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Mechanism of reduction of columnar dendrite spacing in unidirectional solidification caused by electric current passing through solid-liquid interface①

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[Abstract] Considering the S-L interface morphology stability, the S-L interface energy, the Joule heat produced by electric current at the S-L interface, and the change of solute concentration at the S-L interface indirectly caused by electric current, the mechanism of reduction of columnar dendrite spacing in unidirectional solidification caused by electric current passing through solid-liquid interface was studied. The following conclusions can be drawn that: 1) under sub-rapid solidification condition, increasing electric current density will improve the stability of S-L interface, thus decreasing the columnar dendrite spacing; 2) there are two ways by which the increase of electric current decreases the columnar dendrite spacing, one is promoting the splitting of the protruding tips at the S-L interface, the other is promoting the forming of new convex parts at the bottom of the concave interface.

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

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1 INTRODUCTION

Choi et al[1] noted that in the electric slag remelting process, the solidified microstructure fines with increasing electric current density. GU et al[2] found that in the process of direct current passing through the S-L interface, the increase of electric current density would reduce the intercellular spacing. In the study of electric slag induction continuous unidirectional solidification of Cu-Al alloy, the authors[3] also came to the following conclusion that the columnar dendrite spacing almost decreases linearly with increasing the electric current density at a temperature gradient of 100 ℃/cm and a solidification rate of 0.024 cm/s. Although the mechanism of electric current fining solidified microstructure has been studied previously[1,2,4], there exist the problems of theoretic analyses being not complete and experimental evidences being not enough. Considering the following factors such as the stability of S-L interface morphology, the S-L interface energy, the Joule heat produced by current at the interface, and the change of solute concentration distribution at the interface indirectly caused by electric current, the mechanism that the increase of electric current density reduces the columnar dendrite spacing is studied in detail in this work.

2 EXPERIMENTAL METHODS AND RESULTS

The unidirectional solidification experiments were carried out on the self-made equipment[3]. The raw material is a Cu-5%Al alloy. The diameter of the specimens is 10 mm. There are two methods of changing the current density in the solidification process, namely increasing the current density from zero to 256 A/cm^2 directly or increasing the current density from 128 A/cm^2 to 256 A/cm^2. A temperature gradient of 100 ℃/cm and a solidification rate of 0.024 cm/s were used.

The change of solidified microstructure with current density was observed and the effect of current density on columnar dendrite spacing was analysed.

The variation of solidified microstructure with increasing current density is shown in Fig.1, which indicates that the increase of current density reduces the columnar dendrite spacing. The self adjustment of spacing is realized by the following two ways, i.e. the splitting of the protruding tips of the columnar dendrites (see Fig.1(a)), and the splitting of the columnar dendrite interfaces (see Fig.1(b)). When the magnitude of current density increased is large,

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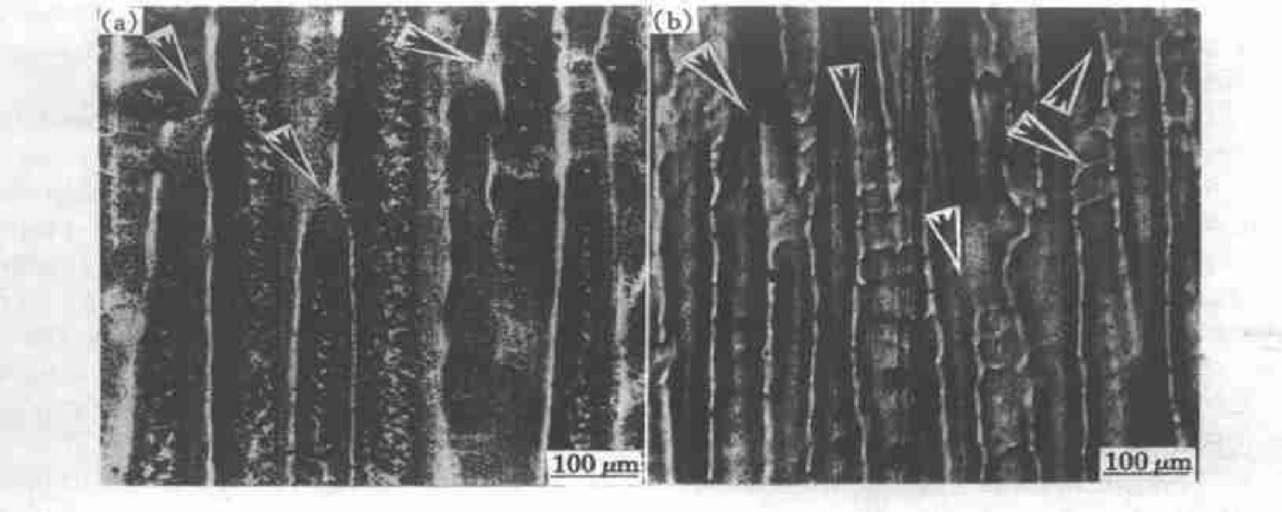

Fig.1 Effect of increasing current density on solidified microstructure
(a) —Increasing current density from 128 A/cm^2 to 256 A/cm^2; (b) —Increasing current density from 0 A/cm^2 to 256 A/cm^2

the splitting of interface is predominant; when the magnitude is small, the splitting of protruding tips is predominant.

3 DISCUSSION

3.1 Effect of current increasing stability of S-L interface

In Ref.[5], the authors established a criterion of the stability of S-L interface morphology under the effect of current, analysed the effect of current on the stability of S-L interface morphology in detail, and drew a conclusion that the stability of S-L interface morphology increases with increasing current density at a temperature gradient of 100 ℃/cm and a solidification rate of 0.024 cm/s. However, the increase of the stability of the interface morphology will produce two contrary effects on the primary dendrite spacing. In the development from planar crystal to coarse dendritic crystal, it increases the primary dendrite spacing; while in the development from coarse dendritic crystal to quasi-planar crystal, it decreases the primary dendrite spacing. The theoretic analyses and the experimental result that the increase of current density decreases the columnar dendrite spacing show that the studied solidification process is in the development stage from coarse dendritic crystal to quasi-planar crystal, but further identification is needed.

Up till now, there is still no reliable criterion to determine the transformation of the solidified structure morphology from coarse dendritic crystal to fine dendritic crystal. WANG and HU[6] recently proposed the nonlinear kinetics theory of the interface morphology stability of single phase alloys. This theory pointed out that under the conditions of sub-rapid and rapid solidification and at a certain temperature gradient, the condition of stability for dendrite is $mG_C < G_L + T_m \Gamma\omega^2$. The term mG_C promotes constitutional undercooling and makes the interface instable; the term $T_m \Gamma\omega^2$ reflects the effect of interface energy and makes the interface stable, and the larger the current density, the larger the value of this term. At a certain G_L, if the above inequality holds, then larger value of $T_m \Gamma\omega^2$ will mean higher stability and higher interface energy, consequently finer dendrites. This description gives out the criterion for the transformation from coarse dendrites to fine dendrites, which cannot be obtained by the linear theory of S-L interface stability. The solidification rate of 24 ℃/s in this work belongs to the sub-rapid solidification range, so it is not difficult to determine that the inequality $mG_C < G_L + T_m \Gamma\omega^2$ is completely satisfied. It can be concluded that the transformation of the solidified structure is at the stage from coarse dendrites to fine dendrites, thus as the current density increases, the increase of stability of the S-L interface morphology will lead to the decrease of columnar dendrite spacing.

3.2 Effects of current segregation in solid phase

3.2.1 Production of Joule heat by current

Due to the difference of electro-conductivity between solid and liquid phases, the current will segregate in the solid phase at the S-L interface, as shown in Fig.2. Therefore, the distribution of Joule heat produced by current will inevitably be nonuniform, which will give rise to different growth rates at different parts. If there is no current, the S-L interface will move towards the liquid phase in the original form of sine wave, while there is current, the Joule heat produced by current will concentrate at the protruding tips of the solid phase, thus slowing down the growth rates at those positions. But at the concave parts of the S-L interface, the higher solute concentrations make the solute atoms diffuse from these positions to the protruding tips of the convex parts. The temperatures at these positions also are relatively lower. Thus, the growth rates at the bottoms of the concave parts will be accelerated relatively, and consequently new little protruding tips are formed, as shown in Fig.2(b). These new protruding tips will run after the original protruding tips and this process will not stop until the formers catch up with the latters, as shown in Fig.2(c) and (d). At this moment, the current completes the process of increasing S-L interface disturbance waves. When the current is increased continuously, the above process will be repeated and thus the disturbance waves will be increased further. According to the viewpoint of Langer et al[7], the columnar dendrite spacing will be decreased further. The splittings of grain boundaries in Fig.1(b) are the traces of newly-formed protruding tips at the concave parts of the interface caused by increasing current density.

3.2.2 Effect of current indirectly changing solute concentration of liquid phase at S-L interface

At the protruding tips of the S-L interface, the temperature is the highest and the solute concentration in the liquid in front of the interface is the lowest, thus the solute atoms at both sides of each protruding tip will diffuse transversely towards its center and the growth rate at the center of the protruding tip will reduce to the lowest, so the protruding tips will become flat, as shown in Fig.3(b). The transverse diffusion of the solute atoms at the center of each tip front becomes more difficult, thus there occurs concaving phenomenon, which is equivalent to forming crack sources at the protruding tips, as shown in Fig.3(c). In the continuous growth process, the solute atoms at the newly-formed concave parts segregate more seriously, thus accelerating the development of the concave parts. In the same time, the current segregate at the newly-formed protruding tips, and the Joule heat produced by current will slow down their growth rates. If the former effect is stronger than the latter, then the splitting of the protruding tips will aggravate continuously. This process will not stop until the Joule heat produced by current makes the growth rate of the newly-formed protruding tips equal that of the original protruding tips, then the current finishes the process of promoting the splitting of the protruding tips, thus increasing the number of the disturbance waves, as shown in Fig.3(d). The branchings at the protruding tips of the columnar dendrites shown in Fig.1(a) are the traces of the splittings of the protruding tips caused by increasing current density.

When the change of current density is larger, it is easier to form new protruding tips at the concave parts of the S-L interface. When the change of current density is smaller, it is easier to produce splittings at the protruding tips due to the easier diffusion of the solute atoms. Therefore when the change of the current density is different, there occur the two cases shown in Fig.1.

3.3 Effect of increasing interface energy by current

It is well known that the S-L interface energy always plays the role of resuming the interface to plane. The relation between the current density and the S-L interface energy can be described as follows: $\sigma_{S-L} = \sigma_0 + wI^2$, where σ_{S-L} and σ_0 are specific S-L interface energies with and without current effect, and w is a coefficient related to solute concentration and electric parameters of solid and liquid phases. It is clear that the increase of current density will increase the interface energy, thus promoting the interface developing towards plane. There are two ways for realizing this development. One is reducing the disturbance magnitude and wave number at the same time, finally

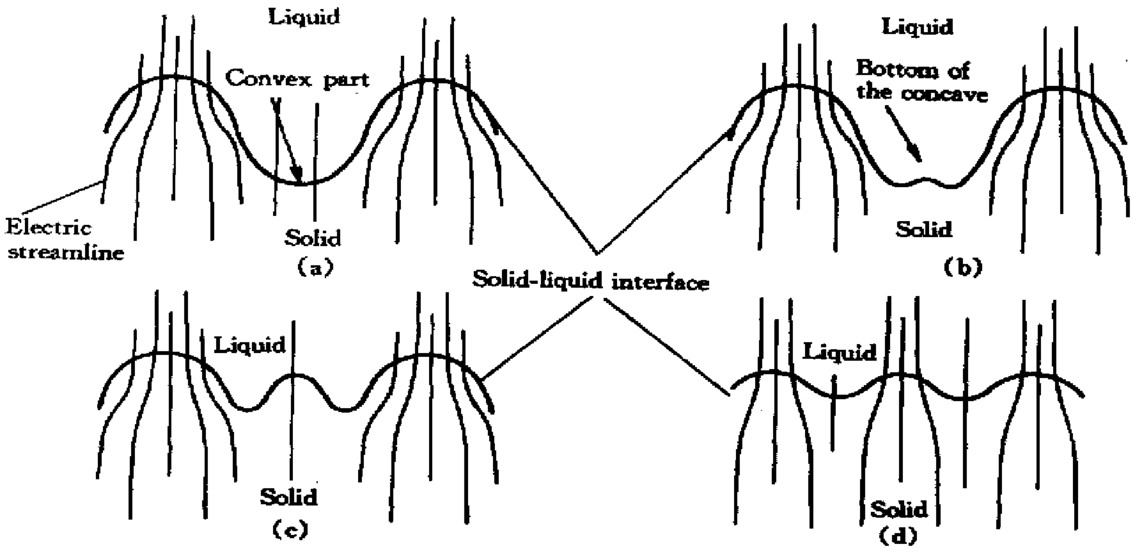

Fig.2 Scheme showing effect of current segregation on disturbance wave number of interface
(a) —Original state; (b) —Appearance of protruding tips; (c) —Development; (d) —Reaching stability

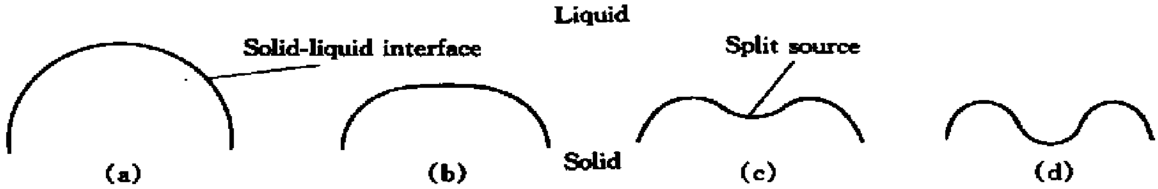

Fig.3 Scheme showing splitting of protruding tips of convex parts of interface
(a) —Original state; (b) —Curvature reduction of protruding tips;
(c) —Appearance of splitting sources; (d) —Reaching stability

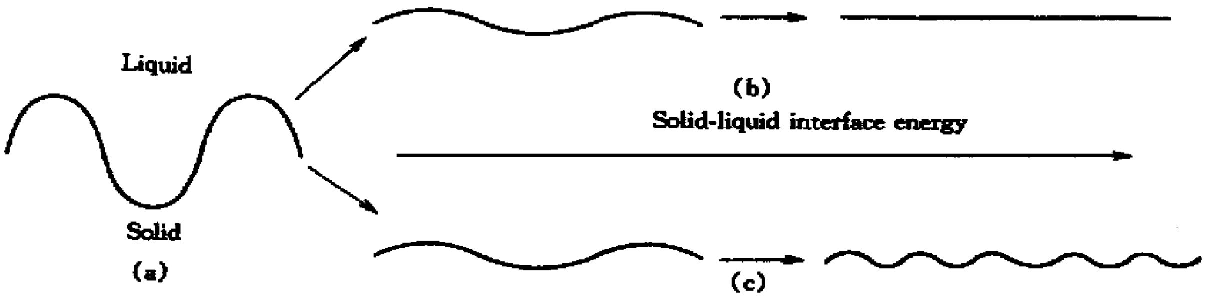

Fig.4 Scheme showing interface developing towards plane caused by L-S interface energy
(a) —Original state; (b) —Approaching equilibrium state; (c) —Deviating from equilibrium state

reaching the planar state, as shown in Fig.4(b); the other is increasing disturbance wave number while reducing disturbance magnitude, thus the interface presents the quasi-planar state with small magnitude and large frequency, as shown in Fig.4(c). The former is the development mode of near equilibrium solidification stage, while the latter is the development mode of the stage far from equilibrium solidification. The solidification in this work belongs to sub-rapid solidification and is in the stage far from equilibrium solidification, thus the effect of S-L interface energy on interface morphology corresponds to the development mode of Fig.4(c). Before this development mode reaches quasi-plane, the solidified morphology is fine columnar dendrites, and the larger the disturbance, the smaller the columnar dendrite spacing. When the quasi-planar state is reached, the interface morphology is macro-plane with small magnitude and large frequency.

4 CONCLUSIONS

1) Under the condition of sub-rapid solidification, the increase of current density increases the stability of the S-L interface morphology, and in the whole reduces the columnar dendrite spacing during solidification.

2) There are two ways by which the increase of current density reduces the columnar dendrite spacing, one is producing splittings at the protruding tips of the interface, the other is forming new protruding tips at the concave parts of the interface.

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