Primary particle movement and change of property of cast iron by centrifugal effect in semi-solid processing

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Abstract

The particle distribution in semi-solid slurry under centrifugal field was simulated and the main factors such as fraction of solid, rotation speed, and holding time effecting the particle distributions are discussed. The simulation results showed that primary particles rich zone is produced in radially outer area and these results are in good agreement with the experiment. The centrifugal effect produces the primary particles distribution along the radial direction. Denser particles are concentrated in outside than inner side. For high fraction of solid samples, ‘wall’ appear in the middle of samples because of high viscosity region making particle difficult to move. Longer holding time gives denser primary particles concentrated more in outside than inner side. Higher rotation speed gives increased gradient of hardness in the radial direction. It is due to that number of primary austenite at inner side at higher rotation speed is less than that at lower rotation one. At higher rotation speed, ledeburite forms more at inner side of specimen than at the other one. It is also shown that semi-solid processing at lower fraction of solid gives higher hardness because smaller number of primary austenite, namely more ledeburite forms in microstructure. The present study gives useful information on producing material with locally changing property by semi-solid processing.

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1. Introduction

Semi-solid processed cast iron can be produced by using mechanical stirring system. The studies of Qiu et al. \cite{1} have shown that at sufficiently slow cooling and high shear conditions, austenite particles become spheroidal and small graphite particles are dispersed uniformly in the sample. A characteristic cast structure is obtained when the slurry is cooled with stirring and centrifugal force acts on liquid/solid particles having different density. In a centrifugal field, the particles move to the outer or inner side depending on the density difference between particle and liquid \cite{2–4}. Using this principle, it is expected to develop functionally graded material targeting cast iron.

In the present paper studies are made on the austenite particle distribution in a centrifugal field during solidification by computer simulation and the influence of operational factors on the characteristics of cast iron structure by semi-solid processing.

2. Experiments

2.1. Materials preparation

Fig. 1 shows a schematic of experimental apparatus for preparation of semi-solid slurry of cast iron. The sample in the magnesia crucible is located within a furnace that can be rapidly heated up to the temperature exceeding 1723 K using silicon-carbide rods. The cast iron with the compositions of Table 1 is melted in the crucible under an argon atmosphere and is continuously stirred, and then cooled from above the liquidus temperature to the predetermined one in the liquid–solid region. The temperature of semi-solid slurry is measured with a thermocouple inserted within stirrer, and the solid fraction corresponding to this temperature is calculated using

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Scheil’s equation [5]. During stirring, the torque is detected using a torque-meter (0 ~ 0.2 N m) and this torque value is estimated as apparent viscosity of semi-solid slurry at the given temperature. In this study, ceramic cylinder tube is used as the stirrer. Finally all of specimens are quenched into water.

The ceramic cylinder tube has a radius \( r_i \) 10.5 mm and the cylindrical crucible has an inner wall of radius \( r_o \) 20 mm and, thus a ring of thickness \( r_o - r_i \) 9.5 mm will appear for semi-solid region as shown in Fig. 2. During mechanical stirring, the solid particles move to outer or inner direction of the crucible under centrifugal field depending on the particle density relative to that of the melt. The experimental conditions are shown in Table 2.

2.2. Microscopic observation and mechanical test

The microstructure and hardness testing of semi-solid cast iron were investigated. Samples were cut and observed for microstructure of matrix. Investigation of microstructure was carried out for water-quenched semi-solid samples cut at 20 mm from the top surface. Average diameter of particle and fraction solid of slurry were measured by using image analyzer. Specimens were investigated on fractional particle area (number of solid particle multiplied by surface area of particle and divided by total area) in every cell area 0.95 mm² along radial distance from stirrer wall. Specimens were tested for Vickers hardness.

### Nomenclature

- \( \gamma \): centrifugal acceleration, \( m/s^2 \)
- \( r_i \): outer radius of ceramic cylinder, \( m \)
- \( r_o \): inner radius of crucible, \( m \)
- \( R_p \): particle radius, \( m \)
- \( \omega \): angular velocity, \( s^{-1} \)
- \( r \): radial position of particle, \( m \)
- \( \rho_p \): density of particle, \( kg/m^3 \)
- \( \rho_l \): density of liquid, \( kg/m^3 \)
- \( \Delta \rho \): density difference between particle and liquid, \( kg/m^3 \)
- \( \eta \): liquid viscosity, \( Pa.s \)
- \( d \): particle diameter, \( m \)
- \( R_0 \): position of particle at initial state, \( m \)
- \( R_1 \): position of particle after moving, \( m \)
- \( \Delta t \): time elapse, \( s \)
- \( \Omega \): rotational speed, \( rpm \)
- \( f_s \): fraction of solid, \( - \)
- \( A = \frac{\omega^2 d^2 \Delta \rho}{18} \)

### Table 1

| Heat No. | C  | Si  | Mn  | P  | S  | CE | Other |
|----------|----|-----|-----|----|----|----|-------|
| FC1      | 2.46 | 2.07 | 0.08 | 0.01 | 0.01 | 3.13 | –     |
| FC2      | 2.60 | 2.48 | 0.18 | 0.04 | 0.01 | 3.44 | 0.70 Cu |
| FC3      | 2.56 | 2.43 | 0.20 | 0.04 | 0.01 | 3.44 | 0.92 Cr |
| FC4      | 2.43 | 2.22 | 0.21 | 0.04 | 0.01 | 3.18 | 0.81 Mo |
| FC5      | 2.70 | 2.45 | 0.14 | 0.04 | 0.01 | 3.53 | 1.79 Ni |

\( CE = C + \frac{1}{3} (Si + P) \).

### Table 2

| Heat No. | Experimental conditions |
|----------|-------------------------|
|          | Rotational speed, \( \Omega \) (rpm) | Holding time (s) | Fraction of solid, \( f_s \) (–) |
| FC1      | 2, 5                     | 0, 300, 600     | 0.22, 0.35, 0.48 |
| FC2–FC5  | 2                       | –               | 0.7               |

Fig. 1. Schematic illustration of experimental apparatus.

Fig. 2. Schematic illustration of ceramic stirrer.
2.3. Chemical analysis

Electron Probe Microanalysis (EPMA) was conducted at an acceleration voltage of 15 kV to identify the chemical compositions of the semi-solid processed specimens for two areas namely; primary austenite and eutectic cell regions.

2.4. Theoretical analysis

The particle segregation occurs during the process with centrifugal force owing to difference in densities between molten metal and particles. Then a particle which is suspended in molten metal is submitted to a vertical acceleration due to gravity $g$ and to centrifugal acceleration $\gamma = \omega^2 r$.

Fig. 3 shows force fields where particles in a liquid will be acted on by the centrifugal, gravity and drag forces. The vertical moving of the particles can be ignored because centrifugal acceleration is much greater than gravity [6].

At steady state, $d^2 r/dt^2 = 0$. The particle velocity can be derived as Eq. (2):

$$V_r = \frac{dr}{dt} = \frac{d^2 \Delta \rho}{18 \pi \eta \omega^2 r}$$  \hspace{1cm} (2)

where $d$ is particle diameter; and $\Delta \rho = \rho_p - \rho_l$, the density difference between particle ($\rho_p$) and liquid ($\rho_l$). If $\Delta \rho = \rho_p - \rho_l > 0$, $V_r > 0$ and the particles move outward. For $\Delta \rho < 0$, $V_r < 0$ and the particles move inward [3].

After integration of Eq. (2),

$$R_1 = R_0 e^{(A)/(\eta) \Delta t} \hspace{1cm} (3)$$

where $A = \omega^2 d^2 \Delta \rho/18$; $R_0$ is position of particle at initial state; $R_1$ is position of particle after moving; $\Delta t$ is time elapse.

Tables 3 and 4 show calculation conditions for the case of FC1 and FC2–FC5, respectively. In the Tables 3 and 4, diameter of particle measured by image analyzer and averaged value was used for both the unalloyed and alloyed cast iron. Fraction solid of slurry is calculated using Scheil’s equation. Its calculation from the average of summation of primary particles volume ratio is done for every 0.95 mm$^2$ along radial distance.

2.5. Numerical procedure

Variation of volume fraction of solid particles due to particle moving under the action of centrifugal force is shown illustratively in Fig. 4. Flow chart of numerical procedure is shown in Fig. 5. The positions of particles are solved iteratively using a numerical solution procedure with 10 fixed-grids in the following order:

1. Set initial condition.
2. Compute volume ratio of primary particle.
3. Next time step (time $= \text{time} + \Delta t$).
4. Calculate position change of particle using Eq. (3).
5. Compute number of particles and fraction of solid at every grid.
6. Check if primary particle in some grids are greater than the maximum primary particle determined by the critical fraction solid.

If not, step 3 through 6 are repeated.

If yes, check if position of particle is over than 9.5 mm (i.e. $r_{i(j)} < 9.5 \text{ mm}$):

Table 3
Calculation condition for FC1 (unalloyed semi-solid cast iron)

| Parameter                              | Value          |
|----------------------------------------|----------------|
| Diameter of particle (m)              | $0.201 \times 10^{-3}$ |
| Density of particle (kg/m$^3$)        | $7.84 \times 10^3$ |
| Density of molten metal (kg/m$^3$)    | $7.23 \times 10^3$ |
| Fraction solid of slurry (–)          | 0.22, 0.35      |
| Cooling rate (K/s)                    | $1.34 \times 10^{-2}$ |
| Rotation speed (rps)                  | 2, 5            |

Table 4
Calculation condition for FC2–FC5 (alloyed semi-solid gray cast iron)

| Parameter                              | Value          |
|----------------------------------------|----------------|
| Diameter of particle (m)              | $0.300 \times 10^{-3}$ |
| Density of particle (kg/m$^3$)        | $7.84 \times 10^3$ |
| Density of molten metal (kg/m$^3$)    | $7.23 \times 10^3$ |
| Fraction solid of slurry (–)          | 0.7            |
| Cooling rate (K/s)                    | $1.34 \times 10^{-2}$ |
| Rotation speed (rps)                  | 2              |
If not, calculation ends \( f_s \) is corrected to \( f_{sc} \).

If yes, add the extra primary particles to another grid backward and repeat step 3 through 6.

7. Stop and put out data.

3. Results and discussion

3.1. Viscosity and microstructures

In these experiments, the slurry is continuously stirred and cooled from above liquidus to the liquid–solid region. The cooling rate is constant while stirring speed is varied as 2 and 5 revolutions per second (rps), respectively. Fig. 6 shows the relationship between apparent viscosity, fraction of solid and rotation speed of stirrer. It can be seen that the apparent viscosity increases with increasing fraction of solid and decrease with increasing rotation speed. Viscosity data in Fig. 6 is used in the present numerical analysis.

Fig. 7 shows the effect of rotation speed and fraction of solid (0.22 and 0.35) on particles distribution of unalloyed semi-solid cast iron (FC1). The higher rotation speed at the same fraction of solid gives the higher centrifugal force and gives denser primary particles concentrated more in outside

Fig. 4. Variation of fractional particle area along radial distance due to particle moving under centrifugal force.

Fig. 5. Flow chart.

Fig. 6. Effect of rotation speed on the apparent viscosity of semi-solid cast iron (FC1).
than inner side. Higher fraction of solid gives higher number of solid particles especially at the outside.

Fig. 8 shows the microstructure of 0.48 fraction of solid of unalloyed semi-solid cast iron (FC1). It is seen that wall appears in the middle of samples. Large primary solid particles are segregated in the inside region, whereas smaller particles are dissociated in the outside region. From viscosity results in Fig. 6 it can be concluded that at higher fraction of solid, viscosity becomes higher so particles are very difficult to move by centrifugal force. This gives such type wall seen in the middle of samples in Fig. 8, which means that particle compacted area is formed in the outer region. Utilizing this phenomena, property graded materials can be produced for cast iron.

Fig. 9 shows the effect of holding time of stirring at semi-solid temperature before water quenching of FC1. The melt is continuously stirred from above the liquidus temperature to the predetermined one in the liquid–solid region and held at that temperature for 0, 300, and 600 s. (So particles distribution is built already.) The results show that longer holding time gives denser primary particles concentrated more in outside than inner side as the effect of centrifugal force.

Fig. 10 shows microstructure of semi-solid alloyed cast iron at fraction of solid 0.7 for FC2–FC5. It is shown again that primary solid particles move more to outer region under the centrifugal force.

The result in Fig. 11 shows that carbon content increase with an increase in distance from outside to inner side. Carbon concentration is greater in the liquid iron than the solid particle.

The effect of alloying elements on the austenite segregation is shown from the EPMA results listed in Table 5. From the result it can be seen that graphite promotion elements such as silicon, nickel, and copper show a tendency to concentrate in the solidifying austenite phase, while carbide-promoting elements such as molybdenum and chromium tend to concentrate in the residual liquid iron. Silicon, nickel, and copper are rejected from the melt during solidification whilst segregation patterns of molybdenum and chromium indicate that these alloying elements associate with complex carbides. The microstructures in
Fig. 10 show that the solid particles condensed at outer side more than at inner side. The observation shows more ledeburite forms around the primary austenite at the inner side than at the outer side and the solid particles at the outer side are rounder than at the inner side. However, any effects of alloying elements are not observed on primary austenite distribution, particles size and number of particle of semi-solid metal at the same rotation speed and cooling rate.

Fig. 9. Effect of isothermally holding time on the structure of cast iron (FC1, Fraction of solid:0.48) (I: Rotation speed 2 rps, Cooling rate 1.34 x 10^{-2} K/s; II : Rotation speed 5 rps, Cooling rate 1.34 x 10^{-2} K/s).

Fig. 8. The structure of semi-solid cast iron (FC1, Fraction of solid:0.48) (I: Rotation speed 2 rps, Cooling rate 1.34 x 10^{-2} K/s; II : Rotation speed 5 rps, Cooling rate 1.34 x 10^{-2} K/s).
3.2. Mechanical properties

Figs. 12 and 13 show the results of Vickers hardness measured at radial position of the sample for unalloyed and alloyed semi-solid cast iron, respectively. Vickers hardness is shown to be dependent on the quantity of ledeburite produced. Namely more formed ledeburite, larger Vickers hardness. In the present rapid cooling condition (quenching) after semi-solid processing, ledeburite tends to form more inside because primary austenite quantity decreases due to centrifugal effect and carbon-rich eutectic liquid increases inside compared with outside. So ledeburite quantity increases inside after quenching, yielding larger values of hardness there. This leads to a Vickers hardness gradient in the radial direction.

The effect of rotation speed on Vickers hardness is that higher rotation speed gives more hardness gradient in the radial direction. The number of primary austenite at

Table 5
Chemical analysis of semi-solid processed iron

| No. | Area    | C  | Si  | Cu  | Cr  | Mo  | Ni  |
|-----|---------|----|-----|-----|-----|-----|-----|
| FC2 | Austenite | 2.14 | 2.43 | 0.71 | –   | –   | –   |
|     | Eutectic | 5.90 | 1.40 | 0.34 | –   | –   | –   |
| FC3 | Austenite | 2.12 | 2.27 | –   | 0.64 | –   | –   |
|     | Eutectic | 6.73 | 0.86 | 2.7  | –   | –   | –   |
| FC4 | Austenite | 1.92 | 2.24 | –   | –   | 0.40 | –   |
|     | Eutectic | 6.60 | 0.49 | 5.16 | –   | –   | 1.85 |
| FC5 | Austenite | 1.90 | 2.24 | –   | –   | –   | 1.85 |
|     | Eutectic | 6.28 | 1.04 | –   | –   | 0.51 | –   |
inner side for higher rotation speed is less than that for lower rotation speed. It causes more ledeburite formation at inner side for higher rotation speed more than the other case. This gives a promising method to produce hardness-graded cast iron that will be applied to parts with surface wearability.

3.3. Comparison of primary particle volume ratio between the experiment and the simulation

Fig. 14 shows the comparison on primary particle volume ratio between the experiments and the simulation, which shows the effect of both the fraction of solid and rotation speed of unalloyed semi-solid cast iron on the solid particle distribution. Here the calculation explains reasonably that primary particles increase steeply along radial distance with the increase of solid fraction and rotation speed.

Fig. 15 shows the comparison result on primary particle volume ratio for alloyed semi-solid cast iron between the experiments and the simulation.

The simulation results show that radial particles distribution is also obtained in the figure by the action of centrifugal force and these results are in good agreement with the experiment.

In this way the characteristic microstructure with primary phase distribution is produced by the effect of centrifugal force, yielding inclined distribution of hardness values. Such a microstructure change is explained reasonably by the simulation. This simulation can be used to detect the optimized condition such as stirring rate, holding time, etc., in order to produce reasonable functionally graded material.
4. Conclusions

In the present study, primary phase distribution produced by centrifugal effect was investigated for semi-solid processing of cast iron. The results obtained are:

1. Centrifugal effect produces the primary particles distribution along the radial direction. Denser particles are concentrated in outside than inner side. It is seen that wall appears in the middle of high fraction of solid samples because high viscosity region is formed outside, so particles are very difficult to move by centrifugal force.

2. Alloying elements promoting carbide formation such as chromium and molybdenum concentrate positively along the cell boundaries. On the other hand silicon, copper and nickel promote graphite formation, and hence show negative segregation giving lower concentration in eutectic than austenite. Alloying elements addition has no effect on primary austenite distribution, particles size and number of particle of semi-solid metal at the same rotation speed and cooling rate.

3. Hardness value changes in radial direction due to the segregation of austenite particles and hence carbon concentration change in radial direction. A property graded material will be produced using this mechanism.

4. Primary austenite particle distribution is explained reasonably by the simulation, considering change of viscosity with solid fraction in the centrifugal processing.

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