EVOLUTION OF MUSHY ZONE IN DIRECTIONAL SOLIDIFICATION OF METALLIC ALLOYS

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ABSTRACT

The evolution of mushy zone during directional solidification has been investigated for three technical metallic alloys: a binary alloy, a ternary one and a multicomponental superalloy. The analysis of transverse sections within the mushy region yielded the solid fraction, f_S, as a function of temperature. It was found, that for alloys with a large amount of interdendritic eutectic the solid fraction curve deviated immense from the prediction of the Scheil model and the lever-rule. Only if the eutectic fraction is negligible, the theory can explain the experimental finding correctly.

KEYWORDS

directional solidification, mushy zone, fraction solid, metallic alloy

1 INTRODUCTION

The microstructure of an alloy has a large influence on the mechanical and physical properties of the material. Therefor a quantitative understanding of the evolution of a microstructure from the melt is of great importance. The change in the concentration of the liquid phase with temperature during the solidification process, defines the so-called solidification path. To calculate the microstructure evolution, and thus the solidification path, only very simple models are available. For technical alloys, which are in general multicomponental, the solidification path can lead to totally different predictions of relative phase amounts and of the occurrence of precipitations, depending on the model used.

In this work the evolution of the fraction solid, f_s, during directional solidification was studied for three technical alloys: a binary Al-7.8wt.%Si alloy, a ternary Al-5.5wt.%Si-1.5wt.%Cu alloy and a Ni-base superalloy SRR 99 (5.5 Al, 2.2 Ti, 8.5 Cr, 5.0 Co, 9.5 W, 2.8 Ta and 66.5 Ni in wt.%.). The experimental results are compared with each other and with the theoretical predictions.

2 EXPERIMENTAL

The experiments were performed in a modified Bridgman furnace. Cylindrical samples, 8 mm in diameter, were directionally solidified in alumina crucibles, with a constant withdrawal velocity of 1, 0.5 and 0.46 mm/min for the three alloys given above. Two thermocouples were located on the axis of the samples at different heights. After having reached steady-state growth conditions, the solidification process was interrupted by rapidly quenching with a Ga-In liquid metal cooling. After quenching, the mushy zone of the samples were metallographically investigated. **Figure 1a-c** shows typical transverse sections of the three alloys, taken from a section within the mushy zone. The microstructure of the quenched liquid between the dendritic structures was very fine and thus clearly be detectable. The dendritic fraction, often called solid fraction, f_S, was quantitatively measured on the transverse sections as a function of position (measured from the dendrite tips), by means of an interactive image analyzing system. Considering the measured temperature curves and assuming a uniform temperature within a section perpendicular to the axis of a sample, a position within the mushy region can be correlated with temperature. Thus f_S is estimated as a function of temperature difference from liquidus, $(T_L\text{-}T)$.

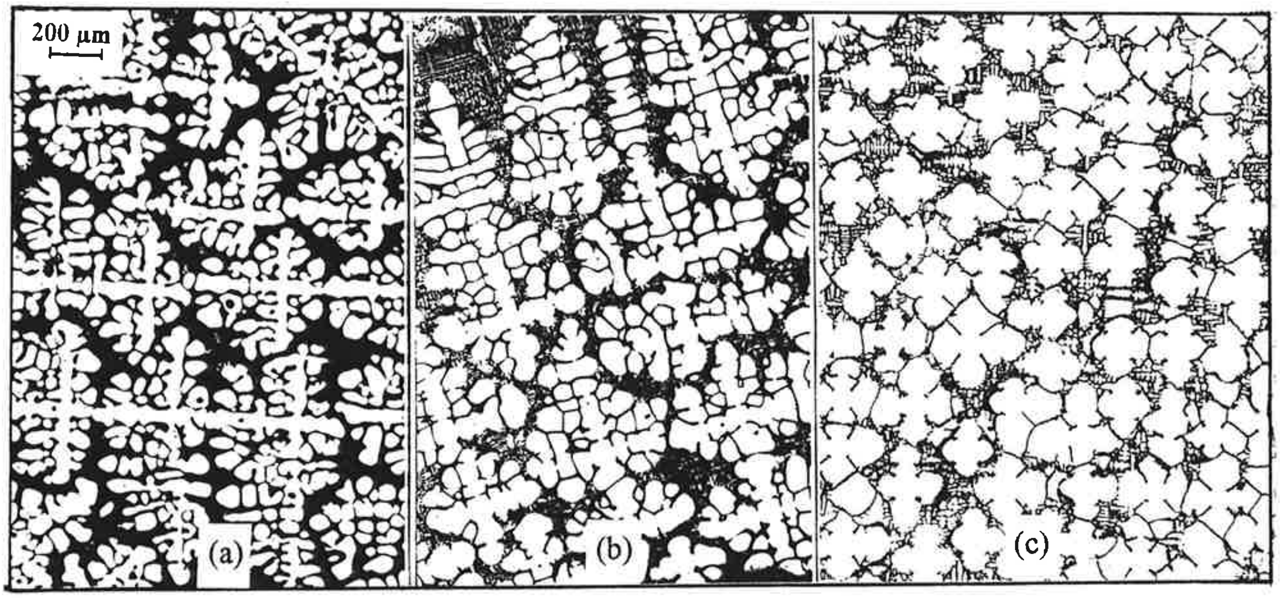


Fig. 1: Typical transverse sections in the quenched mushy zone of the investigated alloys.
(a) Al-7.8wt.%Si, (b) Al-5.5wt.%Si-1.5wt.%Cu and (c) superalloy SRR 99

3 THEORETICAL CALCULATION

For dendritic growth at low solidification velocity, the constitutional undercooling at the dendrite tip is small compared to the solid-liquid temperature interval ΔT_0 [1]. Therefor the tips can be considered to grow at the liquidus temperature T_L. Assuming rapid diffusion in the melt and no diffusion in the solid, the evolution of f_s within the mushy zone can be described, as first approximation, by Scheil´s equation [2]. If rapid diffusion is assumed also in the solid, the Lever-rule has to be used instead. Although both models are very simple, their application for multicomponental alloys is limited to systems where the phase diagram is known. In this work, the differential form of the Scheil model and the Lever-rule were combined with a commercial software program for thermodynamic calculations (ChemSage). Therefore f_s could be determined, according to the Scheil model or to the Lever-rule, even for the ternary Al-5.5wt.%Si-1.5wt.%Cu alloy.

For the superalloy SRR 99 no data for the thermodynamic calculation are available yet. However the following expressions, analog to the Scheil model and the Lever-rule, can be applied to a superalloy :

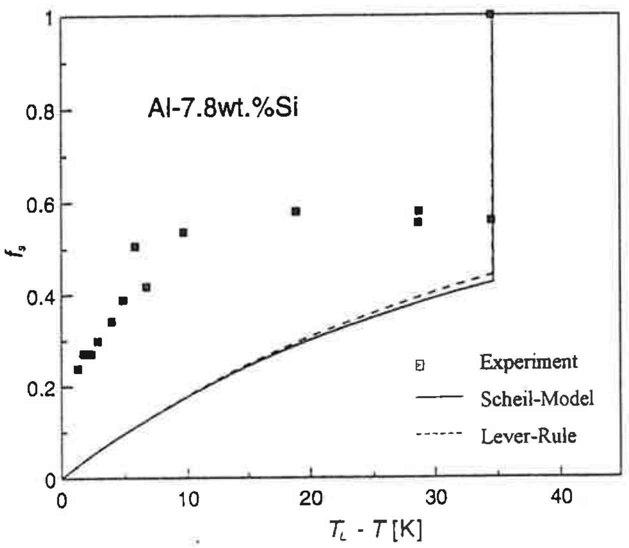
$$T_L - T = \frac{k}{1-k}\Delta T_0\left[(1-f_s)^{k-1} - 1\right] \qquad \text{(Scheil model)}$$

$$T_L - T = \frac{k}{1-k}\Delta T_0\left[\frac{1}{1-f_s(1-k)} - 1\right] \qquad \text{(Lever-rule)}$$

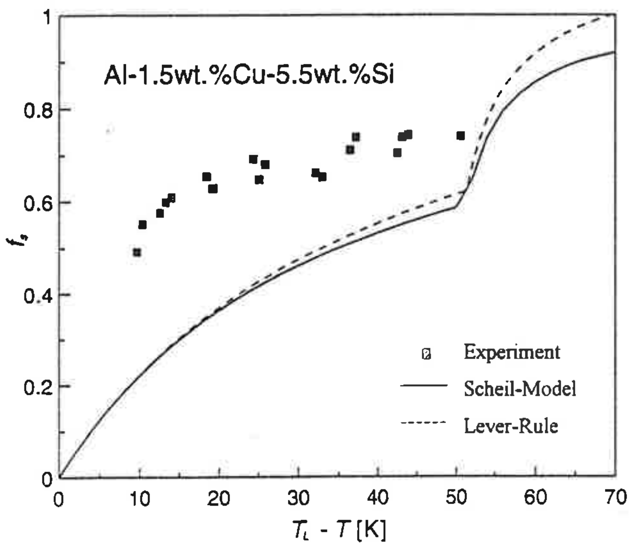
In these equations only the value of an effective distribution coefficient, k, has to be known. According to Kurz and Fisher [3], this effective k can be approximated as $k \approx v_c/v_t$, where v_c and v_t are the critical velocities for the transition from planar to cellular morphology and from cellular to dendritic, respectively. For the superalloy SRR 99, v_c and v_t have been experimentally determined as 0.046 and 0.12 mm/min [4], which results in $k \approx 0.38$. The value of T_L and ΔT_0 were measured as 1357 and 27 °C, respectively [5]. Using these values, the above equations were applied for the f_S - $(T_L\text{-}T)$ relation of the superalloy SRR 99.

4 RESULTS AND DISCUSSION

In **Figure 2a-c** the experimentally determined solid fraction is compared with the predictions of the Scheil model and the Lever-rule, for the three different alloys. At the beginning of solidification a rapid growth of the primary phase (α-phase for Al-7.8wt.%Si and Al-5.5wt.%Si-1.5wt.%Cu, and γ-phase for SRR 99) led to a quick increase of f_S. Then, due to the interfering of the neighboring dendrites, the increase of f_S slowed down. Finally it is terminated with an eutectic precipitation, whose volume fraction is about 70, 55 and 3 percentage for the three investigated alloys at the above mentioned growth conditions. Note that the inderdendritic growth of eutectic in binary alloys has to be isothermal. In multicomponental alloys, however, a temperature interval has to be passed through to complete the eutectic reaction.



(a)



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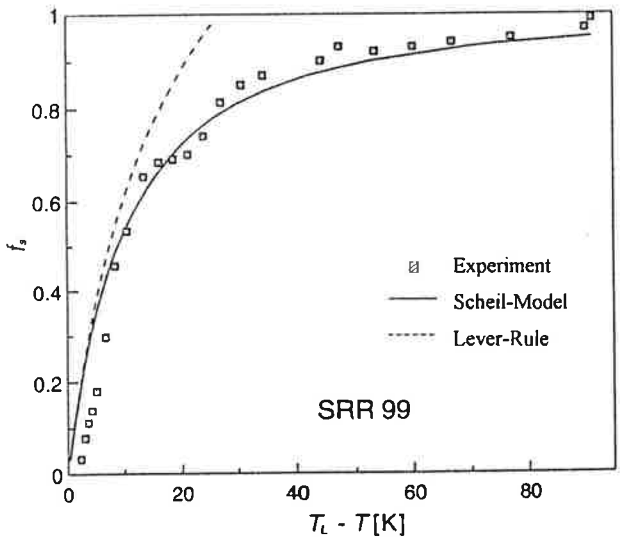

(c)

Figure 2: Experimentally measured evolution of fraction solid f_s as function of temperature, in comparison with theoretical prediction:
(a) Al-7.8wt.%Si
(b) Al-5.5wt.%Si-1.5wt.%Cu
(c) superalloy SRR 99

The thermodynamic calculations for the ternary alloy presented in **Figure 2b** reveal that at about f_s = 0.55 the binary (α+Si)-eutectic should start to form, resulting in a steeper increase of f_s. Considering the Scheil model, the solidification is terminated at the ternary eutectic point at about f_s = 0.97 and T_L-T = 110 °C (which is not shown in the diagram). The eutectic fraction in the superalloy SRR 99 is so small that it can be considered to be completed at a definite temperature, approximately.

For the alloys Al-7.8wt.%Si and Al-5.5wt.%Si-1.5wt.%Cu, which revealed a large (binary) eutectic fraction, the two considered theoretical models do not agree with the experimental results. The increase of the solid fraction is much more pronounced, even than predicted by Lever-rule. However, due to the typically small solid-state diffusion of these substitutional alloys, their solidification behavior should be better described by the Scheil model, which is even less the case. Theoretical models which take a limited back-diffusion into account [6, 7] predict a solid fraction curve between that of the Scheil model and the Lever-rule. Thus the deviation between experimental results and theoretical prediction can not be explained by the diffusion in the solid. A similar result is obtained by Kurz and Grugel [8] for Al-6wt.%Si.

In **Figure 2c** the experimentally determined solid fraction curve for the superalloy is compared with the Scheil model and the Lever-rule. Although this alloy is very complex and a simplified calculation procedure is used, the experimental points coincide well with the prediction of the Scheil model. In this alloy the eutectic precipitation is only three percent. Thus the mushy zone developed nearly completely before the eutectic reaction occurred. Considering the tip undercooling corresponding to the model of Bower et al. [9], the agreement between experimental points and theory in the low solid fraction region can be further improved.

5 SUMMARY

Although the real shape of the phase diagram was taken into consideration, the experimentally determined solid fraction curves of two technical Al-alloys differed immense from both, Scheil model and Lever-rule. Comparison with the good agreement between the experiments and Scheil´s model for a superalloy revealed, that the deviation from the theory is correlated with the amount of (binary) interdendritic eutectic. Ongoing studies will compare our experimental results with a recent model, with takes coarsening and forward diffusion into account [10].

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