Crystallization model of magnesium primary phase in the AZ91/SiC composite

J. Lelito [a], P.L. Zak [a], A.L. Greer [b], J.S. Suchy [a], W.K. Krajewski [a], B. Gracz [a], M. Szucki [a], A.A. Shirzadi [b,c,*]

[a] Department of Foundry Processes Engineering, AGH University of Science and Technology, Faculty of Foundry Engineering, Reymonta 23, 30-059 Krakow, Poland
[b] Department of Materials Science and Metallurgy, University of Cambridge, Pembroke Street, Cambridge CB2 3QZ, UK
[c] Materials Engineering Group (DDEM), The Open University, Walton Hall, Milton Keynes MK7 6AA, UK

ARTICLE INFO

ABSTRACT

Metal–matrix composites are widely applied in the aerospace and automotive industries because of their low density and high rigidity. We focus on composites of magnesium-alloy (AZ91) matrix reinforced with SiC particles, with the aim of building a micro–macro model for their solidification. Heat-flow processes are modelled on the macro-scale, with latent heat release during nucleation and grain growth. Calculations are for a fixed cooling rate, consistent with the micromodel for magnesium primary phase solidification. The population density of grains in the matrix is assumed to depend on the maximum supercooling and on the mass fraction of SiC particles. The present model gives reasonable agreement with measurements.

© 2012 Elsevier Ltd. All rights reserved.

1. Introduction

Metal–matrix composites (MMCs) can be made 'ex situ' by addition of reinforcing particles to a liquid or semi-solid alloy, or 'in situ' by forming the particles during the solidification of the matrix itself. The present work is concerned with 'ex situ' composites consisting of the Mg-based AZ91 alloy (9 Al, 0.6 Zn, 0.2 Mn, 0.03 Si, 0.002 Fe, 0.003 Cu and 0.001 Ni (all wt.%) [1]) reinforced with particles of silicon carbide. These composites solidify with equiaxed dendrites of a Mg-based primary phase and a non-equilibrium eutectic [2]. The eutectic is ignored in our treatment, as the primary phase has the predominant effect on the mechanical properties of the composite. Previous work has shown that the presence of reinforcement particles [3] or fibres [4] can affect the final microstructure of metal–matrix composites because of the effects of the reinforcing phase on the local solidification conditions.

Comprehensive treatments of solidification include macromodelling of fluid flow and heat transfer, and micromodelling of grain nucleation and growth [5]. The literature offers many examples of the application of so-called micro–macro models [6–10]. The most uncertain part of such models is often the nucleation law, which it is convenient to describe in terms of the nucleation rate, i.e. the rate at which the population density of grains increases as the supercooling is increased. In his experiments on cast iron, Oldfield [11] noted that the population of grains was proportional to the square of the supercooling, corresponding to the nucleation rate increasing linearly with supercooling. More recent modelling usually takes the nucleation rate to have a Gaussian form as a function of supercooling, often with superposed Gaussians for different types of nucleation [12].

In such approaches, it is implicit that the number of nucleated grains depends on the maximum supercooling reached and that if the melt were held at a constant supercooling there would be no ongoing nucleation as a function of time. The free-growth model [13] offers a possible explanation of such behaviour. The model is based on there being a size distribution of nucleant particles. Each particle can become an active nucleation centre when a critical supercooling is reached that is inversely proportional to the diameter of the particle.

The model presented in this article is modification of the kinetic-diffusion model of Liu and Elliott [7]. It builds on earlier work on the solidification of AZ91/SiC composites [1,14–17]. The aim is to develop a method for predicting the mean grain size in the matrix of these composites from the mass fraction of SiC particles and the maximum supercooling as input parameters.

2. Mathematical model

A numerical micro–macro model of primary-phase solidification in AZ91/SiC composites must treat diffusion of heat and solute. The primary phase is α-Mg solid solution, approximated here as the binary alloy Mg–9 wt.%Al. The temperature field in the model

* Corresponding author at: Materials Engineering Group (DDEM), The Open University, Walton Hall, Milton Keynes MK7 6AA, UK. Tel.: +44 1908 655799; fax: +44 1908 653858.
E-mail address: a.shirzadi@open.ac.uk (A.A. Shirzadi).

can be described with the Fourier equation taking into account the heat of crystallization of the α-Mg phase:

$$\frac{\partial T}{\partial \tau} = a \nabla^2 T + \frac{q_v}{c_v}, \tag{1}$$

where a is the thermal diffusivity ($m^2\,s^{-1}$), c_V the heat capacity ($J\,m^{-3}K^{-1}$), T the temperature (K), and τ the time (s). The rate of release (per unit volume) of the heat of crystallization (latent heat) is given by:

$$q_v = L \frac{\partial f_S}{\partial \tau}, \tag{2}$$

where L is the latent heat ($J\,m^{-3}$), and $\partial f_S / \partial \tau$ is the rate of increase of solid fraction (s^{-1}). The solid fraction of primary phase, ignoring impingement, is given by:

$$f_S = \sum_{i=1}^{n+1} \frac{4\pi}{3} R_i^3 N_i, \tag{3}$$

where R_i is the radius of a spherical grain of the solid (m), N_i is the volumetric grain density (m^{-3}), and the index i denotes the class of grains that appears during the ith time step since beginning of nucleation. The rate of increase of solid (α-Mg) phase fraction in the $(n+1)$th time step is given by:

$$\frac{\partial f_S}{\partial \tau} = \sum_{i=1}^{n} 4\pi R_i^2 N_i \frac{\partial R_i}{\partial \tau} + \frac{4\pi}{3} R_{n+1}^3 \frac{\partial N_i}{\partial \tau}. \tag{4}$$

The population densities N_i of grains in Eq. (4) are evaluated from an expression of Fraś et al. [9,10] that relates the overall grain population density N_V to the maximum supercooling. This is based on (i) an exponential size distribution of nucleant particles (as also measured by Greer et al. [13]), and (ii) an inverse relationship between the diameter of a nucleant particle and the supercooling necessary for nucleation to occur on it [13].

It is expected that N_V, the overall population density of grains, must also depend on the mass fraction of added SiC particles. A complete expression has been derived for N_V in the matrix phase of AZ91/SiC composites [1,16,17]. The solution of Eq. (4) requires knowledge of N_V and of the local growth rate of the primary-phase grains.

The growth rate is controlled by the supercooling at the solid–liquid interface and by solute diffusion in the liquid. Two stages of solidification are considered. The first ends when the maximum supercooling is reached and from this point no more grains are nucleated. In the second stage, the growth rate of the already nucleated primary-phase grains depends on the supercooling and is controlled by solute (Al) diffusion in the liquid. Diffusion of the base metal (Mg) is not considered in this model. The diffusion of Al in the liquid around a spherical grain of Mg-based primary phase is shown schematically in Fig. 1. The solute concentrations shown in Fig. 1 are C_c in the grain centre, C_a, C_L on the solid and liquid sides of the moving interface, respectively, and C_0 in the liquid away from the computational domain.

The rates of change of aluminium concentration C_α, C_L are, for primary-phase solid:

$$\frac{\partial C_\alpha}{\partial \tau} = D_\alpha \left(\frac{\partial^2 C_\alpha}{\partial r^2} + \frac{2}{r} \frac{\partial C_\alpha}{\partial r} \right) + \frac{r}{R} \frac{\partial C_\alpha}{\partial r} \frac{\partial R}{\partial \tau}, \tag{5}$$

and for liquid:

$$\frac{\partial C_L}{\partial \tau} = D_L \left(\frac{\partial^2 C_L}{\partial r^2} + \frac{2}{r} \frac{\partial C_L}{\partial r} \right) + \frac{R_0 - r}{R_0 - R} \frac{\partial C_L}{\partial r} \frac{\partial R}{\partial \tau}, \tag{6}$$

where D_α, D_L are the diffusion coefficients ($m^2\,s^{-1}$) of aluminium in the Mg-Al primary-phase solid and in the liquid, r is the distance between the centre of the α-Mg grain and the node of the numerical
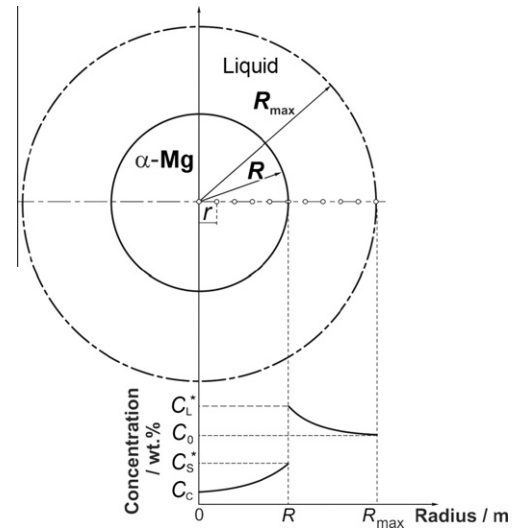

Fig. 1. Aluminium concentration around a spherical grain of primary phase growing into the liquid alloy, treated as binary Mg–9Al (symbols are defined in the text).

grid, and R_0 is the maximum radius, dependent on the population density of grains in the primary phase:

$$R_0 = \sqrt[3]{\frac{3(1 - f_S)V}{4\pi}}. \tag{7}$$

Here, V is the standard analysed volume. The parameter R_0 allows grain impingement to be taken into account. The mass balance at the solid–liquid interface is described by:

$$\left(C_L^* - C_\alpha^* \right) \frac{\partial R}{\partial \tau} = D_\alpha \frac{\partial C_\alpha}{\partial r} \bigg|_{r \to R^-} - D_L \frac{\partial C_L}{\partial r} \bigg|_{r \to R^+}, \tag{8}$$

where C_L^*, C_α^* are equilibrium aluminium concentrations in the liquid and solid (wt.%).

The boundary conditions are as follows:

– at the crystal–liquid interface:

$C = C_\alpha^*$, $r = R^-$, on the grain side,
$C = C_L^*$, $r = R^+$, on the liquid side,

– at the centre of a grain:

$$\frac{\partial C_\alpha}{\partial r} \bigg|_{r=0} = 0, \tag{9}$$

and in the liquid away from the crystal–liquid interface:

$$\frac{\partial C_L}{\partial r} \bigg|_{r=R_0} = 0. \tag{10}$$

To determine the velocity of the solid–liquid interface, Eqs. (5) and (6) are coupled with mass balance, Eq. (8). A similar model has been presented in Ref. [10]. Numerical solution of Eqs. (1)–(8) allows prediction of N_V, the population density of grains in the matrix phase in the MMC, as a function of the mass fraction of SiC particles.

3. Modelling results

Calculations have been performed for the composite AZ91/SiC, with the assumption that the particles of the reinforcing phase SiC are uniformly distributed in the liquid. This assumption facilitates calculation of the effects of the mass fraction and size of the SiC particles on the heterogeneous nucleation of primary