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THE INFLUENCE OF SOLIDIFICATION RATE ON THE MICROSTRUCTURE AND DISTRIBUTION OF SOLUTE DURING SOLIDIFICATION OF Al-Cu ALLOY

UTICAJ BRZINE OČVRŠĆAVANJA NA MIKROSTRUKTURU I RASPODELU PRIMESA TOKOM OČVRŠĆAVANJA LEGURE Al-Cu

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ABSTRACT

In this paper investigation of the influence of solidification rate on the microstructure and distribution of solute during the solidification of Al-2.2%Cu by Vertical Bridgman method was performed. The experiments were carried out over a range of solidification rates from 1.45×10^{-6} m/s to 8.71×10^{-5} m/s, under a protective nitrogen atmosphere. Within the range of investigated solidification rates the change of interface shape from planar-to-cellular has been observed. It was observed that the solidification rate at which the formation of cellular structures begins is greatly different from the calculated one, due to convection. According to distribution of the solute, the effective coefficient of distribution (k_e) was calculated for each solidification rate. Dependence of k_e on solidification rate has been presented.

Key words: Interface shape, solute distribution, Al-Cu alloy, Vertical Bridgman.

IZVOD

U okviru ovog rada ispitivan je uticaj brzine očvršćavanja na mikrostrukturu i raspodelu primesa tokom procesa očvršćavanja legure Al - 2,2%Cu metodom Vertikalni Bridgman. Eksperimenti su izvedeni u opsegu brzina od 1.45×10^{-6} m/s do 8.71×10^{-5} m/s, u zaštitnoj atmosferi azota. U ovom opsegu brzina uočena je promena oblika granične površine od ravne ka ćelijskoj. Konstatovano je da se brzina očvršćavanja pri kojoj počinje formiranje ćelijske strukture znatno razlikuje od teorijski izračunate brzine što je posledica prisutne konvekcije. Na osnovu raspodele koncentracija primese izračunat je efektivni koeficijent raspodele ke za svaku brzinu očvršćavanja. Takođe je predstavljena zavisnost ke od brzine očvršćavanja.

Klju_e reči: Oblik granične površine, raspodela primese, legura Al-Cu, metoda Vertikalni Bridgman

INTRODUCTION

The Bridgman technique has been used for a long time to grow crystals on a laboratory scale. The outstanding results obtained by AT&T with vertical Bridgman growth of large diameter GaAs or InP [1] clearly show the potential of such technique. There is therefore a need for analysing and optimising the growth parameters in the

Bridgman configuration. The results presented in this paper are part of a wider investigation of the influence of solidification conditions on the solidification and its properties. Precisely, the influence of the solidification rate on the shape of interface during solidification from the melt was investigated.

The classical theory of constitutional supercooling (CS) [2] is an important tool in order to predict solid-liquid (S-L) interface instabilities arising during controlled growth from the melt. For a given alloy the decrease of the parameter $\frac{G_L}{R}$, where G_L is the temperature gradient in liquid in front of the interface and R the solidification rate, controls the evolution of the S-L interface morphology. Even when the evolution of the instability is continuous, it is possible recognise discrete stages of the substructures [3]: a) planar interface; b) nodes or depressions at the interface; c) elongated or two-dimensional cells; d) regular or hexagonal cells; e) distorted or branched cells; f) dendritic cells or arrayed dendrites. This evolution operates trough a node mechanism that changes slightly depending on the alloy crystallography [3-5]. It was observed that the planar interface became unstable above a critical growth rate, R_{PC}, given approximately by:

$$\frac{G_L}{R_{PC}} = \frac{m_l\, C_0\, (1-K_0)}{k_0 D_L} \qquad (1)$$

where G_L is the temperature gradient in the liquid phase, R_{PC} the critical growth rate for a planar-to-cellular transition, C_0 the initial solute concentration, m_L the liquid line slope in the equilibrium diagram, k_0 the equilibrium coefficient of distribution, D_L the coefficient of diffusion in the liquid phase.

As the growth rate was increased above R_{PC} the interface morphology became oscillatory unstable, then, after appreciably larger growth rate, cellular and finally, at large velocities, dendritic. Near the critical rate, R_{PC}, the interface exhibited an irregular morphology. The unstable region lies between stable region for a planar interface (equation 1.) and the stable region of regular cells.

In the case of normal solidification (Vertical Bridgman method) the whole charge is molten and it solidifies from one end to other. The distribution of solute concentration in the solid phase can be described by equation (2) [6]:

$$C_S = k_0\, C_0\, (1 - g)^{k_0 - 1} \qquad (2)$$

where C_S is the concentration of solute in the solid phase, C_0 is the initial concentration of solutes in the liquid phase, k_0 is the equilibrium distribution coefficient and g is the fraction of solidified material.

In the real systems the effective distribution coefficient k_e is used instead of the equilibrium one:

$$C_S = k_0\, C_0\, (1 - g)^{k_e - 1} \qquad (3)$$

EXPERIMENTAL

Solidification of Al-2.2%Cu by Vertical Bridgman method was performed. The experiments were performed on the previously described equipment [7]. The experiments were carried out over a range of solidification rates from 1.45×10^{-6} m/s to 8.71×10^{-5} m/s, under a protective nitrogen atmosphere. The solidified samples were

longitudinal cut, polished, etched and then metallographic investigation of the microstructure was performed.

Optical emission spectrometry was used for chemical analysis of distribution of the solute concentration across the specimen.

RESULTS AND DISCUSSION

The assumption was made that the solidification rate is equal to the descending rate of the vessel.

Equation (1) was used to calculate R_{PC}, the rate at which the transition from plane interface to cell growth occurs. The $m = -3.4°C/\%$ was calculated from the
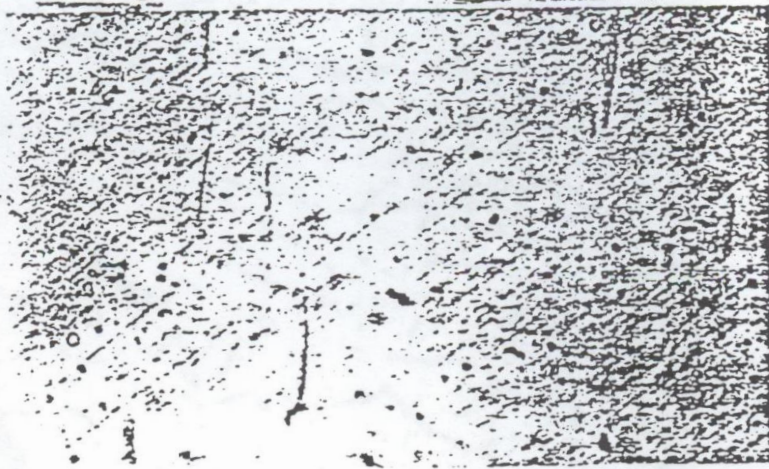

Figure 1 - Microstructure of sample solidified with rate $1.45x10^{-6}m/s$,
planar interface(x 100)
Slika 1 - Mikrostruktura uzorka dobijenog pri brzini očvršćavanja
od 1,45x10⁻⁶m /s, ravna granična površina (x 100)

Figure 2 - Microstructure of sample solidified with rate $2.90x10^{-6}m/s$
unstable interface (x 31.5)
Slika 2 - Mikrostruktura uzorka dobijenog pri brzini očvršćavanja
od 2,90x10⁻⁶m/s, nestabilna granična površina (x 31,5)

Al-Cu equilibrium diagram. Values for k_0 [8] and D_L [9 - 11] were taken from the literature and are 0.153 and 2.2×10^{-5} cm^2/s respectively. It has been assumed that temperature gradient in melt and in furnace is equal. Temperature gradient in the furnace was measured and G is estimated to be 14.5°C/cm. The calculated value for R_{PC} was 7.72×10^{-8} m/s.

It was observed that the solidification rate at which the formation of cellular structures begins is greatly different from the calculated value for R_{PC} [12-13]. Therefore, the experiments were carried out over a range of solidification rates from

Figure 3 - Microstructure of sample solidified with rate 4.35×10^{-6} m/s beginning of cellular structure (x 31.5)
Slika 3 - Mikrostruktura uzorka dobijenog pri brzini očvršćavanja od 4,35x10^{-6}m/s početak ćelijskog rasta (x 31,5)

Figure 4 - Microstructure of sample solidified with rate 8.71×10^{-5} m/s dendrites (x 31.5)
Slika 4 - Mikrostruktura uzorka dobijenog pri brzini očvršćavanja od 8,71x10^{-5}m/s dendriti (x 31,5)

1.45×10^{-6} m/s to 8.71×10^{-5} m/s. The microstructures of samples solidified at different rates are presented in Figures [1-4].

The beginning of connection of irregular cells is observed at rate 4.35×10^{-6} m/s. Therefore the R_{PC}^{exp} - experimentally estimated solidification rate, is 56.35 times grater than calculated R_{PC}. It is in agreement with previous experimental results [7, 13]. The existence of convection, as well as difficulties in the experimental metallographic defining the exact moment of planar-to-cellular transition can be the reasons for this deviation.

According to distribution of the solute across the sample, from the equation (3) the effective distribution coefficient k_e was calculated for each solidification rate [14] and k_e as a function of R was presented (Fig.5.).
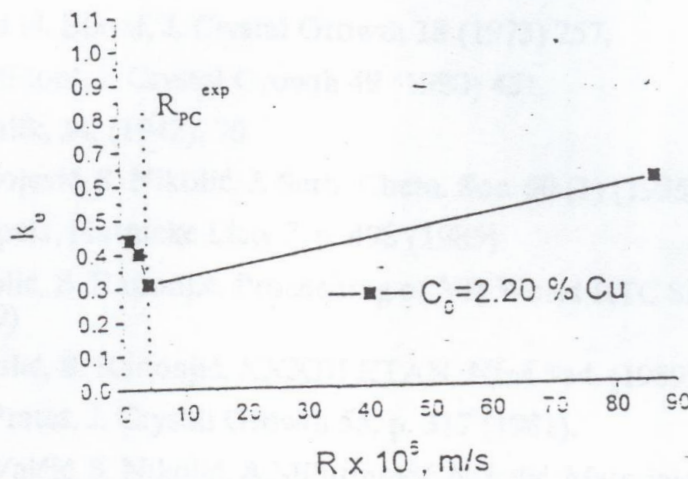

Figure 5 - Dependence of efective coeficient of distribution (k_e)
on solidification rate (R)
Slika 5 - Zavisnost efektivnog koeficijenta raspodele (k_e) od brzine
očvršćavanja (R)

There is a minimum on the dependence of k_e on the solidification rate. The minimum occurs at R_{PC}^{exp}. This indicates that there is a possibility of making correlation between two phenomena: interface shape and distribution of solute.

CONCLUSION

In this study the influence of the solidification rate on the microstructure and solute distribution was investigated. Solidification of Al-2.2%Cu alloy was performed using Vertical Bridgman method.

According to quantitative criterion the critical rate for plane-to-cellular transition (R_{PC}) was calculated to be 7.72×10^{-8} m/s. The beginning of connection of irregular cells is observed at rate 4.35×10^{-6} m/s. Therefore the R_{PC}^{exp} - experimentally estimated solidification rate, is 56.35 times grater than calculated R_{PC}. The existence of convection, as well as difficulties in the experimental metallographic defining the exact moment of planar-to-cellular transition can be the reasons for this deviation.

The effective distribution coefficient k_e was calculated for each solidification rate and k_e as a function of R was presented. The minimum of k_e occurs at R_{PC}^{exp}. This indicates that there is a possibility of making correlation between two phenomena: interface shape and distribution of solute.

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