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Experimental determination of grain growth kinetics during eutectic solidification

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ABSTRACT

In this work a method to estimate grain growth parameters during equiaxed eutectic solidification is presented. The method assumes free grain growth before impingement and an exponential dependence of the grain growth rate on undercooling. In order to validate the method, a heat transfer / solidification kinetics (HT/ SK) model was implemented. Once validated, the method was used to estimate experimentally the kinetic parameters of equiaxed growth of a near eutectic Al-Si alloy not modified an modified with Sr. Results shows that eutectic grain growth parameters of eutectic Al-Si change as a consequence of eutectic modification with Sr.

KEYWORDS

Solidification;
Kinetics;
Growth;
Eutectic;
Al-Si;
Modeling.

INTRODUCTION

Control of solidification microstructures is vital for the development and improvement of the quality and properties of the final cast products. Prediction of the formation and evolution of the cast microstructure is intimately linked to understand the solidification kinetics (SK). For such a purpose, different kinds of models have been developed in the last couple of decades. In the works of Liu et al.[1] and Nakajima et al.[2] are discussed recent methodological progresses in modeling of solidification and casting.

The models used in computer simulations of solidification involve calculations on nucleation and growth kinetics which in turn depend on the availability of nucleation and growth laws capable to reproduce the experimental behavior. Therefore, there is a necessity to develop new experimental methods to obtain data allowing the simulation of SK for specific alloys.

Fourier thermal analysis method (FTA) has been used to study SK of different systems of metallurgical interest like commercial alloys[3] and metal matrix composites[4]. This method has been described in detail by Fras et al.[5]. FTA gives only SK information on the evolution of solid fraction, fs, and solidification rate dfs/dt. In order to obtain more detailed information about kinetics of eutectic equiaxed growth, some efforts have been reported on the development and validation of methodologies allowing grain growth characterization during solidification of undercooled melts. The following exponential equation has been used to describe equiaxed grain growth as a function of the undercooling (ΔT):

$$(\frac{dR}{dt}) = \mu \Delta T^n \qquad (1)$$

In eq.(1) R is the grain radius, t is time, μ and n are the growth parameters and $\Delta T = T_E - T$, defines undercooling, where T_E is the eutectic temperature and T is the instantaneous temperature during solidification. Degand et al.[6] performed experiments with Al-Si eutectic alloy. They found that if the value of the exponential coefficient n is kept constant at 2 in eq. (1), according to the classical theory of eutectic growth, the presence of Sr decreases the value of the pre exponential growth coefficient of modified eutectic.

Diozegi and Svensson[7] have proposed a method to obtain the grain coefficients μ and n from solid fraction evolution and grain density data using the Kolmogorov, Johnson, Mehl and Avrami grain growth model (KJMA). They validated their method using volume grain density and simulated cooling curves, generated by a HT/SK model for eutectic gray iron. However there are not available reports in the open literature on the applications of their method to experimental eutectic alloys.

It has been mentioned that Al-Si eutectic modification with Sr occurs as a consequence of changes in nucleation and growth mechanism of eutectic silicon[8]. Therefore it could be interesting to test any methodology intended to obtain grain growth parameters with Al-Si eutectic alloy, not modified and Sr modified in order to detect possible changes in the eutectic growth coefficients.

Accordingly, the purpose of this work was the development and validation of a method for determination of eutectic grain growth kinetic coefficients and the application of the method to the case of a near eutectic Al-Si alloy, not modified and modified with Sr.

MATHEMATICAL MODELING

Free growth model and method for determining growth coefficients

If it is assumed that Eq. (1) describes the dependence of the grain growth rate on undercooling a plot of log (dR/dt) versus log (ΔT) would give values of n and μ. The growth rate dR/dt can be obtained using a grain growth model linking instantaneous solid fraction and grain radius and known data on solid fraction evolution and grain density. In this way, an equivalent grain radius can be obtained every time step, Δt, recorded in the cooling curve.

It is assumed that during solidification of the sample there are N spherical grains of mean radius R developing freely at the same time. N is the number of grains per unit volume. The solid fraction in the early stages of growth at a time t is given by the following equation where Fs^t and R^t are the solid fraction and radius at time t, respectively:

$$Fs^t = \frac{4}{3}\pi N (R^t)^3 \qquad (2)$$

The density of grains per unit volume in the sample, N, can be determined by metallographic methods and Fs can be obtained from FTA. Thus, it is possible to estimate the instantaneous grain radius, R^t. The evolution of grain radius as a function of time can be obtained from eq.(3).

$$\frac{dR}{dt} = \frac{R^{t+dt} - R^t}{\Delta t} \qquad (3)$$

The instantaneous undercooling during solidification, ΔT, can be obtained from the temperature registered in the cooling curve.

Heat transfer and solidification kinetics (HT/SK) model and inverse heat transfer modeling

The HT/SK model simulates the cooling and solidification of a cylindrical body of metal, thermally isolated at its top and bottom, in order to obtain thermal histories of eutectic Al-Si alloy under cooling conditions close to the experimental conditions present in this work. The model assumes that, at the beginning of the cooling process, the entire metal domain is at the same initial temperature. It is also assumed the description of all heat transfer resistances at the external boundary by a combined heat transfer coefficient, obtained from experimental cooling curves and the use of inverse heat transfer modeling to simulate the cooling of the liquid metal into a metallic mould. The cooling of the melt and the heat flow to its surroundings is simulated assuming that heat transfer within the metal is governed by conductive heat transfer and latent heat generation due to solidification, constant thermo physical properties and unidirectional radial heat flow.

The energy balance applied to the metal system under study described above can be written as:

$$\mathbf{Cp^{v}\frac{\partial T(r,t)}{\partial t}=k^{th}\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial T(r,t)}{\partial r})+L^{v}{}_{f}\frac{\partial fs(r,t)}{\partial t}} \qquad (4)$$

where Cp^v and k^{th} are respectively the volumetric heat capacity and the thermal conductivity of the metal, T (r, t) is the temperature, r is the radial position, t is the time and L_f^v is the volumetric heat of fusion

The boundary conditions are:

$$\mathbf{r=0 \qquad \frac{\partial T}{\partial r}=0} \qquad (5)$$

$$\mathbf{r=r_{max} \quad -k^{th}\frac{\partial T}{\partial r}=h_{eff}A(T-T_{\infty})} \qquad (6)$$

In eq.(6) h_{eff} is the combined heat transfer coefficient, which simulates the heat extraction from the outer surface of the metal, located at $r = r_{MAX}$. To solve eq.(4), fs (r, t), the solid fraction as a function of time and position within the casting must be known. The SK model described in a previous work on Al-Si alloys[9] is coupled to the heat transfer calculations.

The eutectic grains are assumed to be spherical in shape and the rate of growth of eutectic grains is calculated using eq. (1) with $\mu = 5x10^{-6}$ m S^{-1} $^oC^{-2}$ and n = 2. Latent heat of eutectic Al-Si was obtained from FTA applied to the experimental melts. The other thermo physical properties of eutectic Al-Si alloy used during calculations can be found in[9].

Eq. (4) was solved numerically by discretizing the cylindrical metal system in the form of a finite difference mesh composed by a known number of cylindrical volume elements (VE) and using the explicit finite difference method. In order to fulfill the stability needs, time step for calculations was set at 0.0025 s.

For inverse heat transfer modeling, the model previously described and the basic ideas mentioned by Santos et al.[10] were used to obtain the global or combined heat transfer coefficient, h_{eff}, as a function of time or temperature. For instance, the semi empirical heat transfer coefficient used during calculation of the cooling of the probe during the stage of cooling of the liquid probe is given by the following equation where T_{int} is the instantaneous temperature at the outer volume element of the metal domain:

$$\mathbf{h_{eff}=51+4.8x10^{-5}*\exp(-\frac{T_{int}}{-54})} \qquad (7)$$

EXPERIMENTAL PROCEDURE

Melts were produced in an electric furnace with an argon atmosphere to obtain a near eutectic Al-Si alloy using burdens of commercial purity silicon and A356 alloy. Chemical composition was adjusted using spark emission spectroscopy. TABLE 1 shows chemical composition of the experimental base and Sr modified melt.

The effect of the presence of eutectic modification on the grain growth kinetic parameters was explored by triplicate for the same Sr modified, (Sr addition) and non-modified (no Sr addition) near eutectic Al-Si alloy. The addition of strontium was accomplished using Al-10%Sr master alloy. Thermal analysis test samples were taken by submerging a cylindrical stainless steel test cup (0.03 m inner diameter, 0.05m in height and 1.5 mm in thickness, covered with boron nitride) into the melt. The cups were kept submerged for approximately 30 seconds to allow them to reach the bath temperature. Then, they were removed from the melt and placed on a thermal analysis test stand where they were isolated thermally at the top and at the bottom. In order to record the thermal history of the alloy during its cooling and solidification, two thermocouples, type K, with alumina sheath, 0.0015 m OD, were introduced in the liquid sample at the same depth at two different radial positions. Thermocouples output was recorded in a personal computer connected to a NI FieldPoint cFP 1804 data acquisition system. A calibration procedure was performed with 99.9 % aluminum. The experimental cooling curves were numerically processed using FTA method in order to obtain information about the evolution of solid fraction during solidification of the sample.

TABLE 1 : Chemical composition of experimental alloys

	Si	Fe	Cu	Mn	Mg	Zn	Sr	Al
Not Modified	12.6	0.45	0.02	0.1	0.25	0.02	0.0001	Bal
Sr Modified	12.5	0.42	0.02	0.1	0.24	0.02	0.0196	Bal

For microstructural analysis, the samples were sectioned transversally and prepared by standard polishing procedures. The microstructure of the specimens was observed using optical microscopy. In order to reveal the eutectic grain boundaries and determine grain density, a combination of the methods of thermal and chemical etching proposed by Degand et al.[6] was used in order to achieve good grain boundary resolution. Polished, instrumented samples are held in a furnace during 10 minutes at a temperature of about 560°C, and then quenched in water. A second step consists in etching the surface of the samples with Poulton's etchant (60%HCl, 30% HNO_3, 5%HF, 5% H_2O) during 30-60 seconds.

RESULTS AND DISCUSSION

In order to validate the grain growth characterization method proposed in this work, the HT/SK model described previously was used to simulate the cooling and solidification of a cylindrical casting of eutectic Al-Si alloy under cooling conditions similar to the experimental conditions present during experimentation with a heat transfer coefficient obtained from experimental cooling curves using inverse heat transfer modeling.

The radius of the casting was r = 0.015 m and two cooling curves were obtained for volume elements located at two different radial positions (r1= 0 and r2 = 0.005m). The cooling curves were simulated by the model and numerically processed using FTA method. The volume grain density predicted by the model was 5.2 x 10^8 grains/m^3.

Figure 1 shows the cooling curves predicted by the model and the solid fraction evolution obtained from FTA numerical processing of these curves and the parts of the cooling curve and the solid fraction curve that are used for grain growth kinetic calculations. The cooling curve data used to calculate the thermal undercooling includes the section of the curve corresponding to the point of maximum undercooling and a point near to the point of maximum recalescence, shown at times t1 and t2 in Figure 1. During this time interval, using the known number of grains per unit volume and the solid fraction evolution as a function of time predicted by FTA, it is possible to calculate the evolution of the grain radius.

Figure 2 shows the plot of log (dR/dt) versus log (ΔT) generated by the method, where it can be seen a good fit to a linear equation, with an intercept of -5.31, i.e a pre exponential growth coefficient of μ = 4.9 x 10^{-6}, and an exponential coefficient of 2.0 which suggests, when compared to the growth coefficients used by the model (μ = 5 x 10^{-6} and n = 2), that this methodology could be used to obtain the growth coefficients for equiaxed eutectic grain development.

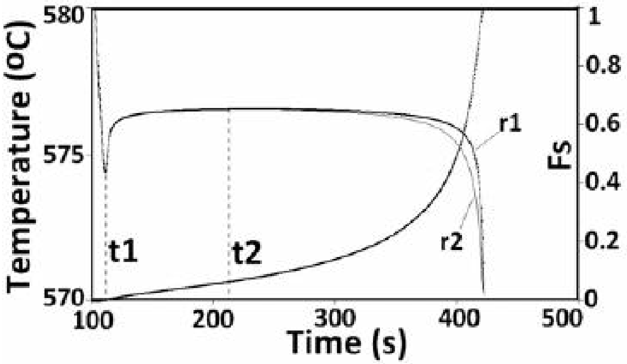

Figure 1 : Cooling curves predicted by the HT/SK eutectic Al-Si model and FTA solid fraction evolution and description of the time interval used to perform grain growth characterization

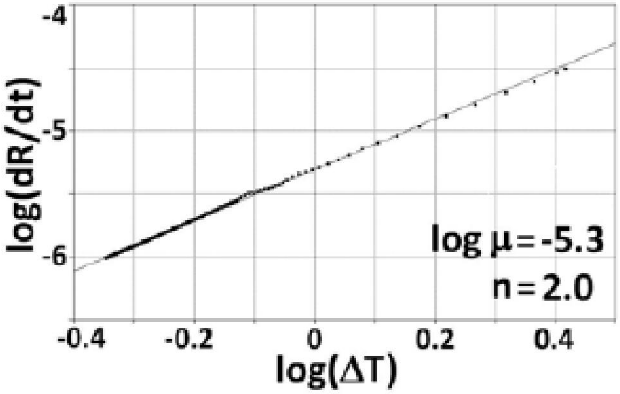

Figure 2 : Logarithmic plot of grain growth rate against undercooling obtained from the method proposed in this work

Accordingly the method was applied to experimental information related to the estimation of the growth coefficients for the near eutectic Al-Si alloy under study, non-modified and modified with Sr. Experimental cooling curves were numerically processed using FTA to obtain the solid fraction evolution. Figure 3 shows typical macroetched, not modified, Figure 3(a), and Sr modified samples, Figure 3(b). The volume grain density determined for not modified and Sr modified samples was roughly 7 x10^8 and 1 x 10^7 grains/m^3 respectively.

Figure 4 shows typical plots of log (dR/dt) versus log (ΔT) for an experimental non modified, Figure 4(a) and Sr modified, Figure 4(b) near eutectic

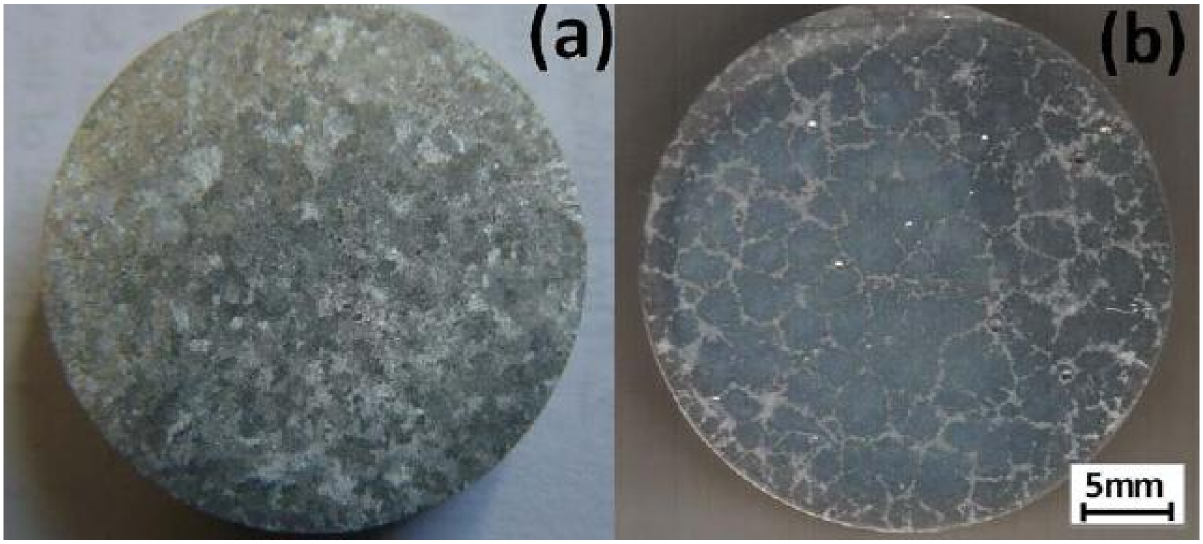

Figure 3 : Typical macroetched samples of (a) not modified and (b) Sr modified experimental alloys

Al-Si alloy.

Pre exponential and exponential coefficients obtained from experimental data are shown in TABLE 2. It can be seen that there is a statistically significant difference between growth coefficients for not modified and Sr modified samples, which suggest that Sr modification has changed the functional relationship between equiaxed grain growth rate and undercooling.

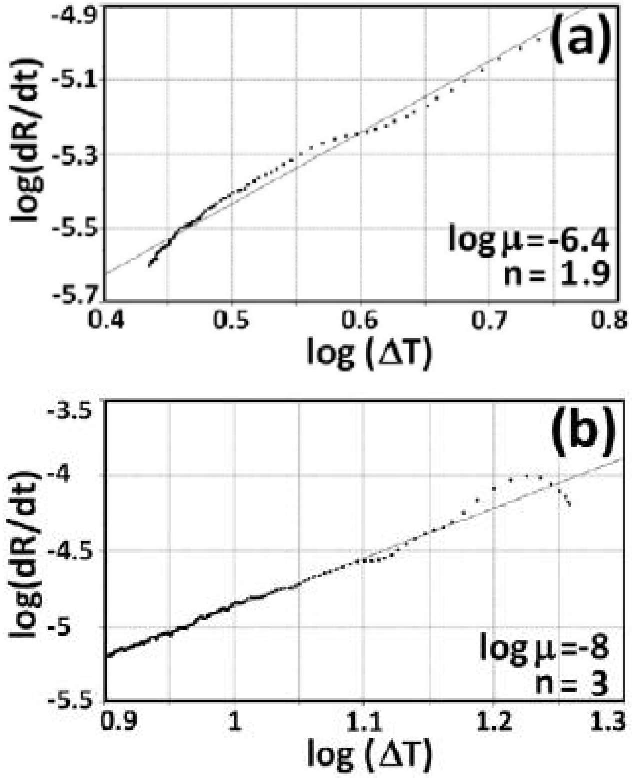

Figure 4 : Logarithmic plot of grain growth rate against undercooling obtained from data for: (a) not modified and (b) Sr modified samples

TABLE 2 : Growth kinetic coefficients obtained for not modified and Sr modified probes

Alloy	Log (μ)	μ	n
Not modified	-6.6 +/-.4	2.5×10^{-7}	2 +/- 0.2
Sr modified	-8 +/- .2	1×10^{-8}	3.1 +/- 0.2

It can be seen that there is some dispersion for the coefficients obtained using this method. In fact this method, as well as the method proposed by Diozegi and Svensson[7], are very sensitive to changes in undercooling and volume grain density data, and these aspects must be evaluated very carefully in order to obtain reproducible results.

In order to explore the validity of the obtained grain growth parameters, these grain growth laws and experimental grain density were introduced into the HT-SK model developed in this work in order to simulate the cooling curves of a unmodified and a Sr modified Al-Si alloys. Figure 5 shows experimental Figure 5(a), and predicted, Figure 5(b), cooling curves for unmodified and Sr modified eutectic, Al-Si alloys. From Figure 5 it is clear that there is a good agreement between experimental and predicted cooling curves for not modified and Sr modified Al-Si eutectic alloy, suggesting that the method developed in this work could be used to obtain kinetic parameters to describe quantitatively grain growth during equiaxed solidification modeling.

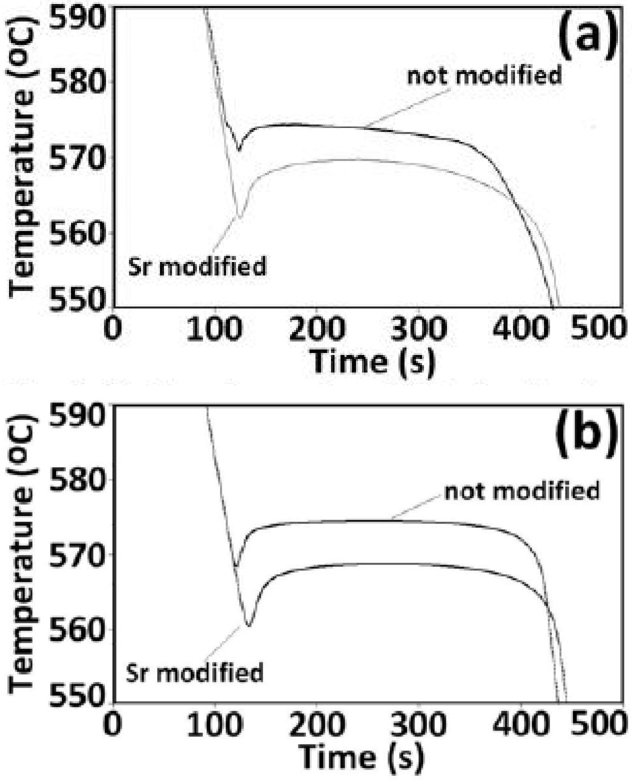

Figure 5 : (a) Experimental and (b) simulated cooling curves of not modified and Sr modified eutectic using nucleation data and grain growth coefficients generated in this work

CONCLUSION

It is proposed a method using a simplified free grain growth model for the determination of eutectic grain growth parameters during eutectic solidification.

The method has been validated using a heat transfer/solidification kinetics model.

The method was used to study a near eutectic Al-Si alloy, with and without Sr additions.

Results suggest that growth parameters of Al-Si eutectic change as a consequence of eutectic modification with Sr.

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