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Mold Filling of Semisolid Metal Slurries

O.J. Ilegbusi and S. Brown

A constitutive model is developed experimentally for Al-7%Si-0.6%Mg alloy slurry. This equation has two parts to represent flow and structure evolution. The model is then used to simulate filling of a chisel-shaped mold of cylindrical cross section. Transport equations are solved for the flow variables, and the free surface is modeled by solving the conservation equation for a scalar "fluid-marker" variable, which is used to update the fluid properties, including density and viscosity.

Keywords
mold filling, semisolid processing, solid particles, liquid matrix

1. Introduction

SEMISOLID PROCESSING involves the production of new materials by exploiting their characteristics in the partially solidified state. It involves partially solidifying a melt or partially melting a solid to produce solid particles in a liquid matrix (Ref 1, 2). The solid fraction typically ranges from about 20% to over 90%. The resulting multiphase system provides opportunities for improvements in both material properties and processing technique (Ref 1, 3).

Among the issues to be addressed in the implementation of semisolid processing are: the rheological behavior of the material; transport phenomena including non-Newtonian flow of the slurry, mold filling, and heat transfer associated with solidification; macrosegregation or fractionation; and the final properties of the fully solidified product. Until the present time, little fundamental work has been done on these issues. Indeed, most of the published research has been concerned with design of equipment for producing the slurries and characterization of the materials produced (Ref 4).

The constitutive behavior of semisolid systems is rather complex. The material exhibits time-dependent thixotropy, where the effective shear resistance decreases as deformation occurs or as the rate of deformation changes. Semisolid slurries also display hysteresis when shear stress is plotted against the shear rate for a cyclic loading cycle.

Flemings and coworkers (Ref 1, 4, 6) pioneered the rheological characterization of melt-solid suspensions. Their work on Sn-Pb alloy was the first to confirm that such slurries exhibit shear thinning behavior, which can be qualitatively represented by a "power-law" constitutive relationship. Other notable efforts in the rheological characterization of melt-solid slurries of metallurgical interest include contributions by Nguyen et al. (Ref 7) and Levaillant (Ref 8).

A number of review articles have surveyed the state of the art of thixotropic flow behavior. Mewis (Ref 9) provides a very complete survey of thixotropic material systems, different models for thixotropy, and experimental methods for characterizing thixotropic behavior. Cheng and Evans (Ref 10) provide a discussion of single internal-variable (or structural model) formulations. An internal-variable model assumes that the time dependence of rheological properties is due to changes in the structure of the system. These changes can in turn be represented by the evolution of a scalar variable.

Experimental work on semisolid rheology is limited. Joly (Ref 5) used hysteresis experiments to characterize the history dependence of slurries. Chijiiwa and Fukuoka (Ref 11) also evaluated apparent viscosities of several metal slurries but paid little attention to the material state. A number of limited experimental results were also presented at the recent conference on semisolid slurry processing (Brown and Flemings, Ref 4).

Very little modeling has been applied to semisolid rheology. Flemings and associated investigators (Ref 6, 12) provided some of the first efforts to characterize the rheology of these systems. While this work has been useful in identifying the fundamental issues, it has not reached the state of completion that would permit formulation of complete constitutive models. Brown (Ref 13) presented a constitutive model based on the "single internal-variable" concept to address thixotropy of semisolid slurries. Specifically, this model assumes that flow resistance is due to hydrodynamic flow of aggregates and deformation of solid particles within the agglomerates and that the degree of agglomeration can be represented by a single scalar (structural) variable. At fully agglomerated state when all particles are well connected together, this variable takes a value of unity; however, in fully disagglomerated state, it is zero. The work presented here is based on this model, and further details are presented in a later section.

While numerous papers have been published in recent years on theoretical studies and mold-filling phenomena, very little work is available on the behavior of non-Newtonian fluids. Notable exceptions are the works of Ilegbusi and Szekely and their coworkers (Ref 14-18). Jonsson et al. (Ref 14) employed a power-law type model to model free surface behavior during mold filling of a non-Newtonian fluid. Ilegbusi and Szekely (Ref 15-17) studied fluid flow, heat transfer, and solidification in both mechanically and electromagnetically stirred slurry systems. In a recent study, Ilegbusi and Szekely (Ref 18) also applied the time-dependent constitutive model of Brown (Ref 19) to the rheocasting of a Sn-15% Pb alloy into which B_4C particles are suspended under assumed isothermal conditions.

The purpose of this paper is to develop further this constitutive model and apply it to the filling of a mold of varying cross section (specifically, a chisel-shaped mold) with Al-7%-0.6%Mg alloy slurry. A series of experiments is performed to establish the exact form and coefficients of the constitutive

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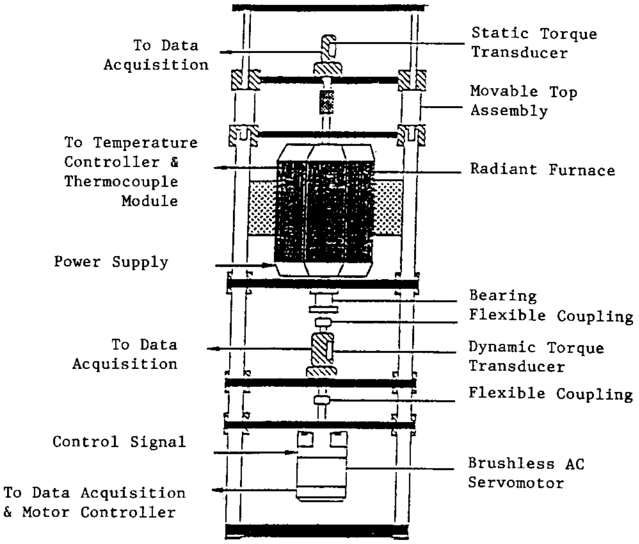

Fig. 1 Schematic of high-temperature rheometer (Kumar, et al., Ref 20)

model. This model is then employed in the numerical scheme to simulate the mold-filling process.

2. Experimental Program

Details of the experimental technique used to determine the rheology of the Al-7%-0.6% alloy slurry were described in several publications by Brown and coworkers (Ref 19, 20) and are not repeated here. The technique involves the use of a high-temperature couette viscometer, which is shown in Fig. 1. A set of rate-change experiments are devised in which each set of rate change from the same set provides a measure of the flow behavior at that state. Each experiment provides a data point for a shear stress/shear rate response of a given structural state. The set of data obtained are analyzed, and a correlation is developed for the shear stress/shear rate relationship. These results are presented in section 3.

3. Mathematical Formulation

3.1 *Governing Equations*

Modeling of mold filling requires a coupling between fluid flow and heat transfer equations. Assuming the slurry is incompressible, these equations are as follows.

3.1.1 Mass Conservation

$$\frac{\partial u_i}{\partial x_i} = 0 \qquad \text{(Eq 1)}$$

3.1.2 Momentum Conservation

$$\rho \frac{\partial}{\partial t}(u_i) + \rho \frac{\partial}{\partial x_i}(u_i u_j) = -\frac{\partial P}{\partial x_i}$$

$$+ \frac{\partial}{\partial x_j}\left[\mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)\right] - F_{\mathrm{d}} + F_{\mathrm{b}} \quad i, j = 1, 2, 3 \qquad \text{(Eq 2)}$$

in which μ is the viscosity, and F_{d} is a drag source per unit volume, used to represent a mushy region. F_{d} is expressed in terms of the drag coefficient C_{d} as:

$$F_{\mathrm{d}} = C_{\mathrm{d}}\mathbf{u} \qquad \text{(Eq 3)}$$

in which C_{d} is varied linearly with solid fraction. F_{b} in Eq 2 is the buoyancy force, which in the present instance is represented by the Boussinesq approximation (Ref 21). Specifically, the density is assumed to be constant in all equations except in the buoyancy term, wherein it is represented by the expression:

$$\rho = \rho_{\mathrm{o}}\,[1 - \beta(T - T_{\mathrm{o}})] \qquad \text{(Eq 4)}$$

where ρ_{o} and β are the density and coefficient of expansion, respectively, at reference temperature, T_{o}. The viscosity of the slurry is calculated from an internal variable constitutive relationship established in the experimental program and described in a subsequent section.

3.1.3 Energy Conservation

We employ the enthalpy method so that the following transport equation is solved for the specific enthalpy h:

$$\rho \frac{\partial h}{\partial t} + \rho \frac{\partial}{\partial x_i}(u_i h) = \frac{\partial}{\partial x_j}\left[\kappa\left(\frac{\partial h}{\partial x_j}\right)\right] + S_h \qquad \text{(Eq 5)}$$

in which the enthalpy source, S_h, takes the form (Ilegbusi and Szekely, Ref 17):

$$S_h = \rho \frac{\partial \Delta H}{\partial t} - \nabla \cdot (\Delta H \rho \mathbf{u}) \qquad \text{(Eq 6)}$$

In Eq 5 and 6, κ is the thermal conductivity, and ΔH is the latent heat. Details of this method were presented by Ilegbusi and Szekely (Ref 15). Note that the enthalpy method is generally more numerically stable than the direct calculation of temperature for systems with sharp discontinuities of the thermophysical properties (such as specific heat). It therefore precludes the need for interpolation of these properties at the interfaces.

The solid fraction, f_{s}, is calculated from the correlation based on the experimental results of Joly (Ref 5) on solidification of Sn-15%Pb alloy. This correlation is expressed as:

$$T(\mathrm{K}) = -45.933 f_{\mathrm{s}}^2 - 7.22 f_{\mathrm{s}} + 481.76 \qquad \text{(Eq 7)}$$

Table 1 Principal input parameters used in the computation

Maximum die diameter	2.5 cm
Minimum die diameter	1.6 cm
mold height	15 cm
Ram speed	25 cm/s
Ram diameter	7.3 cm
Slurry inlet temperature	200 °C
Die initial temperature	20 °C
Fluid viscosity	2.5×10^{-2} Pa·s

Table 1 presents the principal input parameters employed in the computation.

3.2 *Free Surface*

Mold-filling process involves free surfaces or the interaction of two or more distinctly different media (i.e., the semislurry and air) separated by sharply defined interfaces. The position of this interface is not known a priori. Thus, the mathematical model must be able to address the discontinuities that exist in flow quantities, satisfy the above field equations governing conservation of mass, momentum, and energy, and be consistent with the boundary conditions. In this study, we employed a modification of the scalar-equation method (SEM) embodies in the PHOENICS computer code (Rosten and Spalding, Ref 22).

In SEM, the above transport equations are sill valid, but the density and viscosity fields, which are dependent on the local fluid type, are required for the solution of these equations. The transient convection of the scalar φ satisfies the equation:

$$\frac{\partial \varphi}{\partial t} + \frac{\partial}{\partial x_i}(\varphi u_i) = 0 \qquad \text{(Eq 8)}$$

such that;

$$\varphi = 0 \text{ in fluid 1 (air)}$$

$$\varphi = 1 \text{ in fluid 2 (slurry)} \qquad \text{(Eq 9)}$$

In situations with such a large difference in fluid properties across an interface, numerical diffusion is a problem. This problem is reduced by assuming that the fluid properties (ρ and μ) vary linearly from those of fluid 1 to those of fluid 2 at the interface. The gradient of the property interface can be varied by limiting the range of φ within which properties can vary. It is well known in computational fluid dynamics (CFD) that the numerical discretization of the scalar variable transport equation, Eq 8, creates unphysical smearing of the discontinuity at the gas/slurry interface. This mechanism is called false numerical diffusion and may produce unstable and inaccurate results. The solution for φ is here rendered numerically stable by using an explicit-finite-difference scheme as described in Jun and Spalding (Ref 23).

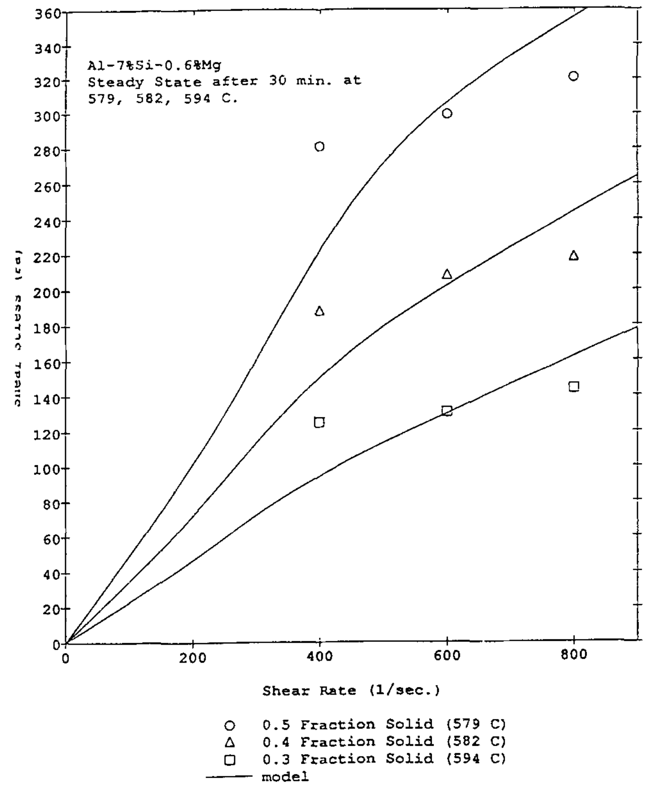

Fig. 2 Shear response at steady state

3.3 *Constitutive Behavior*

We employ a modified version of the model based on the internal (structural) variable constitutive relationship proposed by Brown (Ref 13). This model assumes that flow resistance is due to the hydrodynamic flow of agglomerates and deformation of solid particles within the agglomerates. Based on experimental information, Brown (Ref 13) assumed that the single structural variable, s, varies between 0 and 1. $s = 0$ represents a fully separated structure, and $s = 1$ represents a condition of fully agglomerated structure. Further details of this model are found in Ref 13, 19, and 20.

In general, the flow equation can be written as:

$$\tau = \tau_d + \tau_a \qquad \text{(Eq 10)}$$

where τ_d characterizes hydrodynamic dissipation of energy in the slurry system, and τ_a represents the disruption of pair bonds (or degree of agglomeration). Both terms are related to the shear rate and the internal variable, s, which is governed by an evolution equation. The exact forms of the resulting model equations and coefficients are presented in section 4.1 on Experimental Results.

3.4 *Estimation of Heat Transfer Coefficient*

The heat transfer coefficient is obtained by considering an overall macroscopic heat balance, assuming there are no steep temperature gradients at both sides of the mold/slurry interface or that the temperature jump due to the contact resistance is

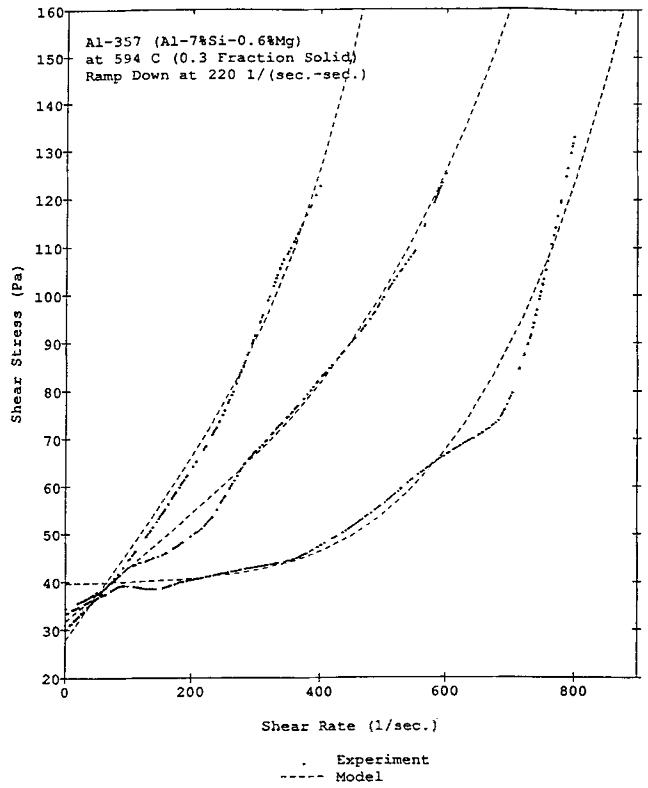

Fig. 3 Shear response at 594 °C

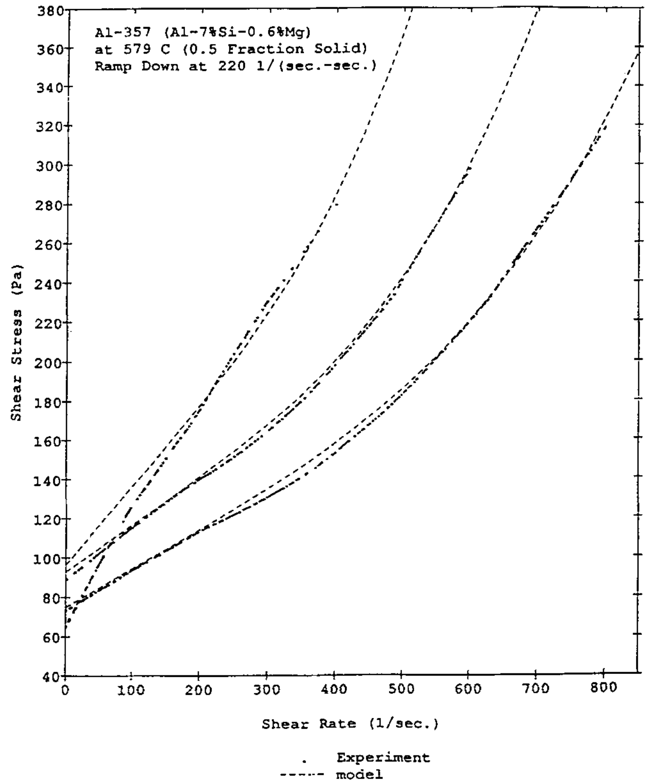

Fig. 4 Shear response at 579 °C

large compared with the gradients in that vicinity. Note that this approach can only be used to obtain an approximate heat transfer coefficient when limited experimental data exist. In the present situation, we are interested in a parametric study of mold filling, and the approximate value will suffice. The average heat flux transferred into the mold is thus estimated by integrating over time the instantaneous rate of heat transfer between neighbor points for which temperature histories are available from measurements provided by ALUMAX. This overall heat transfer rate is balanced with the convective heat transfer rate, which is estimated using average temperature differences between the closest measurement points at both sides of the interface. The estimated heat transfer coefficient is 2600 W/m^2·K.

3.5 *Boundary Conditions*

At the inlet plane to the die, the slurry velocity is calculated from the ram or punch speed. Uniform slurry temperature of 200 °C is also prescribed. At the mold walls, the heat flux is calculated from the estimated heat transfer coefficient. A no-slip condition is imposed on the velocity field. The free surface is assumed to be stress free and adiabatic. The latter conditions are used as first approximations and can be relaxed in a subsequent investigation. However, the assumptions are not expected to significantly affect the principal objective of the study, which is the examination of history-dependent constitutive model on mold filling.

4. Results and Discussion

4.1 *Experimental Results*

Figures 2 to 5 show the measured shear stresses, τ, as a function of shear rates, γ, under varying experimental conditions. These conditions include steady-state and shear-rate ramp down. These data are correlated by the flow equation:

$$\tau = \tau_y + A\,(s)\,\frac{(c/c_{\max})^{1/3}}{1-(c/c_{\max})^{1/3}}\,\mu_f\gamma + (n+1)C(T)\,sf_s\mu_f^{n+1}\,\gamma^n \qquad \text{(Eq 11)}$$

in which τ_y is mean yield stress, and μ_f is fluid viscosity. A is a hydrodynamic coefficient with approximate value being the scaling factor based on simple cubic spatial arrangement (=9/8 from Frankel and Acrivos, Ref 24). $c = f_s(1+0.1\,s)$ is the effective volume packing fraction solid. $c_{\max}$ is the maximum effective volume packing fraction solid. n is an exponent. f_s is fraction solid. μ_f is liquid viscosity. $C(T) = C_o \exp(nQ_v/\mathrm{RT})$. $C_o = 8.9 \times 10^{-15}$Pa -ns^{-1}. $n = 4$. R, universal gas constant, is 8.314 J/mol·K. Q_v, activation energy, is 78.25 kJ/mol.

The term s in Eq 11 is the internal variable used to characterize the structure evolution governed by the equation:

$$\frac{ds}{dt} = H(T,f_s)(1-s) - G(T,f_s)s\gamma^n \qquad \text{(Eq 12)}$$

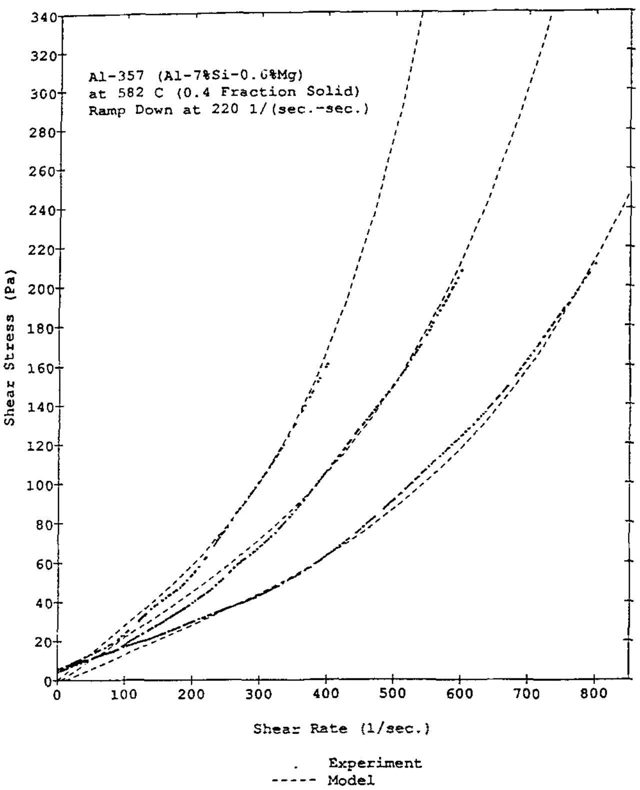

Fig. 5 Shear response at 582 °C

in which H is the agglomeration function, and G is the disagglomeration (or agglomerate-breaking) function. Table 2 presents the experimentally determined values of the above variables at different solid fractions (or temperatures).

Figure 2 shows that at steady state, the response of the slurry is pseudoplastic; i.e., the shear stress increases with shear rate at a decreasing rate. This result is consistent with the conclusion reached previously by other investigators (Ref 5, 25). Figures 3 to 5 on the other hand, show that the slurry behavior under transient conditions is dilatant (the shear stress/shear rate curve has an increasing slope). Figures 2 to 5 show that while the above model adequately reproduces the rheology of the slurry over the range of conditions considered, the transient correlations are better. This observation may be attributed to the assumption inherent in the steady state experiment. Production of semisolid slurries typically occurs over a relatively short time interval (measured in seconds) while steady state data is typically collected over a period of hours. Thus, ramp (transient) experiments are more representative of slurry behavior. Notwithstanding, this model can now be used with confidence in the numerical modeling of the mold-filling process.

4.2 *Mold-Filling Calculation*

Figure 6 shows the plot of the velocity vectors at three different instances as the slurry advances into the chisel-shaped die. Note that this plot does not include any movement of the air above the advancing slurry. The progressive development of the boundary layer near the wall is clearly evident especially at the later stage despite the relatively rapid filling rate. Also, the flow pattern is apparently dominated by the forcing action of the ram, rather than the natural convection at the walls.

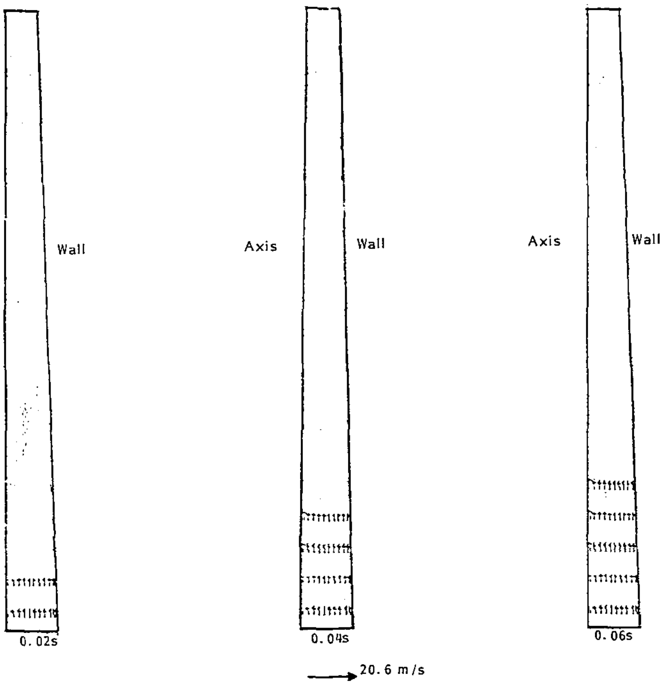

Fig. 6 Velocity vectors after 0.02 s, 0.04 s, and 0.06 s (h = 2600 W/m^2·K)

Figure 7 shows the corresponding plot of the temperature distribution. As expected, the hot region in the vicinity of the incoming stream is quite evident, as is the low-temperature region near the walls. Significant temperature stratification occurs near the walls and above the slurry front due to the high thermal gradients in these regions. The low-temperature isotherm appears to extend farther into the mold at the central axis. Note that the thermal stratification (rather than the profile) above the slurry is a result of our assumption that the air is originally at the temperature of the die (20°C). From a practical standpoint, the near-wall distribution is more significant. Examination of Eq 7 shows that solidification will occur at about 459 K, which implies that a solid shell will form near the walls. Figure 7 shows that this solid shell increases in size as the slurry advances further into the die.

Figure 8 shows the advance of the slurry corresponding to Fig. 6 and 7. The jetting effect at the central region is due to the temperature distribution observed in Fig. 7. This result clearly indicates significant effect of the temperature on the rheology or flowability of the slurry.

Further calculation with the same flow rate and heat transfer coefficient was found to be impossible due to rapid buildup of solid and subsequent choking of the die. Thus, to fully fill the die, the heat transfer rate was reduced by an order of magnitude to 260 W/m^2·K to reduce the intensity of heat transfer at the wall, and the filling rate was increased by 50%. Figures 9 and 10 show the plots of the slurry advance under these conditions. The jetting effect progressively increases as the slurry advances farther into the narrow section of the die. In other words, the free-surface distortion appears to be greater towards the end

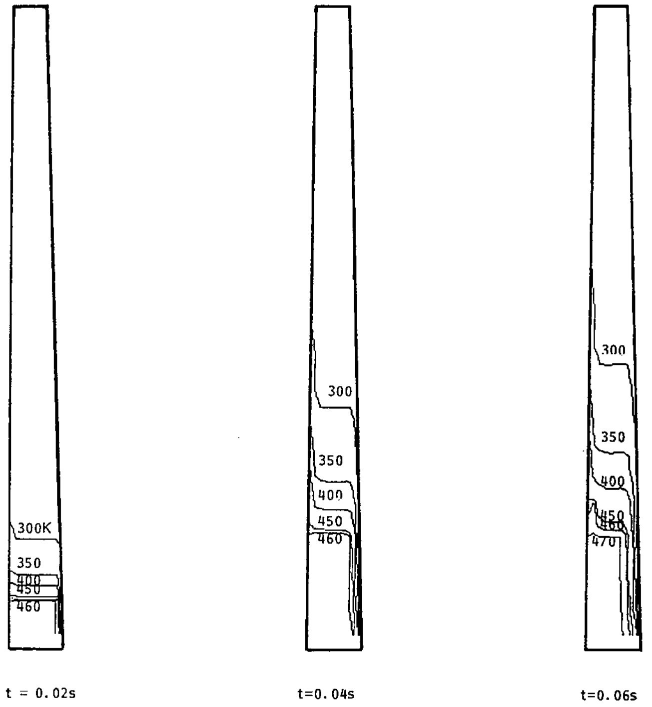

Fig. 7 Temperature distribution after 0.02 s, 0.04 s, and 0.06 s (h = 2600 W/m^2·K)

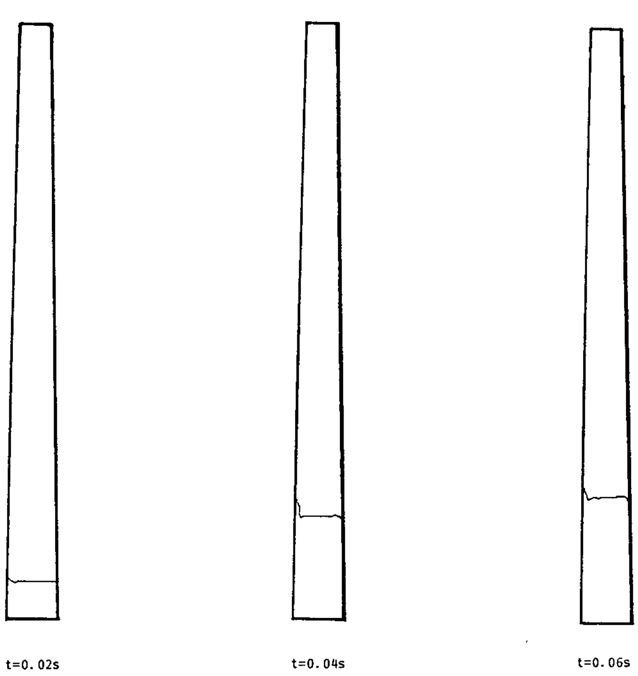

Fig. 8 Advance of slurry after 0.02 s, 0.04 s, and 0.06 s (h = 2600 W/m^2·K)

Table 2 Experimental results

f_s	Temperature,°C	C, Pa^{-n}s^{-1}	G, s^{-1}	H, s$^{-(n+1)}$	τ_y, Pa	A
0.5	579	138,000	1.25×10^{-12}	2.5×10^{-2}	65.0	12.0
0.4	582	120,000	4.18×10^{-13}	5.23×10^{-3}	0	20.0
0.3	594	64,000	2.8×10^{-13}	3.4×10^{-3}	30.0	27.0

of the die. This finding is in direct contrast to the often assumed behavior for mold filling of pure metals. Indeed, one of the reasons for employing bottom pouring in practice is that it is generally thought that free-surface disturbances are reduced under such conditions. Therefore, for semisolid slurries, the allowance for history and temperature effect in the rheology combined with the possible formation of solid shells can produce significant distortion of the free surface, especially at narrow sections. In addition, the observed behavior may be related to the geometry and the possible fractionation of the slurry at the narrow end of the die.

5. Concluding Remarks

A constitutive model based on the single, internal-variable concept, was developed to represent the rheology of Al-7%Si-0.6%Mg alloy slurry. This model was employed in a numerical scheme to simulate the filling of a chisel-shaped die under a range of operating conditions.

The results show that due to the thermal gradient near the wall, the slurry advances into the die as a jet. This jetting effect becomes enhanced as the slurry advances further into the narrow section of the die. This behavior may be related either to fractionation at such sections or the effect of temperature on the rheology of the slurry. This result has significant practical implication in the mold filling of semisolid slurries. It clearly indicates that special care should be taken to incorporate the effect of temperature on the rheology to avoid fractionation of the slurry or choking of the die prior to complete filling. Such an effort, which is the subject of our next study, would represent a major advance in our understanding of semisolid slurry processing.

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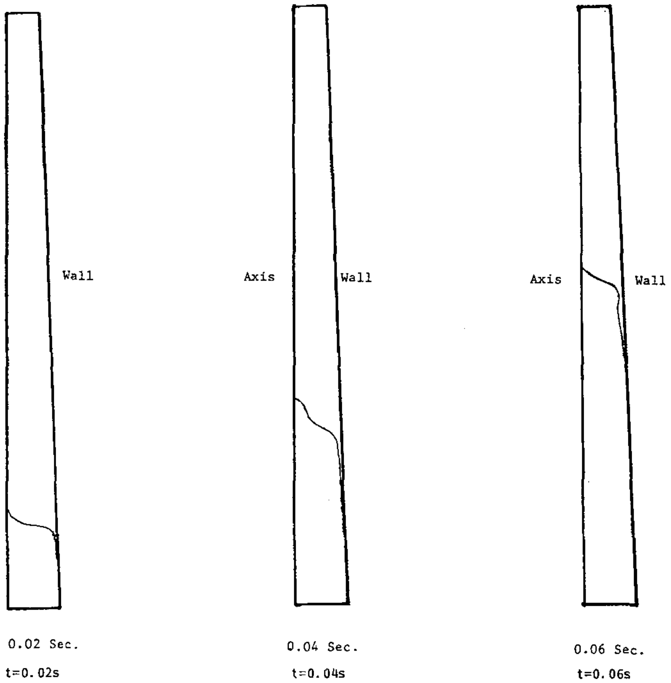

Fig. 9 Advance of slurry after 0.02 s, 0.04 s, and 0.06 s (h = 260 W/m^2·K)

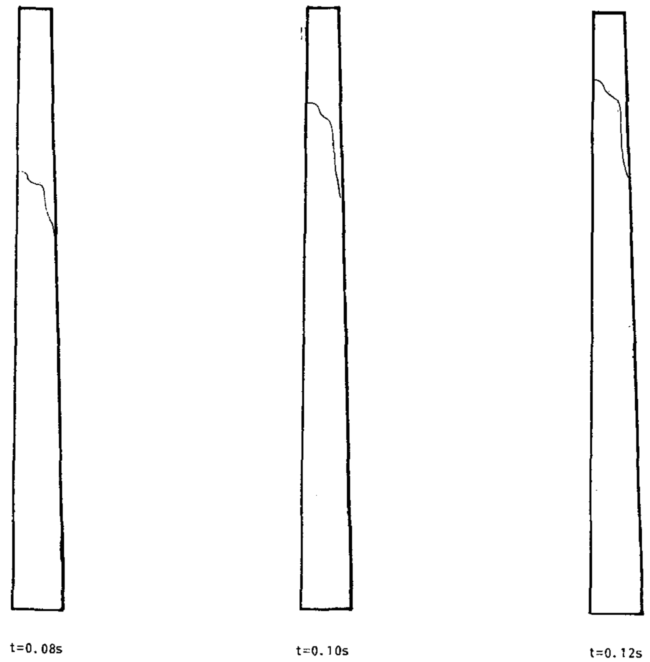

Fig. 10 Advance of slurry after 0.08 s, 0.10 s, and 0.12 s (h = 260 W/m^2·K)

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