



HOKKAIDO UNIVERSITY

Title	Centrifugal Casting of Al-Cr Semi-Solid Slurry Produced by Stir-Mix-Quenching
Author(s)	Ohmi, Tatsuya; Murota, Yasuhiro; Kudoh, Masayuki
Citation	Materials Transactions, JIM, 40(10), 1063-1068 https://doi.org/10.2320/matertrans1989.40.1063
Issue Date	1999-10
Doc URL	https://hdl.handle.net/2115/74707
Type	journal article
File Information	2-6.pdf



Centrifugal Casting of Al-Cr Semi-Solid Slurry Produced by Stir-Mix-Quenching

Tatsuya Ohmi, Yasuhiro Murota[†] and Masayuki Kudoh

*Division of Materials Science and Engineering, Graduate School of Engineering,
Hokkaido University, Sapporo 060-8628, Japan*

We propose a novel process for producing tubular *in-situ* composite castings, in which fine intermetallic crystals are concentrated near the outside surface. In this process, two kinds of molten metals with different compositions are mixed with agitation in a mixing vessel to produce semi-solid slurry containing fine intermetallic crystals, and then the slurry is centrifugally cast. The authors call the first stage of the process "stir-mix-quenching." In the stir-mix-quenching experiments, molten aluminum was used for "the first melt" which acts as a coolant, and a hyperperitectic Al-Cr alloy melt with a higher liquidus temperature was used for "the second melt," which is rapidly quenched to precipitate numerous fine primary crystals when it is brought into contact with the first melt. The structure of the composite layer in the cast pipe produced by the centrifugal casting of the mixed-alloy slurry varies with the casting temperature of the slurry. When the casting temperature is higher than the final peritectic temperature, the intermetallic crystals are distributed uniformly in the composite layer. However, when the casting temperature is excessively high (for instance, 50 K higher than the final peritectic temperature), relatively coarse intermetallic crystals form near the boundary between the composite layer and the intermetallic-free region. On the other hand, the growth of the intermetallic crystals is suppressed when the casting temperature is in the neighborhood of the final peritectic temperature.

(Received March 18, 1999; In Final Form June 15, 1999)

Keywords: semi-solid processing, centrifugal casting, mixing, rapid solidification, aluminum alloy, composite material, intermetallic compound, structure control, refinement, solidification

I. Introduction

The dispersion of fine hard particles is effective in improving the hardness and wear resistance of aluminum alloys. Recently, we proposed "centrifugal duplex casting" (CDC)⁽¹⁾ to produce a tubular *in-situ* composite casting in which fine particles of an intermetallic compound are concentrated near the outside surface. The CDC process is a hybrid of Duplex Casting⁽²⁾ and the centrifugal casting of aluminum-transition metal alloys. The former is effective for the refinement of primary crystals in cast alloys,⁽²⁾ and the latter produces *in-situ* functionally graded materials in which primary intermetallic crystals are distributed with gradient volume fraction.⁽³⁾⁻⁽⁵⁾ We demonstrated the feasibility of the CDC process using an Al-Cr alloy.⁽¹⁾ In the CDC process, two kinds of molten metals with different compositions are cast in sequence at a given interval into the rotating mold of a centrifugal caster. Molten aluminum was used for "the first melt", which was cast first, and a hyperperitectic Al-Cr alloy melt with a higher liquidus temperature was used for "the second melt," which was cast later.

The basic concept of the CDC process is as follows. When the second melt is cast, it collides with the meniscus of the first melt, which is already cooling in the mold. Then, the second melt is dispersed as a great number of small fluid clumps. These fluid clumps are

rapidly quenched; they then migrate toward the outer periphery due to centrifugal force and accumulate to form a composite layer with fine primary crystals.

The solidification structure of the composite layer is controlled by the cooling capacity of the first melt⁽¹⁾. For instance, when the first melt is superheated at the moment of the second-melt casting, coarse intermetallic crystals grow on the inner side of the refined composite layer. On the other hand, when the first melt has partially solidified, the growth of the intermetallic crystals is suppressed. That is to say, we can increase the cooling capacity of the first melt through the effect of latent heat absorption.

The CDC process, however, is not suitable for the fabrication of castings of complex design because mixing in an intricate mold is difficult. In the present study, therefore, we examined a new process that is suitable for such cases. In this process, the first melt and the second melt are mixed with agitation in a mixing vessel to produce semi-solid slurry containing fine intermetallic crystals, and then the slurry is centrifugally cast. We call the first step of the process, in which the semi-solid slurry is produced, "stir-mix-quenching." The state of the first melt at the moment of mixing is liquid or semi-solid slurry in stir-mix-quenching. The concept of this process is an extended version of the slurry-melt mixing process⁽⁶⁾⁻⁽⁸⁾ in which the first melt in a semi-solid state is mixed with the second melt.

[†] Graduate Student, Hokkaido University.

II. Experimental Procedure

1. Stir-mix-quenching

Figure 1(a) shows a schematic view of the apparatus for stir-mix-quenching. The apparatus consists of stirring equipment and a mixing vessel made of insulation brick (60 mm inside diameter, 85 mm depth, and 10 mm radial thickness). The stirrer has two paddles, and its rotation speed is controlled electronically. In order to monitor the temperature of the alloy in the mixing vessel, a thermocouple is located near the inner wall of the vessel.

Figure 2 shows an aluminum-side portion of an Al-Cr phase diagram⁽⁹⁾. We used pure aluminum as the first melt and Al-10 mass%Cr alloy as the second melt. The mass ratio of the first melt to the second melt, W_1/W_2 , was 3, and the total mass of the mixed-alloy slurry was 336 g. The temperature of the first melt just before the pouring into the vessel was 100 K above the melting point of aluminum, and that of the second melt was 50 K above the liquidus temperature of Al-10 mass%Cr alloy.

The experimental procedure for stir-mix-quenching was as follows. The first melt was poured into the mixing vessel, and the stirrer was rotated at 2 s^{-1} . When the

temperature of the first melt fell down to 933 K, that is, just before the first melt began to solidify, the rotational speed of the stirrer was increased to 6 s^{-1} , and immediately after that, the second melt was mixed with the first melt. The mixed alloy was air-cooled with agitation and then centrifugally cast at a given temperature.

In order to examine the structure of the mixed-alloy slurry, an independent experiment of stir-mix-quenching was carried out. In this experiment, the stirring was terminated when the temperature of the mixed alloy fell down to the final peritectic temperature (934 K), and immediately after that, a small amount of the mixed alloy was water-quenched. The remainder of the mixed alloy was air-cooled and solidified in the mixing vessel.

The conventional stir-casting⁽¹⁰⁾ of Al-2.5 mass%Cr alloy was also carried out, and the stir-cast slurry was water-quenched or air-cooled to prepare comparison specimens.

2. Centrifugal casting of the mixed-alloy slurry

Figure 1(b) shows the configuration of the mold of the vertical centrifugal caster used in the experiments. A cylindrical container made of insulation brick (80 mm inside diameter, 40 mm depth, and 10 mm radial thickness) was used for the mold. The mold was not preheated.

The mixed-alloy slurry was cast at a given temperature into the rotating mold and solidified to form a tubular specimen. The rotation speed of the mold was 19 s^{-1} , which produced a gravity multiple of 59G as the centrifugal force at the outer periphery of the specimen. The radial thickness of the specimen was about 16 mm. An etchant, mixed in the ratio of $\text{HCl}:\text{HNO}_3:\text{C}_2\text{H}_5(\text{OH})_3 = 3:1:2$, was used to reveal the macrostructure of the specimen. The particle size of intermetallic crystals and Vickers hardness were measured along the radius direction on the transverse cross section at the 1/2 height of the specimen.

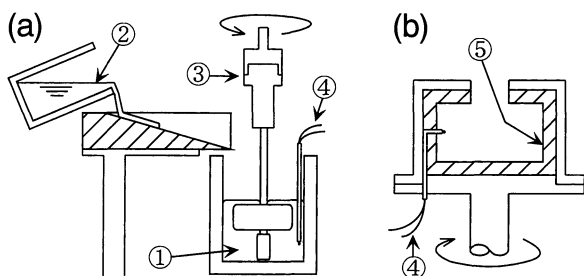


Fig. 1 Schematic illustration of the experimental apparatus. (a) Apparatus for stir-mix-quenching; (b) mold part of the vertical centrifugal caster. ① first melt, ② second melt, ③ stirrer, ④ thermocouple, ⑤ mold.

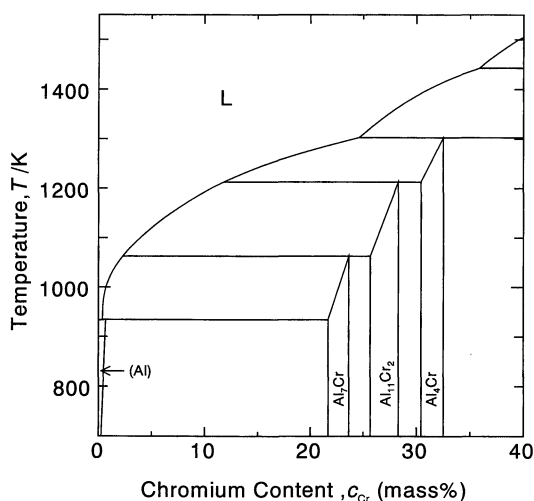


Fig. 2 Aluminum-side portion of the Al-Cr phase diagram⁽⁹⁾.

III. Results and Discussion

1. Solidification structure of the mixed-alloy slurry

Figures 3(a) and (b) show the microstructures of the conventionally stir-cast slurry. They show the back-scattered electron images obtained by using a scanning electron microscope. The bright particles are Al_7Cr intermetallic crystals, and the dark matrix is aluminum solid solution (α -phase). In both of the water-quenched slurry (a) and the air-cooled slurry (b), there are coarse particles which are of the order of $10^2 \mu\text{m}$.

Figure 4 shows a typical time-temperature curve measured during the stir-mix-quenching experiment. The temperature of the melt in the mixing vessel rapidly increased after the melt mixing, and the maximum temperature was 1013 K, which is 62 K lower than the liquidus temperature for the mixed-alloy composition Al-2.5 mass%Cr. This result indicates that the thermal condition adopted in the stir-mix-quenching experiment

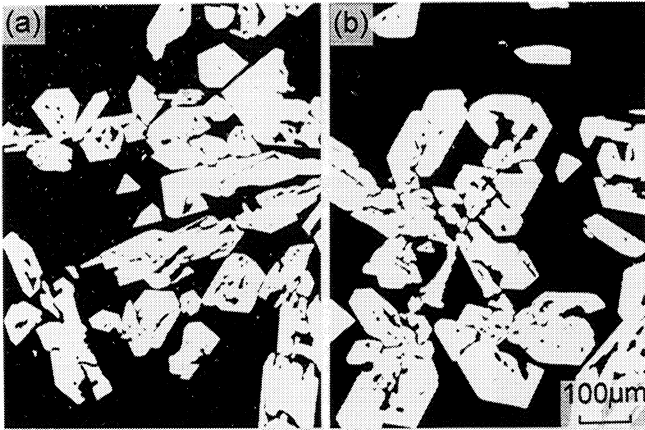


Fig. 3 Microstructures of the Al-2.5 mass%Cr alloy slurry produced by the conventional stir-casting. (a) Water-quenched slurry; (b) air-cooled slurry.

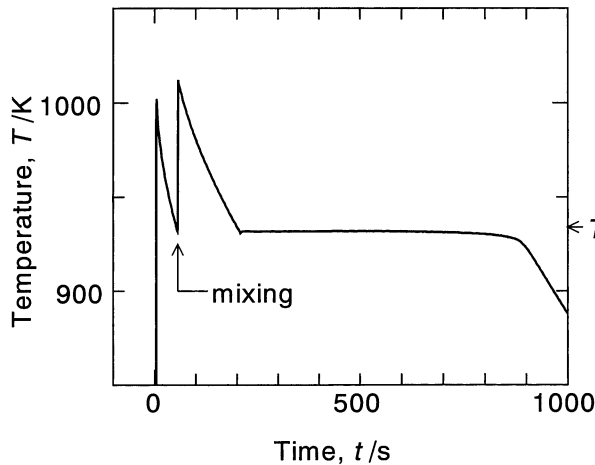


Fig. 4 Typical time-temperature curve measured during stir-mix-quenching. T_P : final peritectic temperature.

led to the formation of stable semi-solid slurry.

Figure 5 shows the microstructures of the mixed-alloy slurry. In both of the water-quenched slurry (a) and the air-cooled slurry (b), fine intermetallic crystals are dispersed in an α -phase matrix. The particle size, which is the arithmetic mean of the area equivalent diameter, is $9.6\text{ }\mu\text{m}$ in the water-quenched sample and $10.0\text{ }\mu\text{m}$ in the air-cooled one. These results show that the particle size is not affected by the cooling condition after the completion of intermetallic crystallization. It is also noteworthy that these particle sizes are comparable to those of the intermetallic crystals in the fine composite layer in the cast pipes produced by the CDC process⁽¹⁾.

2. Solidification structure of the centrifugally cast slurry

Figure 6 shows the macrostructures of a vertical cross section of the cast pipes produced by the centrifugal casting of the mixed-alloy slurry. The casting temperatures were (a) 984 K, (b) 944 K, and (c) 933 K, respectively. In the case of (c), the casting temperature was 1 K

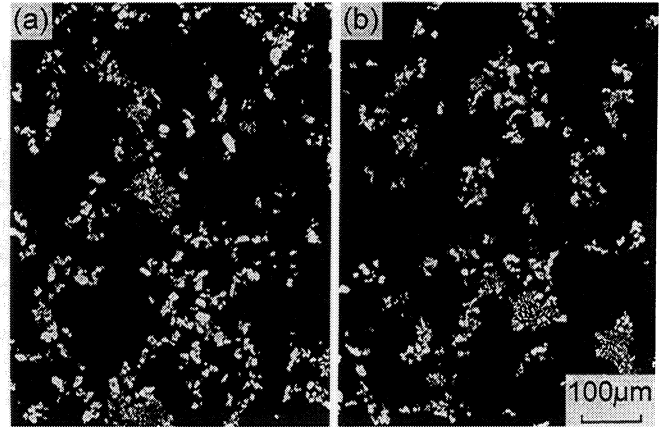


Fig. 5 Microstructures of the mixed-alloy slurry produced by stir-mix-quenching. (a) Water-quenched slurry; (b) air-cooled slurry.

lower than the final peritectic temperature, and the slurry was cast 15 s after the beginning of α -phase solidification. In each specimen, intermetallic crystals are concentrated on the outer side to form a composite layer. However, the structures of the composite layer vary with the casting temperature of the slurry as shown in **Fig. 7**. For the specimens shown in **Figs. 7(a) and (b)**, whose casting temperatures were higher than the final peritectic temperature, the intermetallic crystals are distributed uniformly in the composite layer. On the other hand, in **Fig. 7(c)**, α -phase grains are observed in the composite layer. This fact indicates that the α -phase grains contained in the slurry were accumulated accompanying the intermetallic crystals.

Figures 8(a), (b) and (c) show the microstructures near the boundary between the composite layer and the intermetallic-free region in the specimens shown in **Figs. 6(a), (b) and (c)**, respectively. Particularly noteworthy is that the size of the intermetallic crystals in the immediate vicinity of the boundary varies with the casting temperature. When the casting temperature was in the neighborhood of the final peritectic temperature, as in the case of **Fig. 8(b) or (c)**, the size of the particles in the close vicinity of the boundary is comparable to that of particles in the interior of the composite layer. On the other hand, relatively coarse particles are observed in the boundary region in **Fig. 8(a)**. As mentioned above, similar coarse particles form in the CDC pipes when the cooling capacity of the first melt is insufficient to completely quench the second melt. However, the coarse particles of the specimen shown in **Fig. 8(a)** are smaller than the particles in the CDC pipe⁽¹⁾ produced under similar thermal conditions, that is, using the same mold material and the same combination of the first melt just above the liquidus temperature and the second melt with a superheat of 50 K. Therefore, it is easier to suppress the formation of the coarse particles in the present process than in the CDC process.

3. Mechanism of coarse-particle formation

We propose the following mechanism for the forma-

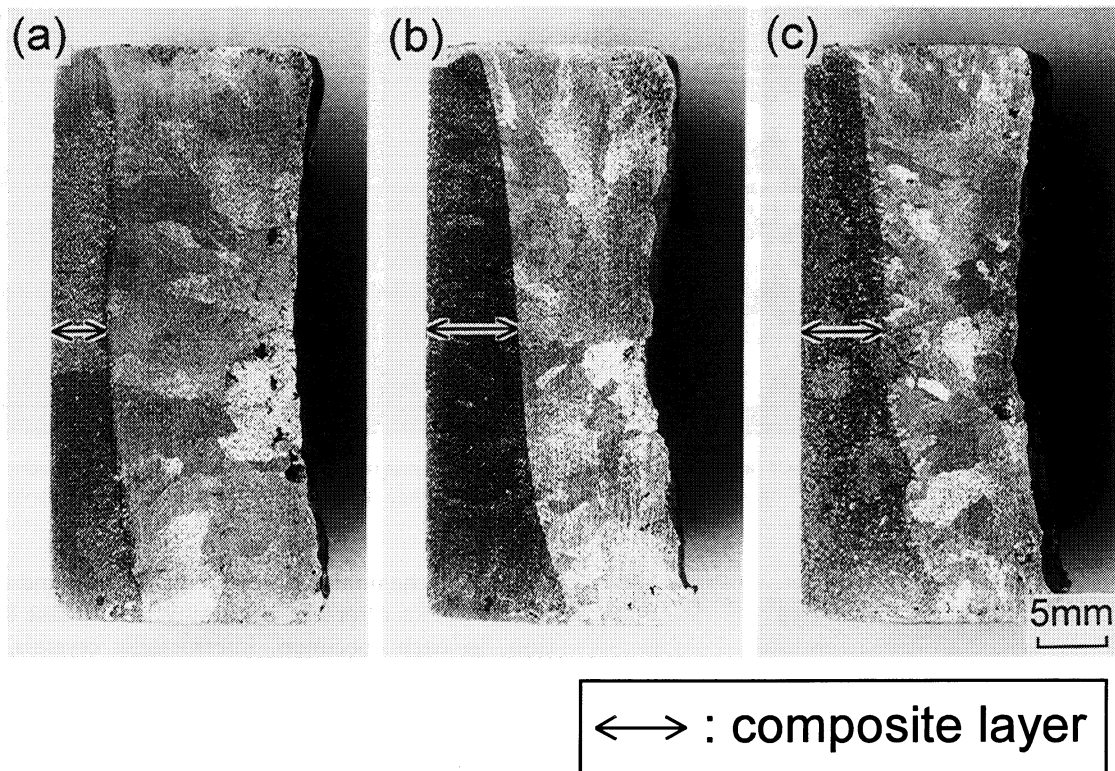


Fig. 6 Macrostructures of vertical cross section of the cast pipes produced by the centrifugal casting of the mixed-alloy slurry. Casting temperature: (a) 984 K, (b) 944 K, (c) 933 K.

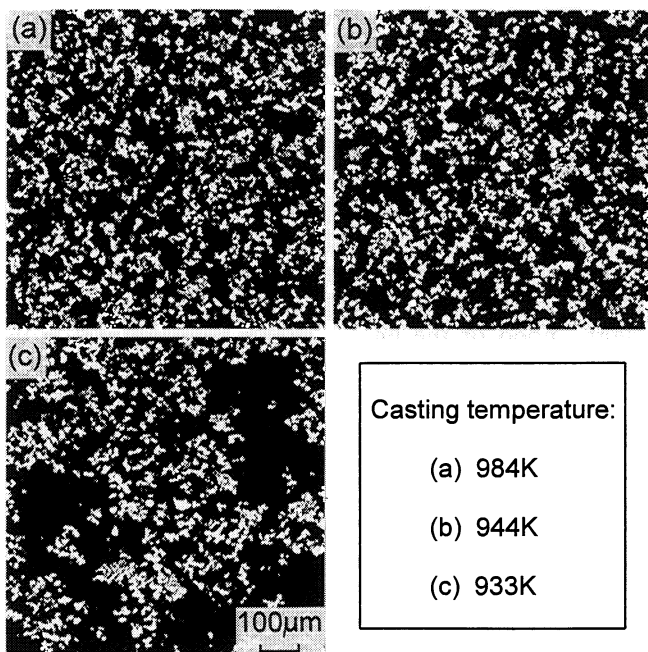


Fig. 7 Microstructures of the composite layer in the specimens shown in Fig. 6.

tion of coarse particles near the boundary when the casting temperature of the slurry is much higher than the final peritectic temperature. In stir-mix-quenching, intermetallic crystals nucleate copiously in the fluid clumps of the second melt, when the second melt is quenched in

the first melt, as illustrated in Fig. 9(a). The intermetallic crystals are dispersed throughout the mixed alloy, and then a semi-solid slurry, which consists of fine intermetallic crystals and liquid with a hyperperitectic composition, is formed (Fig. 9(b)).

When the slurry keeps the homogeneous distribution of the particles during cooling down to the neighborhood of the final peritectic temperature, it is not possible that only a part of the particles are coarsened. Therefore, visible coarsening of the particles near the boundary is not observed in Fig. 8(b) or (c).

On the other hand, in the case of Fig. 8(a), the residual liquid in the mixed-alloy slurry contained a considerable amount of solute Cr at the moment of the centrifugal casting. The mass of the intermetallic phase crystallized from the residual liquid was 1.5% of the mixed alloy in the case of Fig. 8(a), while that was 0.1% in the case of Fig. 8(b) and 0% in the case of Fig. 8(c). In the case of Fig. 8(a), therefore, the segregation of intermetallic particles can cause a part of the particles to grow coarser than in the other cases.

The sedimentation behavior of the intermetallic particles in the centrifugally cast slurry is as follows. A particle that is suspended in molten metal is submitted to a centrifugal acceleration of $\gamma(=\omega^2 r)$, where r is the distance between the particle and the rotation axis and ω is the angular velocity of the mold. The force balance equation on the particle due to centrifugal and viscous forces can be expressed as follows:

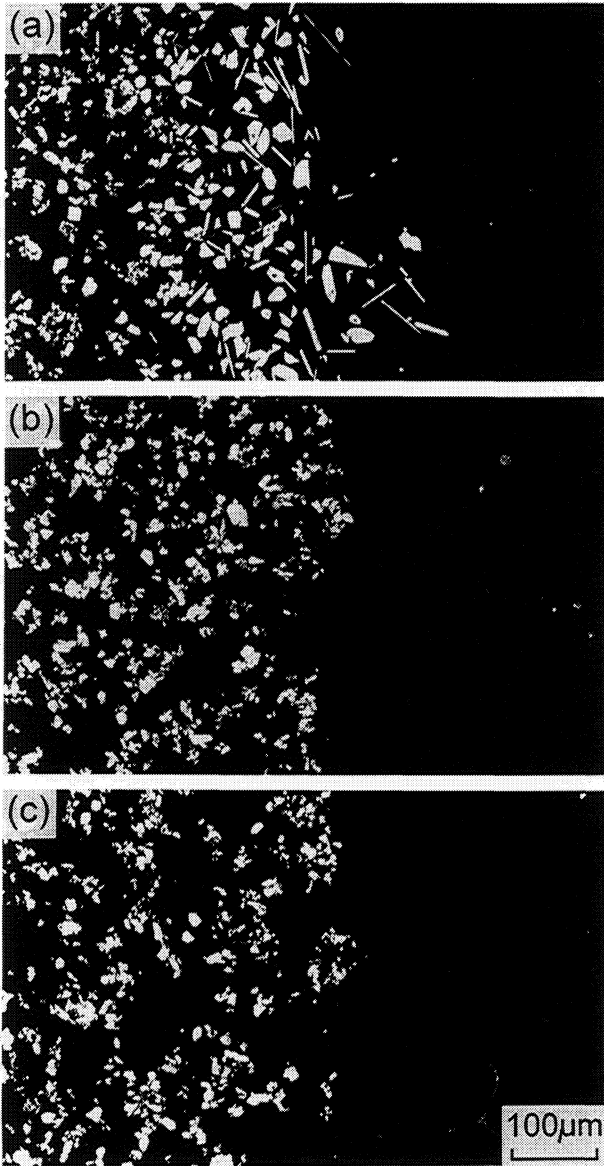


Fig. 8 Microstructures near the boundary between the composite layer and the intermetallic-free region in the specimens shown in Fig. 6. Casting temperature: (a) 984 K, (b) 944 K, (c) 933 K.

$$\begin{aligned} & (4/3)\pi R^3 \omega^2 r (\rho_P - \rho_L) - 6\pi \eta R (dr/dt) \\ & = (4/3)\pi R^3 \rho_P (d^2 r/dt^2) \end{aligned} \quad (1)$$

where R is the radius of the particle, η is the viscosity of the molten metal, and ρ_P and ρ_L are the density of the particle and the molten metal, respectively.

Equation (1) can be solved to get the position and migration speed, respectively, of the particle at any time t as follows:

$$\begin{aligned} r = r_0 \{ & [(a+s)/2s] \exp [(-a+s)t/2] \\ & + [(-a+s)/2s] \exp [(-a-s)t/2] \} \end{aligned} \quad (2)$$

$$dr/dt = r_0(b/s) \{ \exp [(-a+s)t/2] - \exp [(-a-s)t/2] \} \quad (3)$$

where r_0 is the distance of the particle from the rotation axis at $t=0$, and

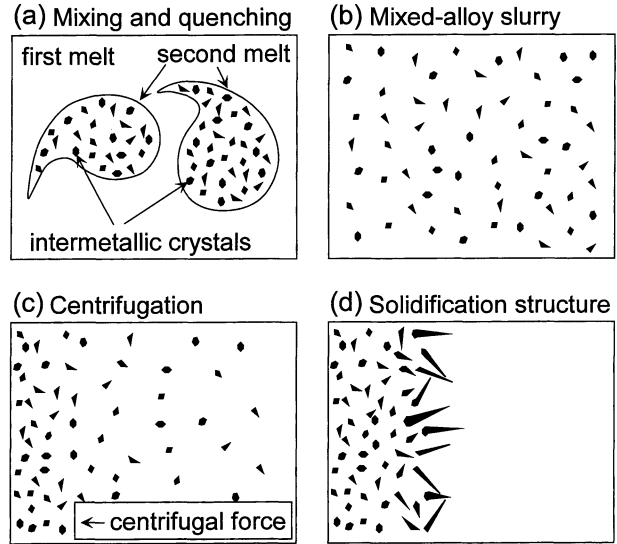


Fig. 9 Schematic illustration of the formation mechanism of the coarse composite layer.

$$a = 9\eta / 2R^2 \rho_P \quad (4)$$

$$b = \omega^2 (\rho_P - \rho_L) / \rho_P \quad (5)$$

$$s = (a^2 + 4b)^{1/2}. \quad (6)$$

Equation (3) indicates that the migration speed of the particle is proportional to r_0 . Therefore, particles whose initial position is near the inner periphery tend to sediment most slowly. These particles are in favorable circumstances to be supplied with solute Cr from surrounding liquid during and after the sedimentation (Figs. 9(c) and (d)), and as a result, they can grow coarser.

Figure 10 shows the profiles of mean particle size measured along the radial direction of the specimens shown in Figs. 6(a), (b), and (c). It is noteworthy that profiles (a) and (b) exhibit a “V” shape. We presume that the shape of the curves results from the inner-outer increase caused by centrifugation and the outer-inner increase caused by crystal growth during and after the sedimentation. The average size of intermetallic crystals at the moment of centrifugal casting is greater in the case of Fig. 6(b) than in the case of Fig. 6(a). On the contrary, the increase in size after the casting in the case of Fig. 6(a) is greater than that in the case of Fig. 6(b). In Fig. 10, the particle-size level of profile (b) is higher than that of (a) in the neighborhood of the outer periphery, while the opposite relation is observed near the boundary between the composite layer and the intermetallic-free region. These results are consistent with the above presumption.

For profile (c) in Fig. 10, the particle size is relatively small and almost uniform throughout the composite layer. The refinement of the intermetallic crystals may result from fragmentation caused by agitation during the final peritectic reaction (liquid phase + $\text{Al}_7\text{Cr} \rightarrow \alpha$ -phase).

4. Hardness profile in the fine composite layer

Figure 11 shows a typical hardness profile along the

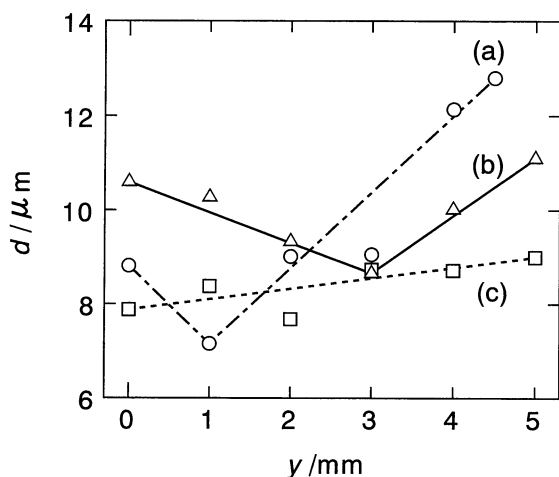


Fig. 10 Profiles of mean particle size in the radial direction of the specimens shown in Figs. 6(a), (b) and (c). y : distance from outer periphery of the specimen. d : mean particle size of intermetallic crystals.

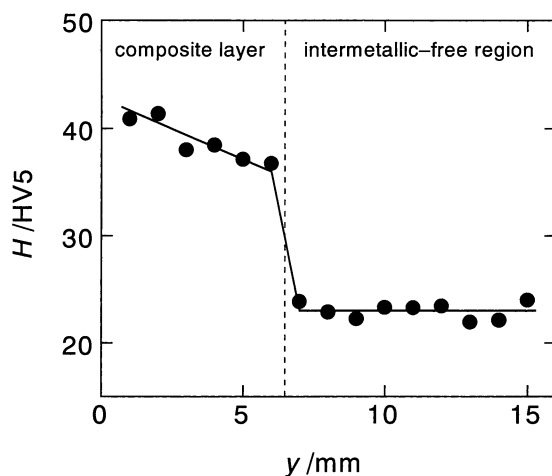


Fig. 11 Hardness profile in the radial direction of the specimen centrifugally cast at 944 K. H : Vickers hardness.

radial direction of the specimen centrifugally cast at 944 K. The measurements were carried out on the transverse cross section at the 1/2 height of the specimen, and the average of five measurements is plotted in the figure. The broken line in the figure indicates the boundary between the composite layer and the intermetallic-free region. In the composite layer, a graded hardness distribution is observed. The hardness decreases gradually as

the distance from the outer periphery increases. The hardness near the outer periphery is double that in the intermetallic-free region.

IV. Conclusions

We attempted to fabricate a tubular surface composite with fine intermetallic crystals on its outer periphery, by the centrifugal casting of semi-solid slurry of an hyperperitectic Al-Cr alloy produced by stir-mix-quenching. The results of our investigation are summarized as follows:

(1) A hyperperitectic Al-Cr alloy slurry containing fine intermetallic crystals can be produced by stir-mix-quenching.

(2) A cast pipe with a fine composite layer along the outer periphery can be produced by centrifugal casting of the mixed-alloy slurry.

(3) The structure of the composite layer is influenced by the casting temperature of the slurry. When the casting temperature is excessively higher than the final peritectic temperature, relatively coarse intermetallic crystals may form near the boundary between the composite layer and the intermetallic-free region.

(4) In the completely refined composite layer, the hardness profile exhibits a inner-outer increase along the radial direction. The hardness near the outer periphery is double that in the intermetallic-free region.

REFERENCES

- (1) T. Ohmi, Y. Kimura, Y. Itoh, M. Kudoh and K. Matsuura: *J. Japan Inst. Metals*, **60** (1996), 490-496. (or *Materials Trans., JIM*, **38** (1997), 879-885.)
- (2) T. Ohmi and M. Kudoh: *J. Japan Inst. Metals*, **54** (1990), 700-705.
- (3) K. Murata, M. Takada, T. Nakata and T. Umeda: *IMONO*, **64** (1992), 537-542.
- (4) K. Murata, H. Harada, T. Nakata and T. Umeda: *IMONO*, **66** (1994), 110-115.
- (5) Y. Fukui, N. Yamanaka, Y. Watanabe and Y. Oya-Seimiya: *J. Japan Inst. Light Metals*, **44** (1994), 622-627.
- (6) T. Ohmi, K. Minoguchi, M. Kudoh, Y. Itoh, K. Matsuura and K. Ishii: *J. Japan Inst. Metals*, **58** (1994), 324-329.
- (7) T. Ohmi, K. Minoguchi, M. Kudoh, Y. Itoh and K. Matsuura: *J. Japan Inst. Light Metals*, **44** (1994), 504-509.
- (8) T. Ohmi, K. Minoguchi, M. Kudoh, Y. Itoh and K. Matsuura: *J. Japan Inst. Metals*, **58** (1994), 1311-1317.
- (9) J. L. Murray: *Binary Alloy Phase Diagrams*, 2nd edition, Ed. by T. B. Massalski, ASM, (1990), pp. 138-140.
- (10) D. M. Smith, J. A. Eady, L. M. Hogan and D. W. Irwin: *Metall. Trans.*, **22A** (1991), 575-584.