SUBCRITICAL BIFURCATION FROM PLANAR TO CELLULAR INTERFACE IN Al - 0.5 wt.% Cu DIRECTIONALLY SOLIDIFIED

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ABSTRACT

Samples of the $Al - 0.5wt.\%\,Cu$ system were directionally grown under controlled conditions, to study the role played by the instabilities in the process relatives to the microstructure selection for a given values of interface velocity, and thermal gradient. Using an interface quenching technique and metallographic analysis in longitudinal and transversal cuts of the samples, we determine the transition mechanisms between the different stages of the growth, and associate them to the stability of the solidification front. We study the planar to a cellular transition in different conditions, and although the solidification parameters are in good agreement with the perturbations theory, when analyzing the amplitude of the
perturbations during the planar to a cellular transition, the same theory is not able to predict certainly the critical wavelength in this case. Also, we found a subcritical behavior during the transition from a planar to a cellular interface for the diluted $Al - Cu$ system, detecting a hysteresis behavior for the amplitude of the perturbations when it is increasing and then decreasing the interface velocity, through the threshold.

INTRODUCTION

It is well known that directional solidification is one of the principal techniques for the production of advanced engineering components since it allows to control the microstructure and the properties of the material [1,2]. In the directional solidification the processing variables are the interface velocity V, thermal gradient at the interface G_L, and the alloy composition C_0. By keeping constant the concentration and the thermal gradient, it is possible to study the microstructure behavior against the interface velocity, that becomes the control parameter of the problem. When an impure material with atomically rough solid-liquid interface is directionally grown, it may exhibit cellular or dendritic structure of solidification front. A planar front of solidification brings a homogenous solid, while a cellular or dendritic front forms a periodic microsegregation map in it. For this reason, the instabilities of a planar front and its development into a periodic array of cells and dendrites have received a particular attention from the metallurgist [2–4].

The nature of these patterns have been explained by the competitive effect between the surface tension, the solute diffusion, and the temperature gradient. The interpretation of the system stability in the basis of "constitutional supercooling" [5,6] has been improved with a complete dynamical out of equilibrium treatment [7–9]. For these studies, in the immediate vicinity of the transition, it is possible to obtain a Landau-like equation for the amplitude A_k, corresponding to a deformation wavevector k

$$\frac{dA_k}{dt} = a_0\,(k)\,A_k - a_1 A_k^3 \tag{1}$$

and, therefore, four different situations can be distinguished, according to the signs of the parameters a_0 (growth rate) and a_1 (Landau coefficient):

(i) $a_0 < 0; a_1 > 0$: the planar front is stable;
(ii) $a_0 > 0; a_1 > 0$: the stationary cellular deformation of amplitude $(a_0/a_1)^{1/2}$ is stable;
(iii) $a_0 < 0; a_1 < 0$: the planar front is stable but unstable to finite amplitude deformations, it is the subcritical instability case;
(iv) $a_0 > 0; a_1 < 0$: the planar front is completely unstable.

Many experimental investigations have been developed with thin samples of transparent organic systems, such as tetrabromomethane (CBr_4) or succinonitrile $(CH_2CN)_2$ [10,11], since they allow the direct observation of the interface dynamics under a microscope. However, as far as we know there are not any works developed in such sense in metallic samples. Available data on the planar to cellular transition have been reviewed by Cheveigné et al. [12] showing a reasonable accordance with the bifurcation concept. For this reason, we have performed a systematic study on the metallic system $Al - 0.5wt.\%\,Cu$ around the "constitutional stability", to determine the interface behavior during the planar to cellular transition and compare the results with the theoretical predictions of Cheveigné et al. for metallic systems.

EXPERIMENTAL

Directional solidification of the $Al-0.5wt.\%\,Cu$ samples was carried out in a Bridgman-like apparatus. The device consists of a mobile vertical chamber furnace of alumina core, where two heating coils with independent temperature controls have been located, maintained at fixed temperatures. This disposition brings a thermal gradient inside the chamber, where we placed the specimens to be directionally grown. The pulling system of the furnace was based in a steeper motor and a gear box, which permitted a translation velocity in the $0 - 10\ \mu m\, s^{-1}$ range. The displacement velocity was measured and controlled with a linear variable differential transformer (LVDT) and a signal conditioning interface, and the data was sent to a Personal Computer which drives the motor. This schemes provides a very precise displacement motion, resulting in a precision of $\pm\, 0.1\ \mu m\, s^{-1}$ [13,14].

Temperature measurements in the specimen were taken by using transverse CrNi-AlNi thermocouples which were made from $0.2\,mm$ diameter wire, isolated by using a ceramic coating. Temperature data was collected by using a data acquisition system, consisting of a 12 bits accuracy analog to digital converter and a multiplexor-low noise differential programmable preamplifier. This assembly also permits the cold junction correction to be carried out. Setting a reader window in $400 - 1000\,^{\circ}C$ range, the experimental error

Table I: Physical properties of $Al - Cu$ system

m	-2.6	$K\ (wt\%)^{-1}$
k	0.14	—
D_L	$3.0 \cdot 10^{-9}$	$m^2\, s^{-1}$
Γ	$0.9 \cdot 10^{-7}$	$K\, m$
k_S	210	$W\ (K\, m)^{-1}$
k_L	95	$W\ (K\, m)^{-1}$

was estimated in at $\pm 0.5\,^{\circ}C$. In our work, we fixed the thermal gradient at a value of $G_L = 2.5 \pm 0.05\,^{\circ}C\,mm^{-1}$, taking the velocity as the parameter control of the problem. The interface velocity was calculated in each time from the profile temperature vs. time obtained, through heat balance equations.

The alloy was prepared melting purity 99.999 % Al and adding the required amounts of eutectic $Al - 33.2\,wt.\%\,Cu$, this work was performed under continuous Ar atmosphere. To avoid or minimize the microsegregation in the preingots, they were cast in cylindrical quartz tubes of $8\,mm$ internal diameter by $250\,mm$ long, directly from the melt by making use of a depression of $4\,mm\,Hg$. The quartz tubes were internally coated using a based graphite paint. Some samples were sectioned and were chemically analyzed using X ray spectro-photometry. Physical properties of the used system can be seen in Table I.

RESULTS AND DISCUSSION

As an introduction to the problem, several runs were developed around the critical value at which the planar to cellular transition occurs. We found a value of $V_{C,Exp} = 0.5 \pm 0.05\ \mu m\,s^{-1}$; below this velocity value the microstructure was planar and stable, while above this velocity, the instabilities tend to grow, as can be seen in Figure **??** [14,15]. In both figures, the crystals were grown from bottom to top, the lower part is the resultant homogenous solid and the upper part is the quenched liquid. According to the linear stability theory we expected a theoretical value of

$$V_C = \frac{2kD_L}{(k_S/k_L + 1)\,m\,(k-1)}\frac{G_L}{C_0} \tag{2}$$

In our case, it brings a threshold velocity of $V_C = 0.58\ [\mu m\,s^{-1}]$, in such a way the experimental obtained value is in the same order than the theoretical one.

With this threshold value, we designed experiences which starting with a planar stable front at a velocity lesser than V_C and an increasing velocity, promoting the microstructure change, ending above the threshold. In this case, the initial velocity was $0.1\ \mu m\,s^{-1}$, and establishing a final velocity of $3\ \mu m\,s^{-1}$, the rate of velocity change was estimated in $2 \pm 0.1 \cdot 10^{-4}\ \mu m\,s^{-2}$. The microstructure evolved from planar to cellular and dendritic at quenching time. In Figure 1 a), the planar to cellular transition can be clearly seen. For this case, we determined a threshold value given by $V^{+}_{C,Exp} = 0.57 \pm 0.05\ \mu m\,s^{-1}$, which is a bit greater than the obtained under stable growth conditions.

In the opposite form, experiences were made starting from a velocity value greater than V_C and cellular microstructure with decreasing rate of growth, looking for a planar front of solidification at a velocity lesser than V_C. We started from a velocity of $4\ \mu m\,s^{-1}$ and then decreased the velocity at a rate that was estimated in $3 \pm 0.1 \cdot 10^{-4}\ \mu m\,s^{-2}$ until a final value of $0.1\ \mu m\,s^{-1}$. In this first case, the initial microstructure, which was composed by dendritic cells, experienced a change at a value of $V^{-}_{C,Exp} = 0.3 \pm 0.05\ \mu m\,s^{-1}$. Then we repeat from an initial velocity of $2\ \mu m\,s^{-1}$ to $0.1\ \mu m\,s^{-1}$ and a deceleration which was estimated in $1.8 \pm 0.1 \cdot 10^{-4}\ \mu m\,s^{-2}$. Here, the cellular to planar transition was more evident, as can be seen in Figure 1 b).

The results can be summarized by noting that the transition threshold shows a hysteresis behavior if the wavelength of the perturbations is represented against the velocity, increased and then decreased through the threshold value V_C, i. e. $V^{-}_{C,Exp} < V_{C,Exp} \lesssim V^{+}_{C,Exp}$,

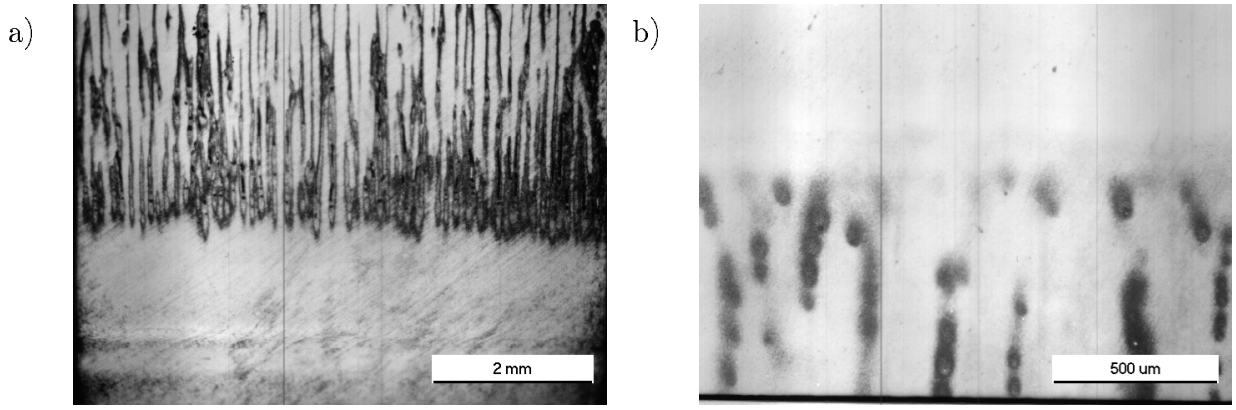

Figure 1: *a) Planar to cellullar transition at $V^{+}_{C,Exp}$; Cellullar to planar transition at $V^{-}_{C,Exp}$, starting from $V \simeq 2V_C$.*

as can be seen in Figure 2. In Figure 2, the arrows show the path of bifurcation, from a planar to a cellular interface as the velocity is increasing and the cellular to a planar interface when the velocity is decreasing, assuming a subcritical transition [14, 15]. The complete behavior could be understood in the following way: when the system starts from a planar interface the use of arbitrary small perturbations is valid, in such a way that the threshold is closer to the value given by the linear stability theory. Close to V_C there exists a set of wavenumbers which could grow and define a characteristic spacing. This fact is characteristic of a subcritical transition. When it starts from an existing spacing, the hypothesis of small perturbations no longer holds because a cellular state is predicted.

On other hand, in accordance with the linear stability theory, we expected a critical wavelength given by

$$\lambda_C = 2\pi \left(\frac{2\Gamma k D_L^2}{m\,(k-1)} \right)^{1/3} \frac{1}{C_0^{1/3}} \frac{1}{V_C^{2/3}} \tag{3}$$

which in this case is $\lambda_C \approx 520\ \mu m$. The experimental wavelength of the perturbations $\lambda_{C,Exp} \approx 125\ \mu m$, which is below in a factor of 4.2. This fact is in accordance with the suggestion made by Cheveigné et al. [12] who predicted a relation in the range 4.5 to 6 for the rate $\lambda_C/\lambda_{C,Exp}$. In addition to that, based in the comparison between the capilarity and composition length, the authors expect a range of variation of approximately $0.1\,V_C$. In this work, we found a range greater than the suggested by the authors [14, 15].

CONCLUSIONS

We have studied experimentally the system $Al - 0.5\,wt.\%\,Cu$ to determine the behavior of the interface during the transition from a planar to a cellular and from a cellular to a planar morphology. At first, when the velocity is increasing, we determined that the threshold $V^{+}_{C,Exp}$ appears at a value very close to that predicted by the linear stability criterium, V_C. At this value, the instabilities at the interface grow quickly, determining a spacing of the $\lambda_{C,Exp} \approx 125\,\mu m$, approximately 4.2 times below the theoretical value expected. At a decreasing interface velocity, the threshold appears at a value lesser than V_C, determining a hysteresis behavior for the transition [14, 15]. With this evidence we can justify that the transition for

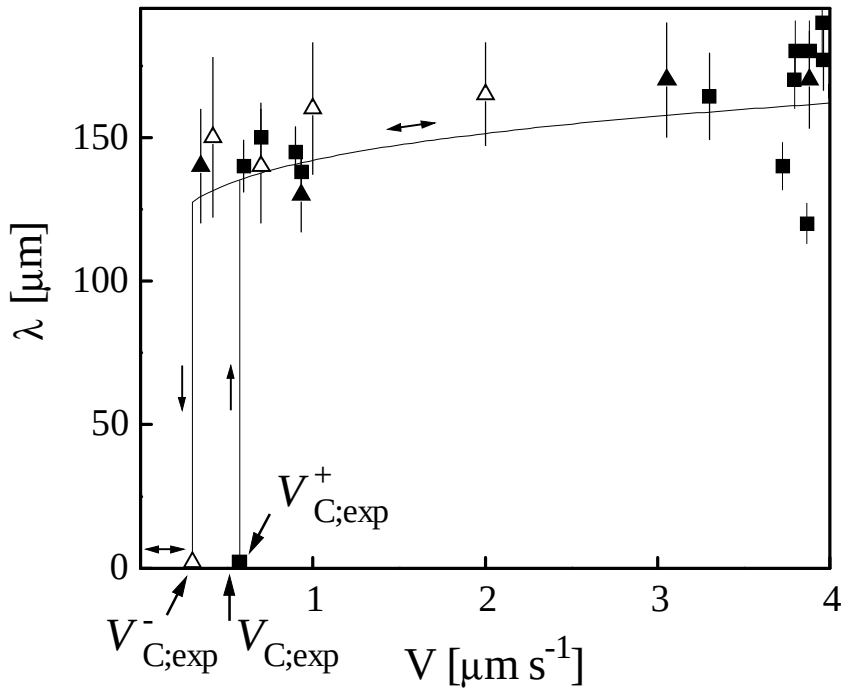

Figure 2: *Wavelength of perturbations vs. interface velocity. Squares: increasing speed. Triangles, open and closed: decreasing speed. Full line: interpretation as a subcritical bifurcation.*

this system is subcritical, as expected [12, 16]. The threshold variation found was greater than the $0.1\,V_C$ range proposed theoretically by Cheveigné [12].

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