Comparison Studies of the Flow Characteristics of the Newtonian and Thixotropic Fluids

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Comparison studies of the effects that the viscosity and the injection speed of the fluids have on the die filling behaviors have been performed via an approach that involves both experiments and simulations. Two different fluids, i.e., thixotropic fluid (paint) and Newtonian fluid (water), were selected as the model fluids to monitor the differences in the rheological behaviors and the associated flow patterns during die filling. According to high-speed photography recorded for the two fluids, in comparison to the Newtonian fluid, the thixotropic fluid flow with higher viscosity proceeded into the die cavity in a more controllable manner while maintaining its free surface. Such experimental observations were then compared with the analytical results obtained from computer simulations. [doi:10.2320/matertrans.M2013157]

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1. Introduction

Since the discovery that semi-solid metals also exhibit thixotropy and pseudoplasticity to an extent similar to those of polymeric materials,¹ extensive studies have been carried out to apply these rheological properties of metal slurries for producing high-integrity structural parts via a technique known as “semi-solid forming”. Metal slurries used for semi-solid forming normally contain approximately 65% of spherical solid particles surrounded by the liquid eutectic phase. At this state, the metal slurry exhibits the viscosity high enough to maintain its own shape during free standing. However, when the external force in the form of shear is applied to the semi-solid slurry, a sudden drop in the viscosity takes place in the slurry, making injection forming possible in a conventional die casting machine.

One of the major concerns in the conventional die casting process is to maintain the flow of the molten metal in a laminar manner, so that the molten metal fills a die cavity in a way to avoid any casting defects (e.g., pores) associated with the turbulent flow. In general, the flow characteristics of a fluid during die filling can be judged by the value of Reynolds number (Re)² as expressed in eq. (1). Experiments showed that a fluid flowing through a circular channel maintains a laminar flow if the value is less than 2100, and otherwise, the flow characteristics tend to become turbulent.

\[ Re = \frac{\rho v D}{\eta} \]  

where \( \rho \), \( v \) and \( \eta \) are the density, velocity and viscosity of the fluid, respectively and \( D \) is the characteristic length such as the gate thickness.

Semi-solid forming, which utilizes highly viscous metal slurries as the raw material, has been regarded as the processing technique that can control the fluid flow in a laminar manner. The viscosity of the semi-solid metals depends on the shear rate, microstructures, cooling rate and solid fraction. Studies on how these variables influence the rheological properties of various semi-solid metals have been conducted extensively in the past.³⁻¹² Studies of the effects that the viscosity has on the flow characteristics of the thixotropic fluid have also been reported.¹³⁻¹⁵ However, for the later issue, much remains to be uncovered and analyzed in detail. In this study, we demonstrated the different flow behaviors caused by the different rheological properties of the two fluids, i.e., the thixotropic and Newtonian fluids. The water (Newtonian fluid) and the paint (thixotropic fluid), both having the rheological properties similar to that of the molten Al alloy and the semi-solid Al alloy, respectively, were selected as the model fluids. The rheological behaviors and the associated flow characteristics of the fluids were analyzed via the experiments and the computational technique.

2. Experimental Procedures

2.1 Measurement of rheological behaviors

Rheological behaviors of the fluids used in this study were measured using the Searle-type viscometer equipped with a stirrer (outer diameter of 54 mm) and a crucible (inner diameter of 60 mm),¹⁶ which allows the Couette flow and thus, makes the measurement of the pseudoplastic and thixotropic behaviors of the semi-solid slurry feasible. Before measuring the viscosity, the viscometer was calibrated using the standard silicon oils, so that the measurements do not exceed ±2% error range. In order to select an appropriate model fluid exhibiting rheological properties similar to those of the typical Al alloys used for conventional die casting (in this study, Al–7Si, commonly known as A357 Al alloy), the rheological behaviors of the A357 Al alloy were measured both during continuous cooling and under isothermal holding.

In the continuous cooling experiments, the molten A357 alloy was sheared at a predetermined shear rate during cooling with the cooling rate of 5 K/min. The torque, temperature of the alloy, and stirring speed measured during the experiments were converted into viscosity, solid fraction, shear stress and shear rate using the relations reported elsewhere.¹⁶ Isothermal holding experiments were conducted...
to measure the pseudoplastic and the thixotropic behaviors of the semi-solid A357 alloy. After melting the alloy at 915 K, it was cooled down to 860 K at a cooling rate of 1 K/min to get a desired solid fraction ($F_s$, in this study, $\sim 0.65$). During the cooling stage, the alloy was continuously sheared at a constant initial shear rate of 75 s$^{-1}$ to promote the globalization of the primary particles. When the temperature of the slurry reaches the predetermined temperature (860 K) or solid fraction ($F_s = 0.65$), the slurry was kept at the same temperature without stirring to allow the agglomeration of globalized particles. After holding the slurry at 860 K for a sufficient time (in this study, 10 min), the slurry was then stirred by increasing the shear rate from 0 to 1045 s$^{-1}$ within a given period of time (here termed the “up-time”) followed by decreasing the rate to 0 s$^{-1}$ within the same period of time (here termed the “down-time”). The measured data, i.e., torque and stirring speed, were converted into viscosity, shear stress and shear rate to evaluate the rheological behaviors of the fluids.

In order to study the effect of the viscosity on the flow behaviors, two different simulator fluids, i.e., thixotropic fluid (paint) and Newtonian fluid (water), were selected as the model fluids. The water ($\rho = 1.0$ g/cm$^3$, $\eta = 0.001$ Pa·s) was chosen as the model fluid to simulate the molten A357 Al alloy ($\rho = 2.5$ g/cm$^3$, $\eta = 0.001$ Pa·s). The paint ($\rho = 1.34$ g/cm$^3$, $\eta = 0.2$ Pa·s) was selected as the thixotropic fluid, because it exhibits rheological behaviors similar to those of the semi-solid A357 Al alloy ($\rho = 2.5$ g/cm$^3$, $\eta \approx 1$–20 Pa·s depending on shear rate, cooling rate and solid fraction). The thixotropic behaviors of the paint were also measured using the same procedures as described above and later used as an input data for the simulation.

### 2.2 Visualization of the fluid flow

Figure 1 shows the mold geometry used to visualize the flow behaviors during die filling. Two circular obstacles with a diameter of 40 mm were placed in the mold cavity. A gate having a rectangular cross section ($2 \times 20$ mm) was positioned at the middle of the bottom wall. A transparent window glass was placed on the mold to record the real-time flow behaviors during filling the mold cavity. The mold was then mounted on a die casting machine having a shot sleeve with a diameter of 35 mm. The model fluids were poured into the shot sleeve and injected into the mold cavity at a predetermined plunger speed. This plunger speed was converted to the gate speed by multiplying the ratio of the cross sectional area between the sleeve and the gate with the plunger speed. Flow patterns of the model fluids during die filling were recorded using high-speed photography at the speed of 500–2000 frames per second. The recorded flow patterns were later compared with the simulation results.

### 3. Results

#### 3.1 Rheological behaviors of the A357 Al alloy

During die filling, the molten Al alloy proceeds to the die cavity, during which it is subjected to shear strain at different rates depending on the injection speed. The shear rate imposed on the melt/slurry can be given by eq. (2.1) and promotes the shear thinning of the slurry, making die casting/filling feasible. The viscosity, along with the gate speed, of the melt/slurry is an important parameter that directly affects the flow characteristics during die filling. Before investigating the approximate range of the critical gate speed, under which the flow characteristic maintains a laminar flow, it is important to have a rough estimation of the shear rate exerted to the slurry. When measuring the speed of the incoming fluid, the speed of the fluid is the maximum at the gate along the longitudinal direction (z-direction in Fig. 1) of the die. Assuming that the gate speed ($v_1$) is much larger than the speed measured along the transverse direction, i.e., $v_z$, eq. (2.1) can further be simplified to eq. (2.2).

$$\dot{\gamma} = \frac{1}{2} \left( \frac{\partial v_z}{\partial y} + \frac{\partial v_y}{\partial y} \right)$$  \hspace{1cm} (2.1)

$$\dot{\gamma} \approx \frac{1}{2} \frac{\partial v_z}{\partial y}$$ \hspace{1cm} (2.2)

In the present study, a rough estimation of the shear rate exerted to the slurry was performed at the gate and the middle of the die using eq. (2.2). When the gate speed ($v_1$) is 5 m·s$^{-1}$, the shear rate has the maximum value of $\sim 2500$ s$^{-1}$ by the time when the slurry enters the gate and, then, it decreased to 200–300 s$^{-1}$, before it comes to stop. Meanwhile, in the case of the fluid with an average injection gate velocity of 10 m·s$^{-1}$, the shear deformation rate applied to the slurry varied from about 5000 s$^{-1}$ at the gate to about 400–500 s$^{-1}$ in the die cavity. Therefore, it is reasonable to measure/estimate the viscosity of the semi-solid slurry by imposing it to the shear rate of 2500–5000 s$^{-1}$. In this study, we measured the viscosity of the fluids experimentally by increasing the shear rate from 0 to 1045 s$^{-1}$, beyond which it was estimated by extrapolating the measured values.

Before selecting the model fluid that allows the simulation of the flow behaviors of semi-solid A357 alloy, the rheological behaviors of the A357 alloy were measured. Given the cooling rate, the viscosity of the semi-solid metals depends not only on the solid fraction, but also on the shear rate imposed on the semi-solid metals. Therefore, the rheological properties of the semi-solid A357 alloy were measured during the cooling stage both to select the model fluid and to utilize the properties as the input data for the
simulations. Figure 2 is the changes in the viscosity of the semi-solid A357 alloy measured as a function of the solid fraction and the shear rate. The measured viscosity was nearly zero even at temperatures where partial solidification began to start. However, beyond critical solid fractions, the viscosity increased drastically with the increasing amount of solid. Note that the critical solid fraction varied depending on the alloy composition, shear rate and cooling rate.18) The raw material used in the actual semi-solid forming contains approximately 60–70% of solid particles. At this solid fraction, the metal slurry, when it is at rest, exhibits a viscosity high enough to make free standing. However, once the slurry is subjected to shearing action, the viscosity of the slurry decreases with increasing shear rate, such that the viscosity of the slurry reduced to less than 1 Pa·s when it is sheared at the shear rate of 1045 s⁻¹ (refer to Fig. 2 and will be shown in more detail in Fig. 4).

In order to measure the pseudoplastic and thixotropic behaviors of the semi-solid A357 alloy, the slurry with \( F_s = 0.65 \) was stirred using a stirrer to impose shear strain at various rates; shear strain was applied to the slurry at an increasing rate \( (0 \rightarrow 1045 \text{ s}^{-1}) \) followed by decreasing rate \( (1045 \rightarrow 0 \text{ s}^{-1}) \). During stirring, the up-time, \( T_u \), required to reach the maximum preset shear rate \( (1045 \text{ s}^{-1}) \) was varied to observe its influence on the rheological behaviors of the alloy. Figure 3 is the typical hysteresis loops showing the pseudoplastic nature of the semi-solid A357 alloy with \( F_s = 0.65 \). The area within the loop became larger as the up-time became shorter. Note that the area enclosed by the loop corresponds to the energy absorbed by the unit volume of the slurry that is required to break down the thixotropic structure of the slurry under a given time. Therefore, these areas enclosed by the hysteresis loops can be used to quantify the degree of the pseudoplasticity, which normally is associated with the rearrangement and fracture of the agglomerated solid particles within the slurry, and thus makes the slurry shear-thinned.16,19)

It is noted from Fig. 3 that, upon measuring the pseudoplasticity of the semi-solid A357 alloy, the flow resistance, i.e., shear stress, increased with increasing shear rate, reached the maximum at \( \sim 300–400 \text{ s}^{-1} \) and then gradually decreased afterward. Further decrease in the flow resistance was observed as the shear rate decreased to zero. In order to demonstrate thixotropic nature of the slurry, the shear stress in the \( y \)-axis of Fig. 3 was converted to the viscosity by dividing it with shear strain rate \( (\dot{\gamma}) \) following the Newton’s law of viscosity, \( \tau = \mu \dot{\gamma} \). Figure 4 shows the representative plots that were obtained by converting the \( \tau - \dot{\gamma} \) curves in Fig. 3, showing the changes in the viscosity of the slurry measured according to the shear rate and different up-time. The results show that at the early stage of shearing (i.e., before the shear rate reaches 150 s⁻¹), the viscosity of the slurry tended to decrease abruptly (from 20 PPa·s to less than 1 Pa·s) as the shear rate increased. Beyond the shear rate of 150 s⁻¹, the viscosity gradually decreased and saturated to a value of 0.2–0.7 Pa·s when the shear rate reached to 900 s⁻¹. This viscosity value did not seem to change further even with the increase in the shear rate. It is interesting to note from Fig. 4 that, during the down-time measurements, the slurry still maintains low viscosity, of which characteristics are commonly known as the thixotropy and render die casting in the semi-solid state feasible.
3.2 Die filling characteristics of the thixotropic paint

Now that the thixotropy of the semi-solid A357 alloy was measured, a fluid exhibiting properties similar to those of the semi-solid A357 alloy is to be sought to use it as the simulator fluid for monitoring the die filling characteristics. The thixotropic behaviors of the paint were recorded using the same procedures employed for the semi-solid A357 alloy and the results are shown in Fig. 5. Although the initial viscosity of the paint was much smaller than that of the semi-solid A357 alloy, the general tendency observed from Fig. 5 was very similar to that of the A357 Al alloy as seen from Fig. 4; the viscosity decreased rapidly at the early stage of shearing (i.e., <150 s⁻¹) and then gradually decreased until it saturated to a value of 0.2 Pa·s. During the down-time experiment, the paint still maintained the viscosity at the level of ~0.2 Pa·s. Such rheological properties observed from the paint are considered to be similar to those observed from the A357 alloy in Fig. 4. Therefore, despite the slightly lower viscosity of the paint at the initial state, we consider it a plausible candidate to simulate the flow behavior of the semi-solid A357 slurry.

The flow patterns of the water and the paint were recorded during the die filling using high-speed photography. Figure 6(a) is a series of the snap shots taken from the high-speed motion pictures showing the die filling behavior of the water injected at the gate speed of 5.5 m·s⁻¹. The incoming liquid front was shattered upon colliding with the obstacles located within the cavity even at the relatively low injection speed. This turbulent-type die filling pattern became even more severe as the injection speed increased, such that the water exhibited a completely shattered liquid front when injected at the gate speed of 10 m·s⁻¹ (Fig. 6(b)). Therefore, under such a condition, the air trap is inevitable during filling the die cavity, which in turn can degrade the mechanical properties. This is why conventional die casting, where molten metals are normally injected at the gate speed of 20–100 m·s⁻¹, is not suitable for producing structural parts, where the structural integrity is essential.

Figure 7(a) is a series of the snap shots taken from the high-speed motion pictures showing the die filling characteristics of the thixotropic paint injected at the gate speed of 5 m·s⁻¹. Unlike the flow patterns shown by the water in Fig. 6(a), the paint maintains a laminar-type flow even after impacting the obstacles within the cavity. As the gate speed increased to 9 m·s⁻¹, the free surface of the paint begins to be shattered as can be seen in Fig. 7(b). At this point, we evaluated the Reynolds number corresponding to the flows in Figs. 6 and 7 in order to show the ability of the thixotropic paint in suppressing the tendency to a turbulent flow; the maximum values of the Reynolds number corresponding to the flows in Fig. 6 were calculated as ~2300 and 4100 at the gate for the gate speed of 5 and 9 m·s⁻¹, respectively. In contrast, in the case of the flows shown by the water, the same was calculated to be larger by more than two orders, such that it was 4.6 × 10⁵ and 8.2 × 10⁵ for the gate speed of 5.5 and 10 m·s⁻¹, respectively. Based on the results in Fig. 7, it is assumed that the critical injection gate speed of the model fluid (with the viscosity of ~0.2 Pa·s) used in this study is ~5 m·s⁻¹. Considering that the viscosity of semi-solid slurry is at least 2–3 times thicker than that of the paint used in the present experiment, the fluid flow would be more controllable during actual semi-solid forming of structural parts.
4. Discussions

The flow patterns exhibited by the water and the paint were computed using the commercial Flow-3D software whether the die filling behaviors of the thixotropic fluid can be predicted using the measured thixotropic properties. The water is a Newtonian fluid with its viscosity being independent on the shear rate and was used as the model fluid to describe the flow characteristics of the molten Al alloy. On the other hand, the paint is a typical thixotropic fluid with its viscosity highly shear-dependent and was used as a simulator fluid for predicting the general flow characteristics of the semi-solid A357 alloy with $F_s = 0.65$.

During injection, the paint, initially positioned in the shot sleeve, proceeds slowly toward the gate area. When the paint reaches at the gate area, it is subjected to shear strain such that the shear rate gradually increases until the maximum shear rate is attained. The shear rate is the maximum by the time when the paint passes through the gate. Therefore, the viscosity of the paint would become shear-thinned by the time it escapes the gate, causing an abrupt decrease in the viscosity. Once the paint proceeds into the mold cavity, the shear rate exerted on the paint will decrease due to the reduced velocity. However, as was shown in Fig. 5, the paint would retain the low viscosity even under the reduced shear rate. In order to use the thixotropic properties of the paint as an input data for simulating the die filling behavior, the viscosity measured during the down-time experiment in Fig. 5 were fitted to the Carreau model as given in eq. (3). The solid line in Fig. 8 shows the best-fit curve corresponding to the experimentally measured viscosity.

$$\eta(\dot{\gamma}) = \eta_\infty + \frac{\eta_0 - \eta_\infty}{\left[ a_1 + \left( \frac{1}{2}a_2\dot{\gamma} \right)^n \right]^{2/n}}$$

where $\eta_0$ and $\eta_\infty$ are the viscosity measured at rest ($= 3.5 \text{ Pa}\cdot\text{s}$) and under infinite shear rate ($= 0.0005 \text{ Pa}\cdot\text{s}$), respectively. $a_1$, $a_2$, and $n$ are the constants, which were determined to generate the best-fit curve (solid line) in Fig. 8 and are 1.0, 1.5 and 0.47, respectively.

The flow behavior of the thixotropic paint was simulated using the die geometry as shown in Fig. 1. The numbers of grid lines generated on the die cavity along the $x$-, $y$- and $z$-directions were 30, 7 and 50, respectively. The $k$-$\epsilon$ turbulence model was employed to predict the fluid flow patterns during die filling. Figure 9(a) is a series of snap shots of the die filling pattern captured from the simulated results of the thixotropic paint injected at the gate speed of 5 m/s$^{-1}$. In general, the simulated flow patterns are in good agreement with the experimental results in Fig. 7(a); the simulation revealed the die-filling patterns similar to those observed from the experiments, which showed a smooth free surface even after impacting the obstacles within the cavity. Figure 9(b) shows the flow patterns calculated for the paint injected at the gate speed of 9 m/s$^{-1}$, showing that the free surface begins to be shattered upon impacting the obstacle (No. 1) inside the cavity. The observation in this study indicates that the viscosity and the velocity of the fluid are the major parameters that have a significant influence on the flow characteristics.
5. Conclusions

The flow characteristics of the Newtonian water and the thixotropic paint, the simulator fluids for the A357 alloy in its molten state and the semi-solid state, respectively, were monitored to study the effects that the viscosity and the injection speed of the fluids have on the die filling behaviors. Based on the comparison study of the flow patterns between the water and the paint, the following conclusions could be drawn.

(1) The incoming water was shattered upon impacting the obstacles located within the cavity even at the relatively low injection speed \( (5 \text{ m s}^{-1}) \), while the paint maintains a laminar type flow even after impacting the obstacles under the same gate speed.

(2) The viscosity-shear rate relation used to compute the flow behavior of the thixotropic fluid could be fitted on the Carreau model and used as an input data for simulations. When compared the analytical results with what were observed from the experiments, the analytical results based on the \( k-\varepsilon \) turbulence model agreed well with the experimental results.

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