

## Distribution and prediction of solute in Al–Zn–Mg alloys

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Received in revised form 9 August 2005; accepted 10 August 2005

### Abstract

The distribution of solute in  $\alpha$ -Al matrix of directionally solidified Al–5.3 at.% (12 wt.%) Zn master alloy with additions of 5.5–11.5 at.% (4.6–9.2 wt.%) Mg was determined and predicted according to the model for dendrite solidification of multicomponent alloys with unequal liquid diffusion coefficients. Predictions showed a good agreement with experimental data, especially for Al–5.3 at.% Zn master alloy with Mg contents from 5.5 to 6.5 at.%. Furthermore, solute concentration data was used to predict the maximum amount of  $\tau$  phase precipitate in  $\alpha$ -Al matrix which will impact positively in the electrochemical efficiency properties of Al–Zn–Mg alloys which will be employed for cathodic protection applications. © 2005 Elsevier B.V. All rights reserved.

**Keywords:** Aluminium alloys; Solidification; Distribution of solute; Predictions; Sacrificial anode

### 1. Introduction

The as-cast microstructure of most solidified alloys consisted mainly of dendrites with eutectic between dendrite arms. To understand its behavior several dendrite growth models have been developed based on the steady state solution of the solidification problem involving a paraboloidal solid/liquid interface. The diffusion field ahead this interface has been given by the Ivantsov solution [1] and used to describe the growth of dendrites. Kurz et al. [2] based on the Ivantsov solution for the transport problem [3] and the marginal stability criterion [4], modeled the problem of constrained cellular or dendritic growth in the velocity range approaching that for absolute morphological stability. In addition, Gäumann and co-workers [5] considered the growth at the marginal stability and used the Ivantsov's model to determine the composition profile in the liquid ahead of the dendrite interface.

The modeling of solidification of ternary systems has been performed assuming independent diffusion of the solutes, such that the diffusion fields in ternary alloys are then given by similar mathematical functions as in binary systems and the boundary conditions at the solid/liquid interface given by the phase diagram. This approach has been applied to derive a dendrite

growth model using the Ivantsov solution, the marginal stability criterion and independent solute diffusion [6]. Furthermore, by assuming no thermal gradient at the scale of the grain, negligible thermal undercooling, low growth rate, growth at the marginal stability limit, independent solute fields and neglect off-diagonal diffusion terms, it was developed a growth kinetic model for multicomponent dendrite tip [7]. Under those assumptions, the growth of the dendrite tip is governed by the supersaturation associated with the difference between the liquid concentration at the tip and far from the tip, normalized by the solute rejected by the tip. When the dendrite is growing at the marginal stability limit, the dendrite tip radius, dendrite tip concentration and dendrite tip undercooling for a multicomponent system are obtained.

This work presents results of solute distribution (Zn + Mg) in  $\alpha$ -Al dendrites of Al–Zn–Mg alloys and the results compared with predictions of the model of dendrite solidification of multicomponent alloys, and then, experimental results and predictions used to propose an appropriated alloy composition which can be used to produce Al alloys for cathodic protection applications of structures exposed to marine environments against corrosion.

### 2. Prediction of solute distribution

In order to derive the distribution of solute Zn and Mg during dendrite solidification of Al–Zn–Mg alloys, it was employed the

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model for dendrite solidification of multicomponent alloys with unequal liquid diffusion coefficients developed by Rappaz and Boettinger [7], which is an extension of the model previously developed for columnar dendrite growth of ternary alloys [8]. During solidification of the alloy, a dendrite is developed and its tip is governed by the supersaturation:

$$\Omega_j = \frac{(C_{L,j}^* - C_{0,j})}{(C_{L,j}^* - C_{s,j}^*)} = \left[ \frac{(C_{L,j}^* - C_{0,j})}{C_{L,j}^*(1 - k_j)} \right] = Iv(Pe_j) \quad (1)$$

where  $C_L^*$  is liquidus concentration at the solid/liquid interface,  $C_0$  initial alloy concentration,  $C_s^*$  solidus concentration at the solid/liquid interface (in at. % or wt. %),  $Iv(Pe)$  is Ivantsov number =  $Pe \exp(Pe)E_1(Pe)$ , where  $E_1(Pe)$ , the first exponential integral;  $Pe$  Péclet number =  $VR/2D_L$ ,  $V$  solidification growth velocity (m/s),  $R$  the dendrite tip radius (m) and  $D_L$  is the solute liquidus diffusion coefficient ( $m^2/s$ ).

Assuming no thermal gradient at the scale of a grain, negligible thermal undercooling, low growth rate, growth at the marginal stability, independent solute fields given by the Ivantsov solution and neglect off-diagonal diffusion terms, the radius is expressed as:

$$R = 2\pi \left( \frac{\Gamma}{\sum_{j=1}^n m_j G_{c,j}} \right)^{1/2} \quad (2)$$

where  $\Gamma$  is capillarity constant (K m),  $m$  liquidus slope,  $G_c$  concentration gradient (wt. %/m or at. %/m) and  $\pi = 3.1416$ .

At the tip:

$$\begin{aligned} G_{c,j} &= - \left( \frac{V}{D_L} \right) (C_{L,j}^* - C_{s,j}^*) \\ &= - \left( \frac{V}{D_L} \right) \left[ \frac{C_{0,j}(1 - k_j)}{1 - (1 - k_j)Iv(Pe_j)} \right] \end{aligned} \quad (3)$$

where the partition coefficient  $k = (C_s/C_L)$ .

By combining the equation corresponding to the dendrite tip radius and solute gradients at the tip, it is obtained the dendrite tip radius as a function of the Péclet number and from this, the solute concentration at the tip in the liquid as:

$$C_{L,j}^* = \frac{C_{0,j}}{1 - (1 - k_j)Iv(Pe_j)} \quad \text{for } j = 1, n \quad (4)$$

and as pointed out by Rappaz and Boettinger [7], if the undercooling of the alloy is small, all the parameters of the phase diagram can be estimated at the liquidus temperature of the alloy.

### 3. Experimental

Al–Zn–Mg alloys were obtained after placing Al, Zn and Mg elements of commercial purity (99.5%) into a high alumina crucible and melted under a vacuum induction furnace with a constant flux of argon and cast into an experimental arrangement as that shown in Fig. 1. Alloy composition of alloys was obtained by plasma spectroscopy and shown in Table 1.

Microstructure was revealed after grinding the specimens in emery paper wet with methanol instead of water to avoid corro-

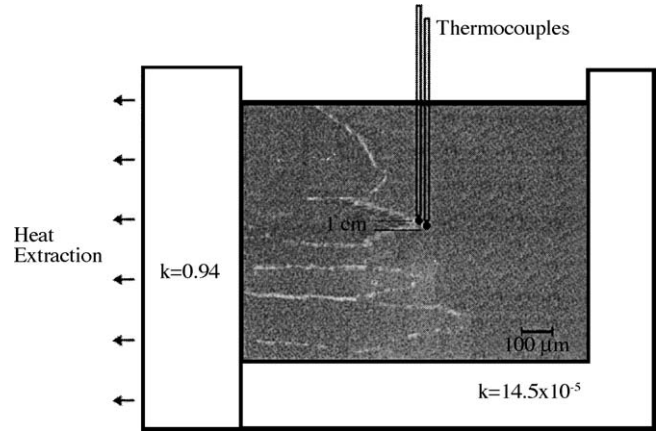


Fig. 1. Experimental arrangement used during solidification of Al-alloys ( $k$  in  $cal/(cm^2 C s)$ ).

Table 1  
Average alloy composition of samples

Elements	Basic			
	Anode 1	Anode 2	Anode 3	Anode 4
Mg	4.6 wt.%, 5.5 at. %	5.4 wt.%, 6.5 at. %	6.2 wt.%, 7.5 at. %	9.2 wt.%, 11.5 at. %
Zn	12.0 wt. %, 5.3 at. %			
Al	Balance			
	Traces			
Si	0.041–0.212 wt. %, 0.0426–0.2206 at. %			
Cu	0.22–0.92 wt. %, 0.10–0.50 at. %			
Fe	0.1 wt. %, 0.104 at. %			

sion of the specimens and electroetched in a solution containing 10%  $HClO_4$  in ethanol and observed under a Stereoscan 440 scanning electron microscope. WDS microanalyses were performed on primary and secondary dendrite arms to determine the distribution of Zn and Mg elements. Microanalyses results were compared with predictions of solute distribution of Zn and Mg in  $\alpha$ -Al solid solution according to the model of dendrite solidification of multicomponent alloys.

The electrochemical behavior of Al-alloys was investigated in 3% NaCl solution. The electrochemical tests were carried out in a three-electrode cell arrangement. The samples of the Al-anode were put in a sample holder presenting an exposing area of  $125 \text{ mm}^2$  to the electrolyte. A platinum gauge was used as a counter electrode and a saturated calomel electrode was employed as a reference electrode.

### 4. Results and discussion

The microstructure obtained after solidification of Al–Zn–Mg alloys is shown in Fig. 2 and consisted mainly of  $\alpha$ -Al dendrites with small precipitates ( $<2.0 \pm 0.3 \mu\text{m}$ , shown by an arrow) of the  $\tau$  phase on it and eutectic in interdendritic regions. As the Mg content increases, the volume percent of  $\tau$  phase in  $\alpha$ -Al matrix and the eutectic in interdendritic regions

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