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ANALYSIS OF REFINING OF ALUMINUM USING FRACTIONAL SOLIDIFICATION WITH FORCED CONVECTION

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Abstract

Segregation of iron atoms during the refining of aluminum using the fractional solidification has been analyzed. The redistribution of the solute atoms was influenced by the solidification and solid-liquid interface tangential velocities. The purity of aluminum increased with decreasing solidification velocity and increasing tangential velocity. The diffusion boundary thickness was inversely proportional to the square root of the tangential velocity in the cylindrical rotating system. Effects of the tangential and solidification velocities on the effective redistribution coefficient were investigated. The calculated refining curves were in good agreement with measured values.

Introduction

Three layer electrolytic refining, zone refining and fractional solidification methods are being used to refine aluminum[1]. The fractional solidification method is becoming popular due to its lower energy consumption. The fractional solidification method is classified into the remelting process and the forced convection process. The remelting process gives rise to refining during solidification and further refining through remelting and removing of higher solute regions by heating of the refined solid. However the remelting method has drawbacks of a lower productivity and necessity of a refined temperature control[2,3]. The forced convection method has advantages of simpler operation and facilities[4,5,6]. Analysis of the forced convection method has not been made due to the fact that the process involves complex interaction between solidification and fluid phenomenon of molten metal. In this study, analyses have been made of effects of solidification velocity and solid-liquid interface tangential velocity on removing of a solute element of iron from aluminum.

Experimental Methods

The chemical composition of aluminum used in the experiment is given in Table 1.

Table 1. Chemical composition of aluminum. (ppm)

Element	Fe	Si	Ti	V	Ga	Cr	Al
Conc.	1200	300	40	100	180	20	bal.

Experimental set up for refining by the forced convection method is shown in Fig.1. Aluminum of 3 kg was melted in a 2 kW electric furnace. The rotating cooling pipe of high purity graphite, with one end being closed, was connected to a DC motor by which rotation could be controlled up to 2000 rpm. Solidification and stirring could be achieved by the rotating graphite pipe, dimension of which was 30mm in outer dia., 16mm in inner dia. and 100mm in length. A 304 stainless pipe was connected to the graphite pipe. Air was blown into the inside of the rotating tube through a gas blowing pipe installed inside the tube. The air flow rate was controlled by a flow meter. The rotating velocity of the cooling graphite pipe was measured by a digital tachometer. The melt was held at 660°C, the melting point of aluminum. The concentration of iron was analyzed by ICP at locations 10mm below the top surface of the refined solid aluminum.

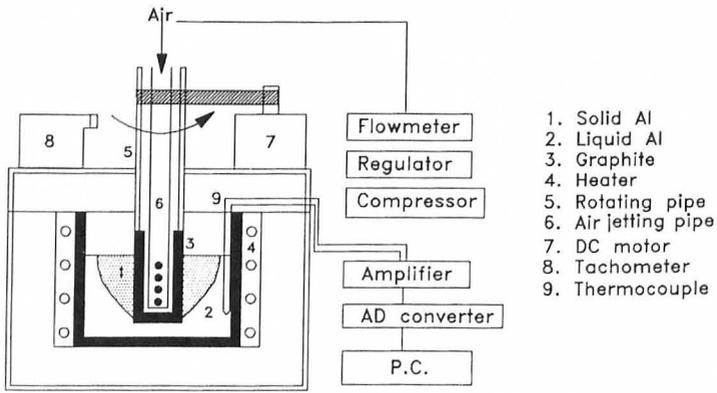


Fig. 1. The Schematic illustration of the apparatus.

Experimental Results

Fig. 2 shows refining ratios, C_s/C_0 with C_s and C_0 being the solute concentration by weight in the solid and the initial solute concentration, respectively, of iron measured at an air flow rate of 70 l min^{-1} as a function of distance from the outer surface of the graphite pipe which rotated at speeds of 500, 1000 and 1500 rpm. The refining ratio of iron measured at a given rotating velocity decreased to a minimum value and then increased with increasing distance from the outer surface of the graphite pipe. For a given distance from the outer surface, the refining ratio decreased with increasing rotating speed. Fig. 3 shows the refining ratios of iron measured at air flow rates of 35, 70 and 300 l min^{-1} as a function of distance from the outer surface of the graphite pipe which rotated at a speed of 1000 rpm. The refining ratio of iron measured at a given air flow rate decreased to a minimum value and then increased with increasing distance from the outer surface of graphite pipe. For a given distance from the surface, the refining ratio decreased with decreasing air flow rate.

Discussion

Refining of Al using the fractional solidification with forced convection is achieved by

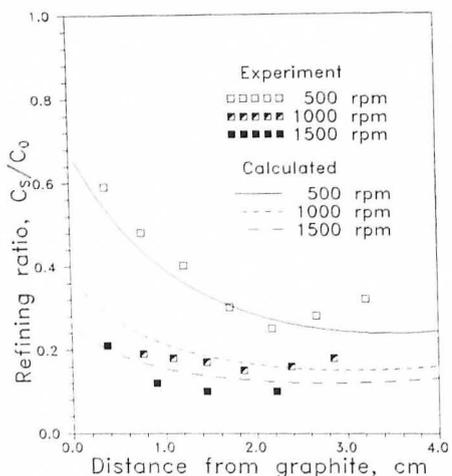


Fig. 2. Effect of rotating speed on refining ratio of Fe at an air flow rate of 70 l/min⁻¹.

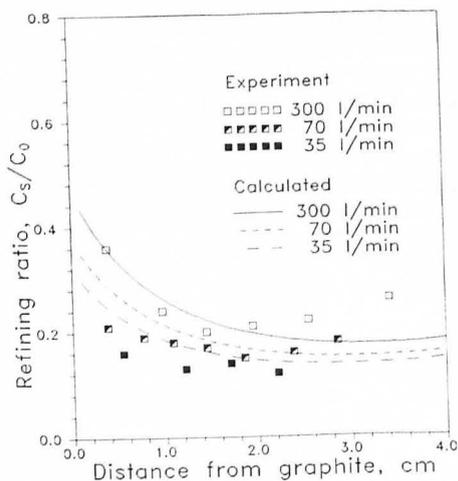


Fig. 3. Effect of air flow rate on refining ratio of Fe at a rotating speed of 1000 rpm.

segregation. An appropriate solidification model is necessary to analyze the process. Diffusion of solute in the solid is usually neglected as compared to its diffusion in the liquid. The assumptions of no solute diffusion in the solid, the two phases across the solid-liquid interface being in local equilibrium, and the rapid and complete solute diffusion in the liquid lead to Scheil's equation.

$$C_s = kC_0(1-f_s)^{k-1} \quad (1)$$

where f_s is the fraction by weight of the solid in the alloy and k the equilibrium redistribution coefficient. However, even in the case of severe convection in the liquid, it is difficult to achieve the perfect mixing. Burton et al. took this fact into account by introducing a diffusion boundary layer[7]. They assumed that the solute transport within the diffusion boundary layer is possible only by diffusion and complete mixing in the liquid outside of the diffusion layer. The assumption lead them to derive the following expression called the effective redistribution coefficient.

$$k_e' = \frac{k}{k + (1-k)\exp(-R\delta/D_L)} = \frac{k}{k + (1-k)\exp(-\Delta)} \quad (2)$$

where R is the solidification velocity, δ the thickness of the diffusion boundary layer and D_L the solute diffusion coefficient in the liquid. The effective redistribution coefficient k_e' approaches the equilibrium redistribution coefficient k with decreasing R and δ . Eq.(2) which is rigorously applicable to the planar interface system may be applied to the cylindrical system as in this study, since the diffusion boundary layer thickness is very small as compared to the melt size. The diffusion boundary layer thickness δ is known to be independent of the solidification rate at very small solidification rates but influenced by the solidification rate as the solidification rate increases[7,8].

When a melt flows on a plate, δ may be express as[9]

$$\delta = 3 D_L^{1/3} \nu^{1/6} U^{-1/2} x^{1/2} \quad (3)$$

where ν is the dynamic viscosity, U the fluid velocity and x the distance from the plate edge. In the case of rotational fluid flow as in this study, the quantity of Δ in eq.(2) is assumed to depend on the following expression.

$$\Delta = a R^n U_S^{-1/2} \quad (4)$$

where a and n are constants in a given experimental condition. Therefore eq.(2) should be modified to the following equation.

$$k_e = \frac{k}{k + (1-k) \exp(-a R^n U_S^{-1/2})} \quad (5)$$

And Scheil's equation can be modified to

$$C_S = k_e C_0 (1 - f_S)^{k_e - 1} \quad (6)$$

Rearranging eq.(5), we obtain

$$\ln a + n \ln R = \ln \left[U_S^{1/2} \ln \frac{k_e(1-k)}{k(1-k_e)} \right] \quad (7)$$

or

$$\ln a - \frac{1}{2} \ln U_S = \ln \left[R^{-n} \ln \frac{k_e(1-k)}{k(1-k_e)} \right] \quad (8)$$

The values of C_S/C_0 are known to depend on the radius of solidified body, r as shown in Figs. 2 and 3. The value of f_S was measured as a function of r from the specimen of Fig. 4, which was obtained at an air flow rate of 70 l min^{-1} . The air flow was interrupted for 10 s in intervals of 2 min, and the solidified specimen was sectioned and etched to measure the interrupted marks, from which f_S was evaluated as a function of r , as shown in Fig. 5. The best fitting of the data in Fig. 5 yields

$$f_S = 0.01571 (r-1.5)^2 + 0.048422 (r-1.5) - 0.001883 \quad (9)$$

where unit of r is cm.

The tangential velocity U_S at the solid-liquid interface can be calculated from the rotational velocity f or the number of rotation per unit time and the value of r , as $U_S = 2 \pi r f$. The solidification velocity is difficult to measure. Therefore the velocity is calculated using a heat transfer equation. If heat flows uniformly through the cooling graphite pipe from the melt, solidification will proceed radially from the graphite pipe and the cylindrical coordinate system can be used in the heat transfer calculation. The temperatures of the solidification front and the melt were assumed to be equal to the melting point of aluminum, 660°C , because of forced convection of the melt. When room temperature air is blown into the graphite pipe, a temperature distribution from the inside of the pipe to the solid-liquid interface can be drawn as in Fig.6. Since the heat flow per unit time through each heat resistance is the same, the total heat flux may be expressed as[10]

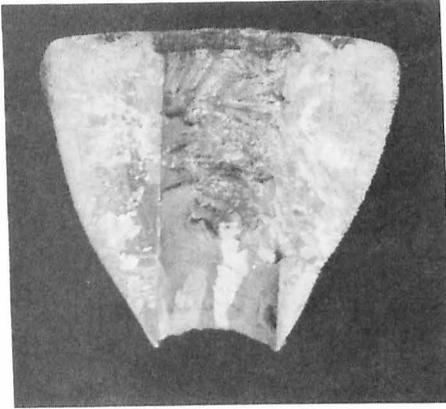


Fig. 4. Longitudinal shape of the specimen solidified under forced convection.

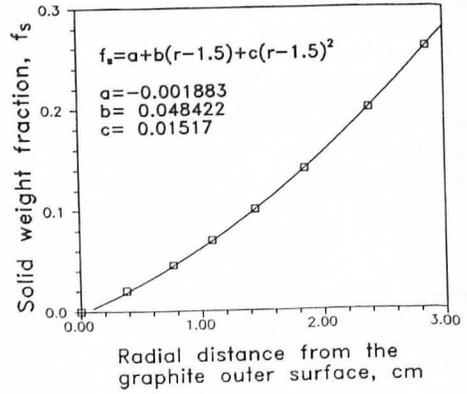


Fig. 5. Relations between distance from the the graphite pipe outer surface and solid weight fraction.

$$Q = \frac{T_f - T_i}{\frac{1}{2\pi r_1 l \bar{h}_i} + \frac{\ln(r_2/r_1)}{2\pi l k_1} + \frac{\ln(r/r_2)}{2\pi l k_2}} \quad (10)$$

where r_1 and r_2 are the inner and outer radii of the graphite pipe, T_f and T_i are the melting point of aluminum and room temperature, k_1 and k_2 are the heat conductivities of the graphite and aluminum, l is the melt height and \bar{h}_i is the mean heat transfer coefficient at the air-graphite interface. The heat flux of the solid-liquid interface q may be expressed as

$$q = \frac{Q}{2\pi r l} = \frac{T_f - T_i}{\frac{r}{r_1 \bar{h}_i} + \frac{r \ln(r_2/r_1)}{k_1} + \frac{r \ln(r/r_2)}{k_2}} = L \frac{dr}{dt} \quad (11)$$

where L is the latent heat of fusion of aluminum. Therefore the radial solidification velocity R can be expressed as

$$R = \frac{dr}{dt} = \frac{A}{r (\ln r - B)} \quad (12)$$

where $A = \frac{T_f - T_i}{L} k_2$, $B = \ln r_2 + \frac{k_2}{k_1} \ln \left(\frac{r_1}{r_2} \right) - \frac{k_2}{r_1 \bar{h}_i}$.

Thermophysical properties used in the calculation are given in Table 2.

Table 2. Thermophysical properties of aluminum and graphite[11].

Property	T_f	T_i	k_1	k_2	L
Units	°C	°C	W/mK	W/mK	J/m ³
Values	660	20	17	210	9.5×10^8

The value of \bar{h}_i depends on the cooling air flow rate. The value of \bar{h}_i at a given air flow rate was obtained so that it could yield the same f_s as the measured. Thus, the \bar{h}_i values were found to be 238, 409 and 919 $\text{Wm}^{-2}\text{K}^{-1}$ at air flow rates of 35, 70 and 300 l min^{-1} . Fig.7 shows the solidification velocities calculated using eq.(12) as a function of the radius of solidified body.

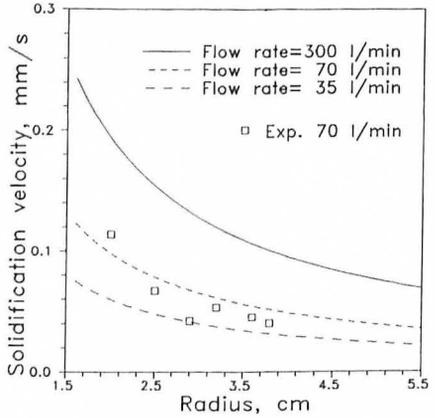
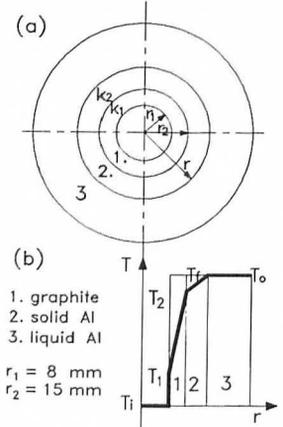


Fig. 6. (a) The schematic shape of the specimen and (b) temperature profile in the solidification system.

Fig. 7. Variation of solidification velocity with the radius of solidifying aluminum.

The solidification velocity decreases with increasing radius and decreasing air flow rate. The measured values of U_s and k_e , and the calculated values of R were used to plot in accordance with eqs. (7) and (8), as shown in Figs. 8 and 9.

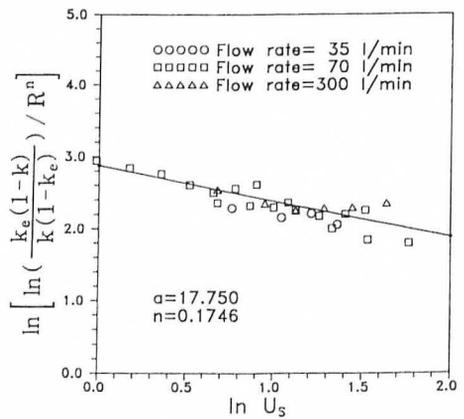
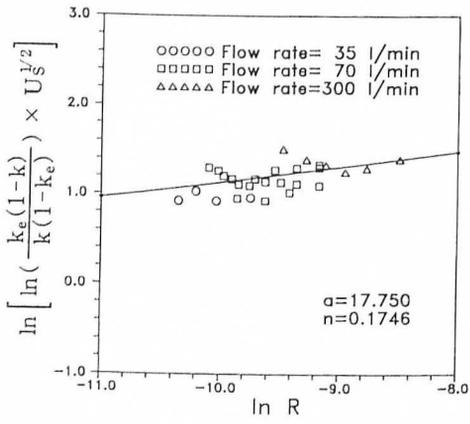


Fig. 8. Relations between tangential velocity of the rotating solid surface and effective redistribution coefficient of Fe for various experimental conditions.

Fig.9. Relations between solidification velocity of the rotating solid surface and effective redistribution coefficient of Fe for various experimental conditions.

The best fitting of the data in Fig.8 gives $n=0.1746$ and $a=17.750$. These values gives straight line in Fig.9, the slope of which is $-1/2$ in a good agreement with the theoretical value. The good linear relations in data in Figs.8 and 9 imply that the assumptions we made are reasonable. Once all the values of parameters in eqs.(5) and (6) are known, we are now in position to calculate the effective redistribution coefficient as a function of tangential velocity(Fig.10) or solidification velocity(Fig.11), within tangible experimental ranges. At a given solidification velocity, the effective redistribution coefficient decreases with increasing tangential velocity. At a given tangential velocity, the redistribution coefficient increases with increasing solidification velocity. The effect of tangential and solidification velocities on effective redistribution coefficient is shown in Fig.12. The contour plots of Fig. 12 may give us useful information on important process variables of solidification and tangential velocities to obtain predetermined effective redistribution coefficient, which in turn determines refining ratios.

In order to know applicability of the values of parameters obtained in the analysis, calculation has been made of the effect of rotation rate of the graphite cooling pipe at an air flow rate on refining of aluminum. The values of R and U_s at a moment are substituted into eq.(5) to give k_e , which is substituted into eq.(6) to yield the refining ratio of Fe, C_s/C_o . The calculated results are shown in Figs. 2 and 3 along with the experimental data. Agreement between the measured and calculated results is satisfactory. It follows from the analysis that the decrease in the refining ratio of iron with increasing distance from the graphite pipe can be attributed to the increase in the tangential velocity which in turn gives rise to the decrease in the effective redistribution coefficient. However as solidification proceeds, the solute concentration in the liquid increases and in turn the refining ratio increases. This effect is negligible in the earlier stage, but becoming important in the later stage.

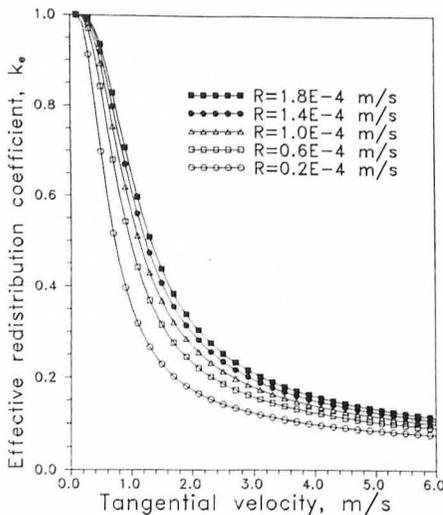


Fig.10. Effect of tangential velocity on effective redistribution coefficient for various solidification velocities.

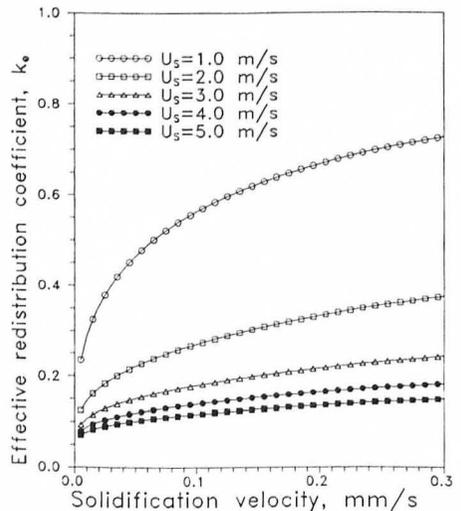


Fig.11. Effect of solidification velocity on effective redistribution coefficient for various tangential velocities.

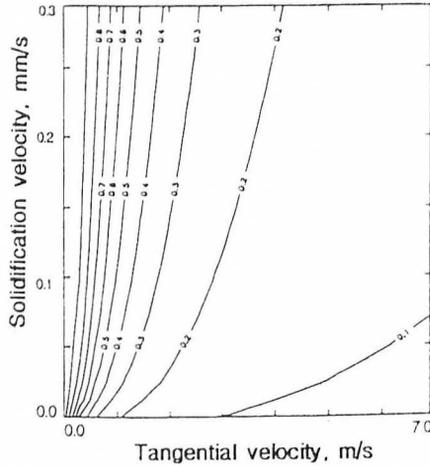


Fig.12. Contours of effective redistribution coefficient with at various solidification and tangential velocities.

Conclusions

1. The effective redistribution coefficient decreases with decreasing solidification velocity and increasing tangential velocity.
2. The diffusion boundary thickness is inversely proportional to the square root of the tangential velocity in a given solidification velocity.
3. The calculated refining curves using the proposed effective redistribution coefficient are in good agreement with measured values.

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