color online) Atomic arrangements of Al_3Ti intermetallic compound of OR-A to OR-F superimposed on atomic arrangements of Al phase indicated by broken lines.

Fig. 15. (Black and White) Composite inverse figure of 001_{Al3Ti} poles in the Al

coordinate. The 001_{Al3Ti} poles with OR-A to OR-F are superimposed.

Table I Casting conditions for Specimens 1 to 6.

Specimen	Temperature of crucible furnace	Temperature of mold preheating furnace	G number
1	1000 °C (solid-particle method)	300 °C	300 G
2	1200 °C (*In situ* method)	478 °C	
3		300 °C	
4		RT	
5		300 °C	600 G
6	1000 °C (Gravity casting)		1 G

Table II Disregistry and *M* values of OR-A to OR-F.

Interface		Principal direction of Al	Principal direction of Al_3Ti	$\lvert \boldsymbol{x}_{Al} \rvert, \lvert \boldsymbol{y}_{Al} \rvert$	$\lvert \boldsymbol{x}_{Al3Ti} \rvert, \lvert \boldsymbol{y}_{Al3Ti} \rvert$	Principal strain ε_x, ε_y	Disregistry δ	M [x10^{-3}]	Ref.
A	$(100)_{Al}$ //$(100)_{Al3Ti}$	[010]	[010]	0.4049 nm	0.3851 nm	–0.0489	3.96 %	4.306	-
		[001]	[001]	0.8098 nm	0.8608 nm	0.0630			
B	$(001)_{Al}$ // $(001)_{Al3Ti}$	[100]	[100]	0.4049 nm	0.3851 nm	–0.0489	4.89 %	6.377	40 51 52
		[010]	[010]	0.4049 nm	0.3851 nm	–0.0489			
C	$(110)_{Al}$ // $(110)_{Al3Ti}$	$[\bar{1}10]$	$[\bar{1}10]$	0.2863 nm	0.2723 nm	–0.0489	4.12 %	4.306	-
		[001]	[001]	0.8098 nm	0.8608 nm	0.0630			
D	$(111)_{Al}$ // $(112)_{Al3Ti}$	$[\bar{1}10]$	$[\bar{1}10]$	0.2863 nm	0.2723 nm	–0.0489	3.08 %	2.240	52
		$[\bar{1}\bar{1}2]$	$[\bar{1}\bar{1}1]$	0.4959 nm	0.5093 nm	0.0270			
E	$(221)_{Al}$ // $(001)_{Al3Ti}$	$[1\bar{1}0]$	[210]	0.8589 nm	0.8611 nm	0.0025	0.26 %	0.017	40
		$[11\bar{4}]$	$[\bar{1}20]$	0.8589 nm	0.8611 nm	0.0025			
F	$(110)_{Al}$ // $(001)_{Al3Ti}$	$[\bar{1}10]$	[110]	0.2863 nm	0.2723 nm	–0.0489	0.24 %	2.182	40
		[001]	$[\bar{1}10]$	0.8098 nm	0.8169 nm	0.0088			