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Behavior of particles in front of metallic solid/liquid interface in electromagnetic field①

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Abstract: The first part deals with the behavior of particles theoretically, and the critical electromagnetic force needed to alter the behavior of particles was deduced under different conditions. It was proposed that applying electromagnetic force would change the distribution coefficient of the particles. By using the data from literatures, the migrating rate of SiC particle by electromagnetic force was calculated, which is far more than the critical rate of solidifying interface which will result in the engulfment of the SiC particle in the Al-SiC matrix metal. Therefore the possibility of controlling the behavior of the particles in front of the solidifying interface by electromagnetic field was confirmed. In the second part, by using simulative experiments, the man-made alternation of the behavior of the particles in front of the solidifying interface under electromagnetic field was observed, and the idea of changing the distribution of the particles in solidified metal by electromagnetic force was verified experimentally. It is shown that, the particle, which would be engulfed by the solidifying interface, would escape from the interface under electromagnetic buoyant force (EMBF), and the particles adherent to the interface would migrate toward it and be engulfed finally under EMBF. Further more, the particles being pushed by the interface would stay at the interface, the repulsive force exerted on the particles would be counteracted by EMBF, and then the particle would turn to be engulfed. Adjusting the direction and magnitude of EMBF could alter the distribution of the particles in the solidifying metal.

Key words: solidification; particle; electromagnetic field; pushing/engulfment; distribution of particles
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1 INTRODUCTION

The interaction between the particles and the solidifying interface is often involved in the processing of metallic materials. For example, during the solidification of the metal casting, the inclusion in the molten metal (including foreign inclusion and secondary inclusion) is possible to be engulfed by solidifying interface to stay in the casting, also it is possible to be pushed by the interface to retain in the molten metal. Another case is, the reinforcing particles will be engulfed to enter the solid and possibly pushed by the interface, which will result in uneven distribution of the reinforcing phase in solid during processing the metal-matrix composites (by the way of casting).

Such pushing/engulfment behavior of the particles determines their distribution in metal and influences the properties of metal material detrimentally, so the phenomenon was widely concerned by researchers[1-3]. The former research showed that the behavior of the particles was affected significantly by the difference of the surface free energy of the system, size and rigidity of the particles, acceleration of gravity[4-7], the ratio between the heat transmitting coefficient of the particles and that of the solidified metal[8], shape of the interface, viscosity of the fluid (liquid metal) and the gradient of the temperature etc. However, Han and Hunt[10, 11] considered that the flow of the fluid in front of the solidifying interface due to the gradient of temperature and concentration led to the pushing of the particle. Consequently, the behavior of the particle could not be controlled artificially up to now[12, 13] since the parameters involved in the process are very complex, and the consistent standpoint on the decisive parameters that determine the behavior of the particle is not acquired.

The authors noticed that Uhlmann considered the particle in front of the solidifying interface was acted by two kinds of forces, one was pushing force, which would urge the particle to be pushed, another was drag force, which would urge the particle to be engulfed, and the behavior of particles would be determined by the preponderant force. Considering the electromagnetic field has the advantage of exerting

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force on things without contact, if the electromagnetic force (EMF) was introduced to the solidifying molten metal, the particle would be acted by electromagnetic buoyant force, and then the force balance of the particle would be broken, which would result in changing the behavior of the particle. By adjusting the EMF and the solidifying process fittly, the behavior of the particle will be controlled, so does the distribution of the particle in solid. This paper dealt with the idea theoretically and experimentally.

2 FORCES EXERTED ON PARTICLE IN MOLTEN METAL

There are three main kinds of force exerted on a particle in front of the solidifying interface[10, 18], the first one is the virtual gravity due to the difference between the density of the particle and that of the fluid, the second one is the pushing force by the interface, and the last one is the viscous force. When there is flow in fluid, force is also exerted on a particle near a surface due to the fluid velocity gradient[10], just as most of the researchers, we assume there is no flow in the fluid. Furthermore, the force exerted on a particle will be different in front of interface with different geometries, and in this paper, only smooth interface and spherical particle was concerned.

The three forces are as follows.

1) virtual gravity

$$F_G = \frac{4}{3}\pi r_p^3 \Delta\rho g \tag{1}$$

where r_p is the radius of a particle, $\Delta\rho$ is the difference between the density of a particle and that of molten metal, and g is the gravity acceleration.

2) pushing force by interface[19]

$$F_I = 2\pi r_p \Delta\sigma_0 \left(\frac{a_0}{a_0 + h}\right)^n \tag{2}$$

where $\Delta\sigma_0$ is the difference among the surface free energy of a particle, solid and liquid phase, a_0 is the distance between two closer atoms, h is the span between a particle and the interface; the value of n is related to the shape of the interface, to smooth one, $n = 7-8$[10], here we assume $n = 7$.

3) viscous force

$$F_D = 6\pi\eta v_p r_p \xi \tag{3}$$

where η is the viscosity of the fluid, v_p is the velocity of the particle, ξ is the modified coefficient, when a particle suspends in fluid freely, $\xi = 1$; when the particle is near the solidifying interface ($h \to 0$)) and migrate toward to it, $\xi = r_p/h$; when the particle moves along the interface, $\xi = \ln \frac{r_p}{h}$[10].

Compared to the direction of the gravity, there are two representative interfaces, that are horizontal interface and vertical interface, the forces exerted on a particle are shown in Fig. 1. This paper mainly deals with the former case to simplify the process.

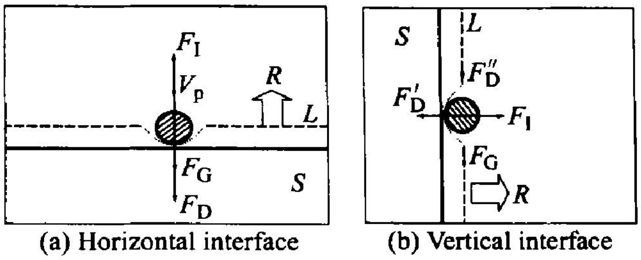


Fig. 1 Sketch map of forces exerted on a particle in front of solidifying interface

When interacting with the interface, a particle must experience the following steps:

a) Solidifying interface advances toward with a velocity of R;

b) The interface approaches the particle;

c) Pushing force is exerted on the particle, then the particle obtains a velocity v, and moves along the interface;

d) When $v \geqslant R$, the particle would either move along with the interface, or escape from it, and then the pushing force exerted on the particle would decrease until the three forces, F_I, F_G, F_D reach a balance, at last the particle also moves along the interface with a velocity of R;

e) When $v < R$, the particle would be engulfed by the advancing interface.

The above three kinds of forces determine the behavior of the particle in usual circumstance. The parameters that the former researchers referred such as surface energy, gradient of temperature, diameter of the particle etc would all influence the force balance of the particle, and the behavior of the particle as well. So, if we introduce a foreign force (such as EMF) to the solidification process, the break down of the force balance of the particle is reasonable. Considering that the velocity of the particle would change also by applying foreign force, we can bring out the following criteria determining the behavior of the particle based on the above steps and the conclusions by Uhlmann,

1) When the velocity of the particle under the above three kinds of forces and foreign force is larger than or equals that of the advancing interface, that is $v_p - R \geqslant 0$, the particle will be pushed by the interface;

2) When $v_p - R < 0$, the particle will be engulfed by the interface.

The following study will be based on the criteria.

3 INFLUENCE OF EMF ON PUSHING/ENGULFMENT BEHAVIOR OF PARTICLE IN FRONT OF SOLIDIFYING INTERFACE

3.1 Force exerted on non-conductive particle in conductive fluid under electromagnetic field

As seen in Refs. [14, 15], if the conductive fluid was acted by EMF ($\boldsymbol{F}_{EMF} = \boldsymbol{J} \times \boldsymbol{B}$), the electromagnetic buoyant force (EMBF) $\boldsymbol{F}_p$ exerted on a sphere particle suspended in the fluid is,

$$F_p = -\frac{\pi \varphi d_p^3}{4} F_{EMF} = -2\pi\varphi r_p^3 F_{EMF} \quad (4)$$

where $\varphi = \frac{\sigma_f - \sigma_p}{2\sigma_f + \sigma_p}$, σ_f and σ_p are the conductivities of fluid and particle separately. When $\sigma_p \ll \sigma_f$ (that is, the particle is non-conductive), $\varphi = \frac{1}{2}$, so F_p is

$$F_p = -\pi r_p^3 F_{EMF} \quad (5)$$

where the minus shows that the direction of F_p is opposite to that of F_{EMF}.

In most of the considerations involved in this paper, the particles (such as inclusions, reinforcing phase) are non-conductive ($\sigma_f \ll \sigma_p$). When applying EMF, the EMBF will exert on the particle, which will break its force balance, and the direction and the magnitude of the velocity of the particle will be changed too, so the pushing/ engulfment behavior of the particle will be changed according to the above criteria.

3.2 Horizontal interface

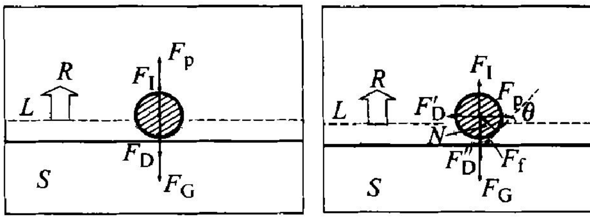
The angle between the EMF and the interface can be controlled to be any value from 0° to 360°. In this paper, only two representative situations are considered, one is that the direction of the EMF is perpendicular to the interface; the other is that the EMF is parallel to the interface. The forces exerted on a particle in front of a horizontal solidifying interface under EMF are shown in Fig. 2.

3.2.1 EMF perpendicular to interface

Because all forces exerted on the particle keep on a beeline, the critical EMF needed to change the original behavior of the particle could be easily deduced according to on the above criteria, as shown in Table 1.

3.2.2 EMBF parallel to interface

If the particle is pushed by the interface originally, applying EMF will not change its behavior. However if the particle is engulfed by the interface originally, things will reverse. When the EMBF is large enough, the particle will roll on the interface, and it can not be engulfed[10]. The forces exerted on the particle are shown in Fig. 2(b). Obviously, the forces in the horizontal and vertical directions should keep balance. So the following equations could be obtained from Fig. 2(b).



(a) EMF perpendicular to interface (b) EMF parallel to interface

Fig. 2 Forces balance of particle in front of horizontal solidifying interface under EMF

1) In the horizontal direction,

$$F_p = F'_D + N\sin\theta + F_f\cos\theta \quad (6)$$

2) Since the particle rolls on the interface and could not be engulfed, then in the vertical direction, it would win a velocity R and advance along the interface, the following equations could be obtained,

$$F_I + N\cos\theta = F''_D + F_G + F_f\sin\theta \quad (7)$$

$$F'_D = 6\pi\eta v_p r_p \ln\frac{r_p}{h} \quad (\xi = \ln\frac{r_p}{h}) \quad (8)$$

$$N = \frac{F_f}{f} \quad (9)$$

$$F''_D = 6\pi\eta R r_p^2/h \quad (\xi = r_p/h) \quad (10)$$

where N is the counterforce exerted on the particle by interface, F_f is the frictional force, f is the roll or slip frictional coefficient, v_p is the roll velocity of the particle. v_p can be obtained by solving the equations from (6) to (10),

$$v_p = \frac{1}{6\eta\ln(r_p/h)}\left\{2F_{EMF}r_p^2 - \frac{f+\mathrm{tg}\,\theta}{1-f\,\mathrm{tg}\,\theta}\left[6\eta R\frac{r_p}{h} + \frac{4}{3}r_p^2\Delta\rho g + 2\Delta\sigma_0\left(\frac{a_0}{a_0+h}\right)^7\right]\right\} \quad (11)$$

In order to keep the particle rolling and not be engulfed, the roll velocity must be larger than zero,

Table 1 Critical EMF needed to change original behavior of particle when EMBF points are perpendicular to interface

Original force relationship	Original behavior of particle	Direction of EMBF	Critical EMF	Behavior of particle under EMF
$F_I \geqslant F_D + F_G$	Pushing	F_p and F_I keep reverse direction	$F_{EMF} > \frac{6\Delta\sigma_0 h\left(\frac{a_0}{a_0+h}\right)^7 - 4r_p^2\Delta\rho g h - 18\eta r_p R}{3hr_p^2}$ ($\xi = r_p/h$)	Engulfment
$F_I < F_D + F_G$	Engulfment	F_p and F_I keep same direction	$F_{EMF} > \frac{18\eta R + 4r_p^2\Delta\rho g - 6\Delta\sigma_0\left(\frac{a_0}{a_0+h}\right)^7}{3r_p^2}$ ($\xi = 1$)	Pushing

and then the EMF should be

$$F_{\mathrm{EMF}} > \frac{f + \mathrm{tg}\ \theta}{1 - f\,\mathrm{tg}\ \theta} \frac{1}{2r_{\mathrm{p}}^{2}} \cdot \left[6\eta R \frac{r_{\mathrm{p}}}{h} + \frac{4}{3} r_{\mathrm{p}}^{2} \Delta\rho g + 2\Delta\sigma_{0} \left(\frac{a_{0}}{a_{0} + h} \right)^{7} \right] \tag{12}$$

3.3 Influence of EMF on distribution coefficient of particles

As shown in Refs. [2, 11], the behavior of the particles would influence the distribution coefficient between solid and liquid,

$$k_0 = P(R - v_{\mathrm{p}})/R \tag{13}$$

where R is the velocity of the advancing interface, P is pushing/engulfment function (for pushing, $P = 0$ for engulfment then $P = 1$), and v_{P} is the velocity of the particle perpendicular to the interface.

Adjusting the direction and magnitude of either magnetic field or electric current can change that of the EMF, and both the magnetic field and electric current can be the function of time t, then the EMF can be the function of time too, as shown in equation (14),

$$F_{\mathrm{EMF}} = Af(t) \tag{14}$$

where A is a constant, $f(t)$ is the function of time. From the above discussion, it is known that the pushing/engulfment function P and v_{P} can be the function of F_{EMF}, then P and v_{P} can be rewritten as

$$P = f'(F_{\mathrm{EMF}}) = f'(Af(t)) \tag{15}$$

$$V_{\mathrm{p}} = Mf''(F_{\mathrm{EMF}}) = Mf''(Af(t)) \tag{16}$$

where M is a constant, both $f'(x)$ and $f''(x)$ are the functions of independent variable x. Substituting equations (15) and (16) into equation (13) yields

$$k_0 = f'[Af(t)]\{R - Mf''[Af(t)]\}/R \tag{17}$$

Equation (17) shows that, applying an EMF varying with time will influence the distribution coefficient of the particles between solid and liquid, that is, influence the distribution of the particle in solidifying metal.

3.4 Comparison between migrating velocity of particle by EMF and critical velocity of solidifying interface

It was reported in Ref. [19] that, the SiC particle with a diameter of 40 μm was pushed by the solidifying interface and won a velocity of 100 μm/s, and the critical velocity of the solidifying interface that would result in the engulfment of the SiC particles was 366 μm/s. In this paper, assuming that the current density is 10^5 A/m^2, magnetic field intensity is 1 T, and the viscosity of the molten aluminum is 0.005 Pa·S[19], then taking into account equation (4), the migrating velocity of SiC particles by EMF can be calculated,

$$v_{\mathrm{p}} = \frac{d_{\mathrm{p}}^{2} J \cdot B}{24\eta} = 1\,333\ \mu\mathrm{m/s} \gg R_{\mathrm{critical}}$$

The calculated velocity of the SiC particle under EMBF is much larger than that of the critical velocity of the solidifying interface that will result in the engulfment of the particles, so applying EMF to alter the pushing/engulfment behavior of the particle in front of the solidifying interface is very possible in theory.

4 EXPERIMENTAL

4.1 Simulative experiment of observing behavior of particles in front of solidifying interface under electromagnetic field

Based on the principle of comparability, we used copper sulfate solution to simulate conductive molten metal, and hollow alumina sphere to simulate non-conductive particles. The container consists of two parallel copper plates by which the current can flow through the electrolyte, cooling end, baffle and viewfinder. One NbFeB (B_{r} = 1 T) was attached on the bottom of the viewfinder to supply magnetic field. The cooling end was cooled by liquid nitrogen, and the cooling velocity (the velocity of the advancing interface) could be adjusted by controlling the flow rate of liquid nitrogen. A microscope and a video capture were used to observe and capture the pictures of the process of solidification. The density of the copper sulfate solution and the hollow alumina spheres is 1.2−1.6 g/cm^3 and 1.0−2.0 g/cm^3, and by floatation we can obtain the spheres whose density are near that of the solution. The velocity of the advancing interface is in the range of 50−80 μm/s, and the range of the diameter of the sphere is 450−550 μm. The sketch map of the experiment is shown in Fig. 3.

4.2 Experiment of changing distribution of particles in solidifying metal by applying EMF

In this experiment, we use hypereutectic aluminum-silicon alloy, and the primary silicon-rich crystal is used to simulate the non-conductive particles[20]. The molten alloy, whose temperature is 750 ℃, is poured into the container consisting of heat-resistant material, heating element, carbon electrode and cooling water pipe (the container is heated to 600 ℃). The current flows in the molten alloy through the two carbon electrodes, and the whole container is put in the magnetic field produced by an electromagnet. When the height of the molten alloy reaches the set value, the current is introduced to the molten alloy, and then the molten alloy is acted by EMF (B = 0.12 T, J = 56 250 A/m^2). At the same time, all sides and top of the container are covered by heat-resistant fibers, and the cooling water begins to flow through the pipe, then the molten alloy would solidify from bottom to top. Adjusting the direction and magnitude of the current could change that of the EMF, and adjusting the flowing rate of the cooling water could change that of the advancing interface.

We begin to counter the time when the water flows, and end the timer when the molten alloy wholly solidifies. Take out the solidified alloy after cooling, and cut it in the middle section perpendicular to the direction of the current. Polish the section and observe the distribution of the silicon-rich particles under microscope. The experiments without EMF are carried out parallelly. The sketch map of the experiment is shown in Fig. 4

4. 3 Results and discussion

4. 3. 1 Behavior of particles in front of solidifying interface by applying electromagnetic field

1) Particles adherent to solidifying interface

The adherent position here involved refers to the position that its distance to the solidifying interface is three to five times of the diameter of the particle, and the behavior of the particles is not known. At this time, we apply EMF in electrolyte with its direction pointing against the solidifying interface, and the particles migrate toward the interface and reach there, then are engulfed by it. The observed pictures are shown in Fig. 5.

2) Behavior of being engulfed particle by mushy area

In Fig. 6, when the particle is being engulfed by solidifying interface (trapped by mushy area), we apply EMF to the electrolyte with the direction toward the interface, then the particle is exerted by a reversed force and tends to escape from the mushy area, which is different from that in Fig. 5.

3) Behavior of being pushed particle

Fig. 7 shows the same thing that the particle is being engulfed by solidifying interface under EMF, however, what different is that the particles are pushed intensively by interface when without EMF.

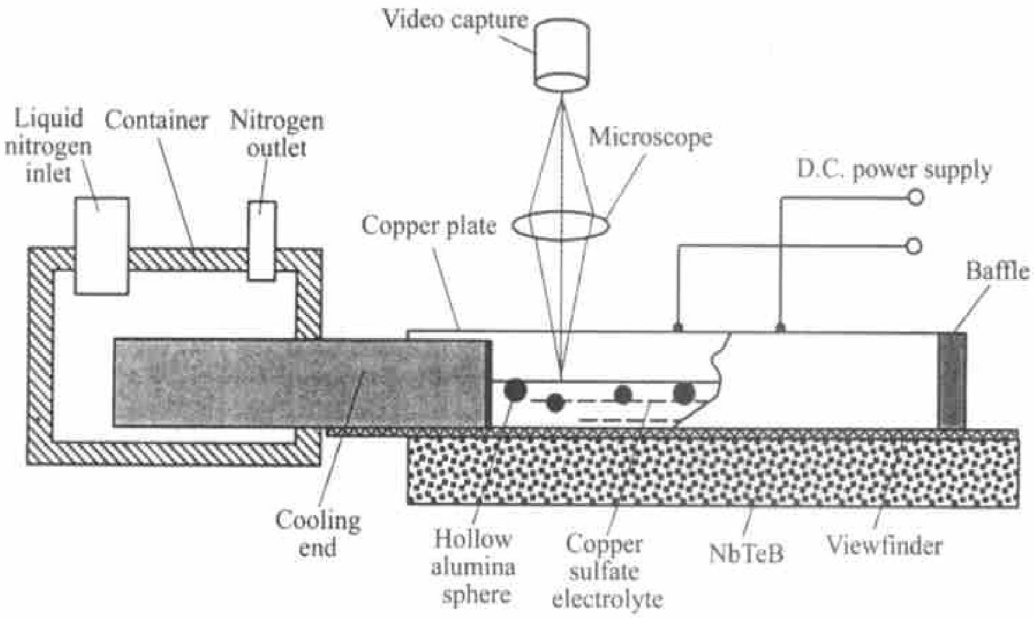


Fig. 3 Sketch map of equipment to observe behavior of particles in front of solidifying interface under electromagnetic field

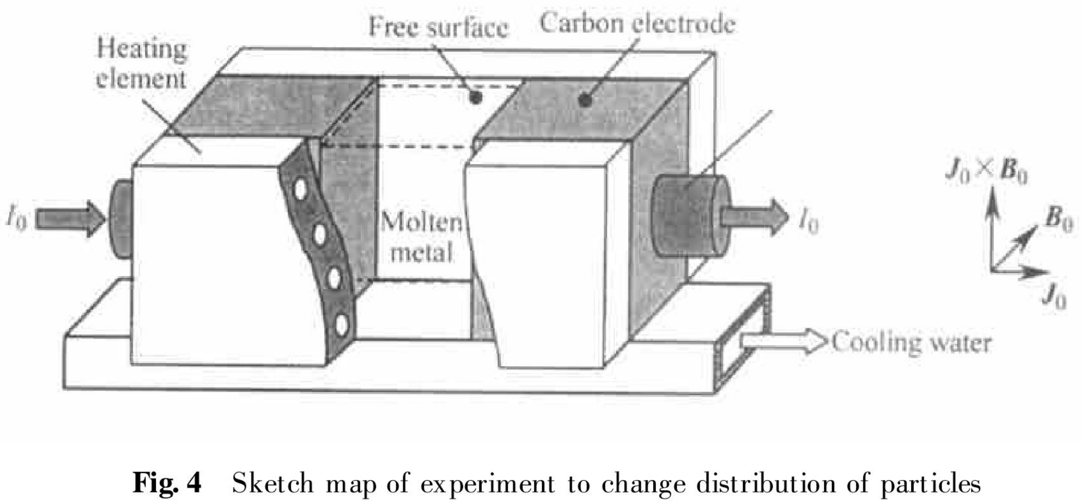


Fig. 4 Sketch map of experiment to change distribution of particles in front of solidifying metal by EMF

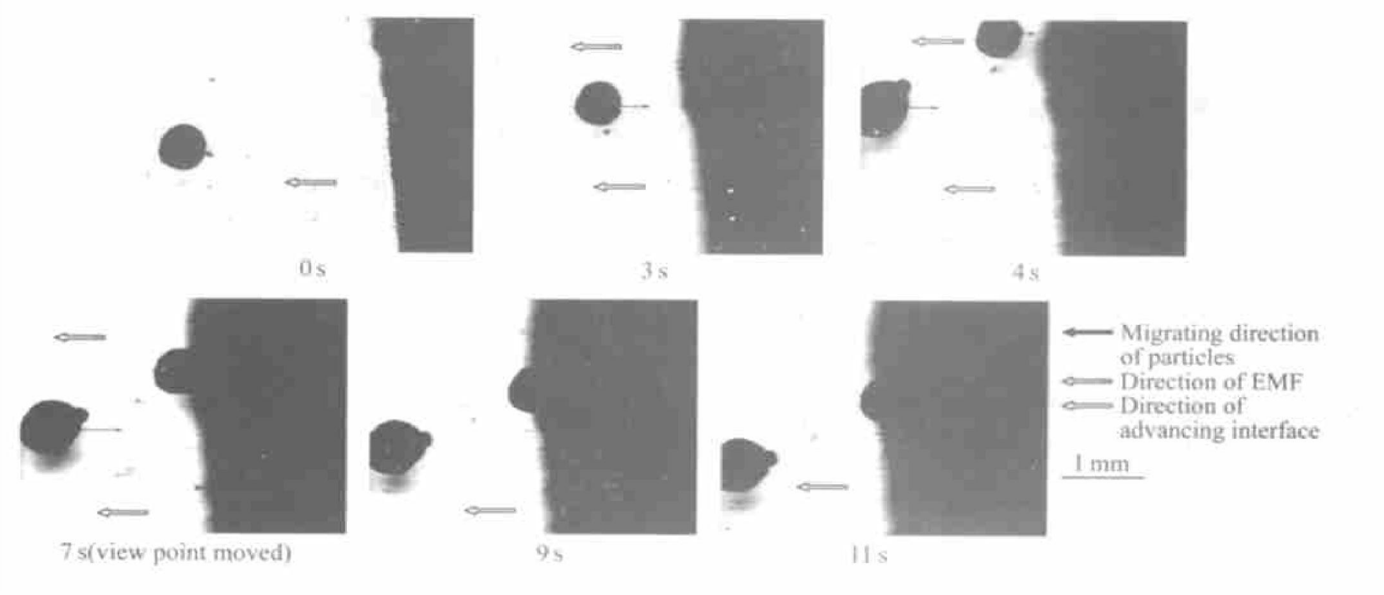


Fig. 5 Particle adherent to solidifying interface migrating to there and being engulfed under EMF

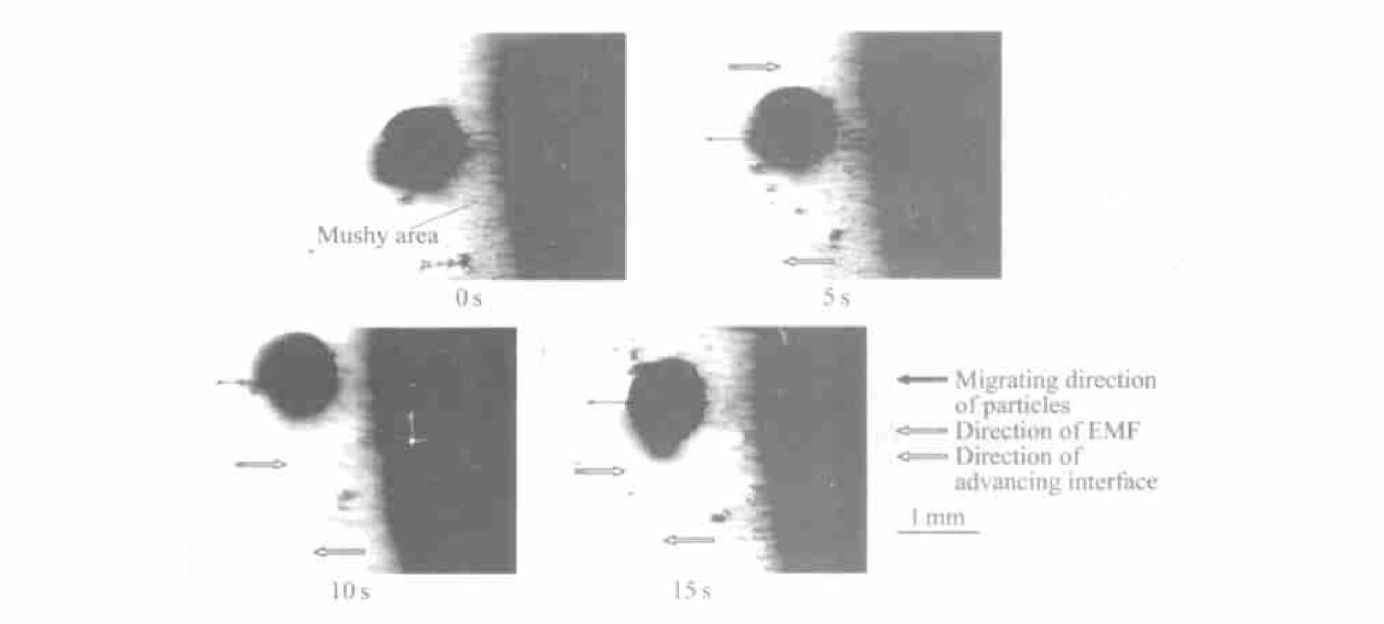


Fig. 6 Being engulfed particle in front of solidifying interface turning to be pushed under EMF

As shown in Fig. 7, the distance between the particles and the interface is elongated, which shows that the particles are pushed away from the interface during 0 to 11 th. In the period from 13 th to 41 st, the EMF is introduced, and the hollow alumina sphere migrates toward the interface, and then is engulfed by it subsequently. While since 41 st, when the EMF is retrieved, the particles would be pushed away from the interface again. The process effectively proves that the pushing/engulfment behavior of the particles in front of the solidifying interface could be altered indeed by applying EMF.

4.3.2 Redistribution of particles in metal under EMF

In order to make sure that the distribution of the particle in the metal could be altered by EMF, the experiment was carried out by using hypereutectic aluminum-silicon alloy (19% Si). The results are shown in Fig. 8. It is shown in Fig. 8 that,

1) When cooled by air and without EMF, the silicon-rich particles would float up due to that its density is lower than that of the molten alloy, then the solidifying interface could not capture the particles, which results in that they distribute in upper area of the solidified metal (Fig. 8(a));

2) When cooled by water and without EMF, the silicon-rich particles would be trapped immediately by interface due to higher velocity of the advancing interface, so they distribute in the lower area of the solid-

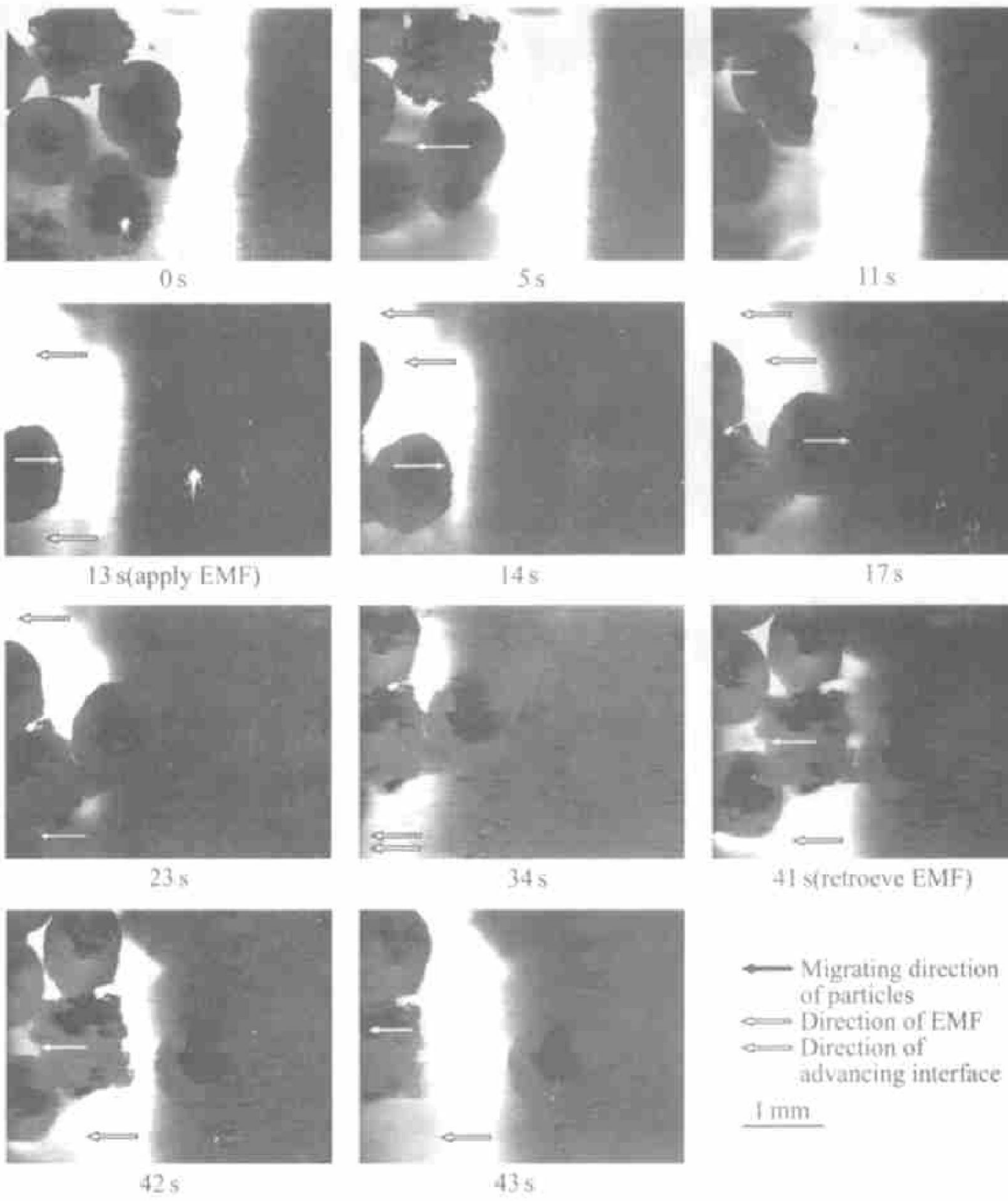


Fig. 7 Being pushed particle turning to be engulfed under EMF

ified metal (Fig. 8(b));

3) When cooled by air and with EMF upward, the silicon-rich particles are acted by electromagnetic buoyant force, and they tend to migrate downward to be trapped by solidifying interface, then distribute in the lower area of the solidified metal (Fig. 8(c));

4) When cooled by water and with EMF downward, one possible result is that the particles would be trapped due to higher cooling speed, another possible one is that the particles would escape from the interface due to EMBF and buoyant force. Fig. 8(d) shows that they distribute in the upper area, compared to Fig. 8(b), we could conclude that the particles escape from the interface due to applying EMF downward.

The relationship between the floating rate by gravity, migrating rate by EMF and advancing rate of interface is evaluated in Table 2. Based on Table 2, Table 3 is deduced to reveal the relationship between the migrating rate of the particles and the solidifying rate of metal. When EMF and the buoyant force keep the same direction, then the migrating rate of the particles equals the difference between the floating rate by gravity and migrating rate by EMF, but if their direction is reverse, then it equals the sum of the two rates.

It is displayed in Table 2 that, to the horizontal solidifying interface, the pushing/engulfment behavior of the particles is determined by the relationship between the migrating rate of the particles and the solidifying rate of metal under EMF. When the former is greater than the latter, the particles tend to escape from the interface, while when the relationship is reversed, they tend to be engulfed by the interface.

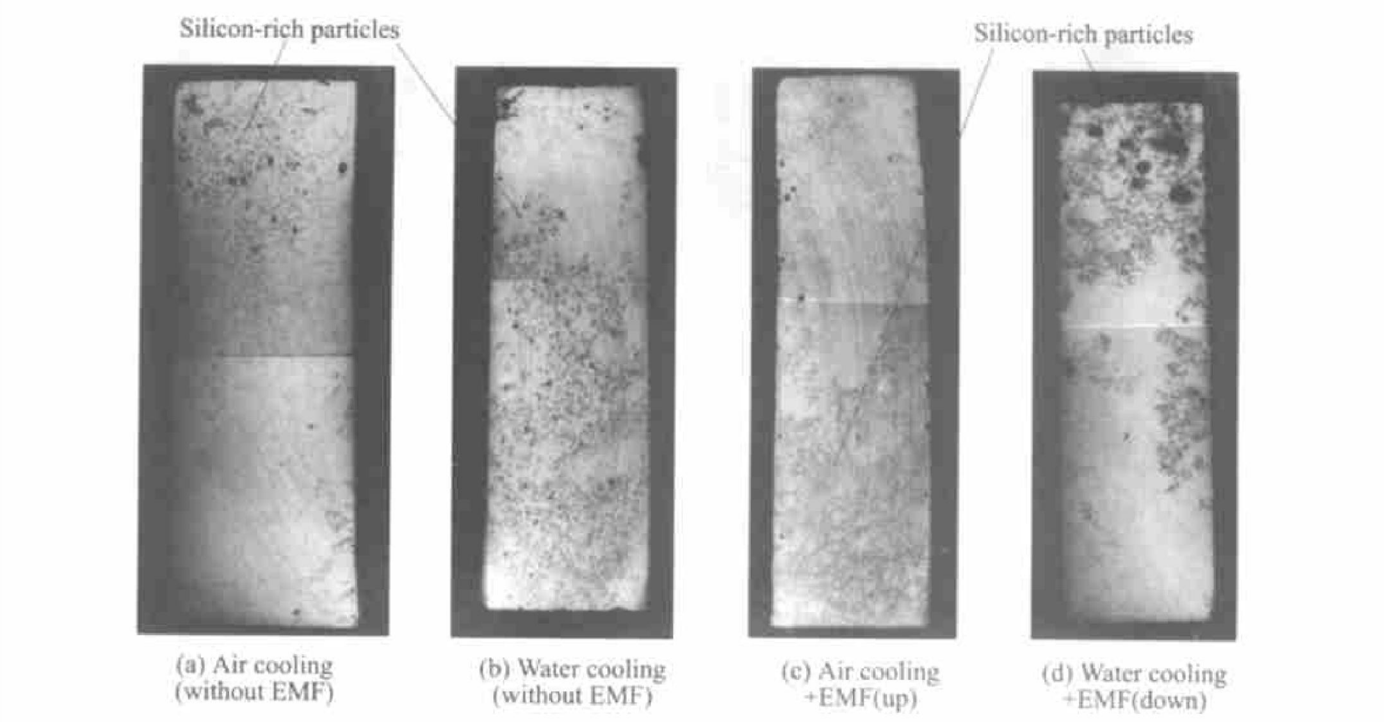


Fig. 8 Distribution of silicon-rich particles in metal under different solidification conditions

Table 2 Some parameters of solidification process of metal under electromagnetic field

Property	Value	Property	Value
Density of silicon-rich particle(750 ℃)/ ($g·cm^{-3}$)	2.31[20]	Resistance of silicon/ (Ω·m)	2.3×10^5
Density of molten aluminum-silicon alloy (750 ℃)/($g·cm^{-3}$)	2.53[20]	Floating rate by gravity/($mm·s^{-1}$)	2.19
Gravity/($N·m^{-3}$)	2.19×10^3	Floating rate by EMF/($mm·s^{-1}$)	4.22
Electromagnetic buoyant force/($N·m^{-3}$)	4.22×10^3	Advancing rate of interface by water cooling/($mm·s^{-1}$)	2.67
Flux of cooling water/h^{-1}	800	Advancing rate of interface by air cooling/($mm·s^{-1}$)	0.44
Average diameter of silicon-rich particles/mm	0.3		

Table 3 Relationship between migrating rate of particles and advancing rate of interface under different solidifying conditions and its influence on behavior of particles

Solidifying condition	Migrating rate of particle v_p/ ($mm·s^{-1}$)	Advancing rate of interface R/ ($mm·s^{-1}$)	Relationship between v_p and R	Behavior of particle
Air cooling and without EMF(Fig. 8(a))	2.19	0.44	$v_p > R$	Floating(pushing)
Water cooling and without EMF(Fig. 8(b))	2.19	2.67	$v_p < R$	Engulfment
Air cooling and with EMF(Fig. 8(c))	−2.03	0.44	$v_p < R$	Sink (engulfment)
Water cooling and with EMF (Fig. 8(d))	6.88	2.67	$v_p > R$	Floating (pushing)

5 CONCLUSIONS

1) The force balance exerted on the particle determines the pushing/engulfment behavior of the particles in front of the solidifying interface, and applying EMF would break the force balance of the particle, so its behavior would be altered too.

2) There is a critical EMF, above which the original behavior of the particles could be changed, and the equilibrium distribution efficient of the particles between solid and liquid phase could be changed, too.

3) It is shown in simulative experiments that,

the particle adherent to the solidifying interface would migrate toward the interface and stay there under EMF, then be engulfed by interface. The particle being engulfed by the solidifying interface would escape from the interface under EMF, and then turn to be pushed. The particles being pushed by the solidifying interface could stay at the interface under EMF, and the pushing force could be counteracted by EMBF, then the particles turn to be engulfed.

4) Applying EMF could change the distribution of the particle in solidified metal. The relationship between the migrating rate of the particles and the solidifying rate of metal under EMF dominats the behavior of the particles.

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