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Relationship between columnar crystal spacing and electric current density in unidirectional solidification of monophase Cu-Al alloy ①

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[Abstract] On the basis of previous theoretical inferential relationship between the columnar crystal spacing and the density of electric current applied during unidirectional solidification, the effect of current density on the columnar crystal spacing was discussed and analyzed, and the experiment was made to verify the theoretical relationship. The results show that at fast solidification speed the columnar crystal spacing decreases with increasing the density of electric current, while at slow solidification speed the columnar crystal spacing increases with increasing the density of electric current. The critical conditions for the evolution of columnar crystal spacing were confirmed. The calculated values concerning the spacing and the density are consistent with the experimental results.

[Key words] unidirectional solidification; columnar crystal spacing; effect of electric current; Cu-Al alloy

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1 INTRODUCTION

In the study of effect of electric current on the unidirectional solidification, it has been found that during the solidification, when the electric current passes through the interface between the liquid and solid, the morphological stability of the interface increases with increasing the electric current[1]; and the relationship formula between the columnar crystal spacing and the density of electric current has been drawn based on the M-S theory. The conclusion that under the condition of sub-rapid solidification the columnar crystal spacing reduces with increasing the current density has been confirmed[2], based on this conclusion, the mechanism of grain-refining of electric current during solidification has been discussed in detail[3~5]. In the previous discussion, the results have not dealt with thoroughly the effect of the speed of solidification. The results in the Ref. [2] dealt with only the conditions of sub-quick solidification. Recently it has been further found that when the stability of solid-liquid interface morphology increases, different solidification speed can cause different influence on the columnar crystal spacing structure formed during solidification. In this paper the effects of both the solidification speed and the density of electric current on the columnar crystal spacing are discussed in detail, and the relevant results are verified by experiments.

2 THEORETICAL ANALYSIS

2.1 Relationship between electric current density and columnar crystal spacing

When electric current passes through the solid-liquid interface, the theoretical relationship between the columnar crystal spacing d_1 and the density of electric current can be expressed as[2]

$$d_1 = (3\Delta T')^{1/2} G_L^{-1/2} (\cos\theta)^{1/2} (-p'/3)^{-1/4} \tag{1}$$

where $\Delta T'$ is the temperature difference between solid frontier and non-equilibrium solidus line; G_L is the temperature gradient at solid-liquid interface frontier;

$$p' = -(g_L + g_S)/[2mG_C D_L^2/k_0 v^2 - 2T_m\Gamma]$$

$$g_L = \lambda_L G_L^0/\bar{\lambda};\ g_S = \lambda_S G_S^0/\bar{\lambda};$$

the parameters G_L^0 and G_S^0 are, respectively, the temperature gradient within the liquid and the solid of solid-liquid interface when the interface is at flat state, and

$$\theta = \frac{1}{3}\arccos[-q'/(-p'/3)^{3/2}]$$

while

$$q' = -(S_e^L - S_e^S)/[\bar{\lambda}(2mG_C D_L^2/k_0 v^2) - 2T_m\Gamma]$$

S_e^L and S_e^S are the amount of exothermic heat of liquid and solid, respectively, within unit volume and unit time caused by electric current[6], and

$$S_e^L = J^2/k_e^L,\ S_e^S = J^2/k_e^S$$

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J is the density of electric current, the parameters k_e^L and k_e^S are the conductivity of liquid and solid, respectively; $\bar{\lambda} = (\lambda_S + \lambda_L)/2$, and the parameters λ_S and λ_L are thermal conductivity of solid and liquid, respectively; m is the slope of liquidus line of the alloy; G_C is the gradient of solute concentration of the liquid at solid-liquid interface; D_L is the diffusion-coefficient of the solute; k_0 is the balance distribution coefficient; v is the solidification speed; T_m is the melting point of the pure metal; $\Gamma = \sigma/L$, and σ is the specific surface energy of the solid-liquid interface, L is the latent heat of solidification.

2.2 Effect of electric current density on columnar crystal spacing

Since the effect of energy coefficient Γ of solid-liquid interface and concentration gradient G_C on d_1 when the electric current passes through the interface have been discussed in detail in Ref. [2], there is no need to re-describe it herewith.

It can be seen from Eqn. (1) that the influencing tendencies of θ on p' and d_1 are identical when θ is of practical meaning. However, the calculated result shows that the influencing effect of θ is far less than that of p'. Thereafter, for the sake of simplicity, the influencing factor of θ is ignored, and the effect of p' on d_1 is only considered in this paper.

When p' increases, d_1 decreases, or vice versa. From qualitative analysis of Eqn. (1), it can be seen that under the conditions of v being constant, the density of electric current J is increased, then p' is increased, resulting in decreasing of the spacing d_1 of columnar crystal. But in reality when there is an electric current passing through solid-liquid interface, the speed of solidification v will be decreased accordingly, thus p' is deceased, resulting in increase of d_1. That is to say that the effect of electric current on p' depends on the comprehensive effect of the two items in the denominator part of the formula of p'.

In the unidirectional solidification processes, when there is an electric current passing through solid-liquid interface, the total quantity of heat released in the metal is sum of the quantity of heat released when the temperature decreases from the molten state to the melting point of the metal, solidifying latent heat, the quantity of heat released when temperature decreases from the melting point to room temperature, and the quantity of heat produced by electric current in the system. Based on this point, the relationship between the solidifying speed and the density of electric current can be formed by

$$v = \left[q - \frac{\pi}{4}a^2 l\left(\frac{1}{k_e^S} + \frac{1}{k_e^L}\right)J^2\right] / \left(\frac{\pi}{4}a^2\rho_L G_L C_L + \frac{\pi}{4}a^2 L\right) \tag{2}$$

where q is the quantity of heat released from metal in unit time; the parameters of C_L and l are the specific heat capacity of the liquid metal and the length of the sample, respectively.

For the sake of simplicity, H is used to express the denominator part of the p' formula, thus

$$H = 2mG_C D_L^2/k_0 v^2 - 2T_m\Gamma \tag{3}$$

If H increases, p' will decease. If Eqn. (2) and formula $\Gamma = (\sigma_0 + wJ^2)/L$ [7] are substituted into Eqn. (3), then

$$H = \frac{2mG_C D_L^2}{k_0} \cdot \left\{\frac{q - \pi a^2 l[1/(1/k_e^S + 1/k_e^L)]J^2/4}{\pi a^2 \rho_L G_L C_L/4 + \pi a^2 L/4}\right\}^{-2} - \frac{2T_m\sigma_0}{L} - \frac{2T_m w}{L}J^2 \tag{4}$$

where σ_0 is the specific surface energy of solid-liquid interface under the condition of no electric current; w is the coefficient related with solute concentration, electrical parameters of both solid and liquid of the metal.

In order to clarify the effect of electric current on H, Eqn. (4) is differentiated with respect to the density of electric current as

$$\frac{dH}{dJ} = J \cdot \left\{\frac{m\pi^3 a^6 l G_C D_L^2(\rho_L G_L C_L + L)^2(1/k_e^S + 1/k_e^L)}{8k_0[q - \pi a^2 l(1/k_e^S + 1/k_e^L)J^2/4]^3} - \frac{4T_m w}{L}\right\} \tag{5}$$

When $dH/dJ = 0$, which means that the columnar crystal spacing d_1 is not affected by the density of electric current, q can be regarded as q_0, and its critical condition can be obtained as

$$q_0 = \sqrt[3]{\frac{32k_0 T_m w}{Lm\pi^3 a^6 l G_C D_L^2(\rho_L G_L C_L + L)^2(1/k_e^S + 1/k_e^L)}} + \frac{\pi a^2 l(1/k_e^S + 1/k_e^L)}{4}J^2 \tag{6}$$

In reality, when $q > q_0$, resulting in $dH/dJ < 0$, it implies that with increasing the density of electric current, the columnar crystal spacing d_1 is decreased; however, when the actual $q < q_0$, resulting in $dH/dJ > 0$, it is illustrated that with increasing the density of electric current, the columnar crystal spacing d_1 is increased.

From the definition of q, it can be found that the increasing of q means the increasing of solidification speed, or vice versa. From the above analysis, it can be concluded that when the density of electric current increases, the different spacing results of columnar crystal can be obtained with respect to different solidification speed, as demonstrated by following experiment.

3 EXPERIMENTAL STUDIES

3.1 Experimental results

Unidirectional solidification experiments were carried out with the Cu-5% Al alloys by using the self-made electro-slag induction continuous unidirectional solidification complex[8]. The diameter of the sample is 10mm. The complex can ensure that the electric current passes through the solid-liquid interface, and the density can be adjusted at will within a certain limit.

The experiment starts at the conditions of the temperature gradient of solid-liquid interface frontier being 70 K/cm and the solidification speed being 0.008 cm/s. When the density of electric current increases from 0 to 128 A/cm^2, the temperature gradient is enhanced to 100 K/cm, at the same time, the solidification speed is increased to 0.024 cm/s, then the density of current is increased gradually to 764 A/cm^2 under these conditions. Observations of the solidified structure were made onto the samples obtained at different electric current density, and the evolution of columnar crystal spacing with respect to the density of electric current was evaluated.

The solidification structures in different density of electric current have been observed, as demonstrated in Ref. [8], and the experimental results of the effect of electric density on the columnar crystal spacing are shown in Fig. 1. It can be seen clearly in the figure that at the left of the dotted line , the cooling speed is 0.56 ℃/s, which can be classified as slow solidification speed; while at the right of the dotted line, the solidification speed is about 3 ℃/s, which can be classified as sub-rapid solidification speed[9]. Under the condition of slow solidification speed, the columnar crystal spacing d_1 increases near linearly with increasing the density of electric current; while under the condition of fast solidification speed, the columnar crystal spacing d_1 decreases almost linearly with increasing the density of electric current. The experimental results are consistent with the above theoretical analysis conclusion.

3.2 Comparison between experimental results and calculated data

Taking the experimental temperature gradient of the solid-liquid interface frontier, the experimental conditions as well as relevant physical parameters (see Table 1) of Cu-5%Al alloy into the Eqn. (6), it can be obtained that when the density of electric current is 46~ 800 A/cm^2, the heat q_0 released from the solidified metal per unit time is between 2 400~ 4 300 J/s. When the solidification speed is 0.008 cm/s and 0.024 cm/s, the actual quantity of heat released is about 1 500 J/s and 4 600 J/s, respectively. Consequently, it can be seen that when the heat q at slow solidifying speed is less than the critical solidifying value q_0, increasing the density of electric current results in the increasing of columnar crystal spacing; while when the heat q at fast solidifying speed is greater than the critical solidifying value q_0, increasing the density of electric current results in the decreasing of columnar crystal spacing . The experimental results agree well with the calculated values.

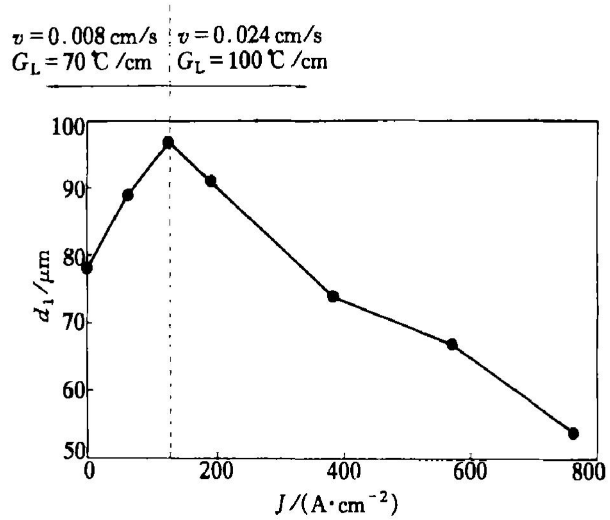

Fig. 1 Influence of electric current density on columnar crystal spacing under different solidification conditions

Table 1 Physical parameters of Cu-5%Al alloy

Parameter	Value	Reference
$D_L/(cm^2 \cdot s^{-1})$	5×10^{-5}	[10]
k_0	0.89	[11]
m	2.7	[11]
T_m/K	1 356	[11]
$\rho_L/(g \cdot cm^{-3})$	7.2	[11]
$k_e^S/(\Omega \cdot cm)$	1.334×10^7	[11]
$k_e^L/(\Omega \cdot cm)$	$1.185\,4 \times 10^8$	[11]
$\sigma_0/(J \cdot cm^{-3})$	1.17×10^{-5}	[11]
$L/(J \cdot cm^{-3})$	1.88×10^3	[12]

4 CONCLUSIONS

1) At the condition of sub-rapid solidification speed the columnar crystal spacing decreases with increasing the density of electric current, while at slow solidification speed the columnar crystal spacing increases with increasing the electric current density.

2) The critical conditions for the evolution of columnar crystal spacing can be theoretically calculated. The calculated values are consistent with the experimental results.

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