Effects of Spiral Magnetic Field on Structures Transformation and Macrosegregation of Sn-Pb Alloy

Xingguo ZHANG^{1)†}, Qian ZHAO¹⁾, Jianshe GUO¹⁾, Qingyuan XING¹⁾, Liang DENG¹⁾, Xiaoguang HOU²⁾ and Canfeng FANG¹⁾

1) School of Materials Science and Engineering, Dalian University of Technology, Dalian 116024, China

2) Baoshan Iron and Steel Company Limited, Shanghai 201900, China

[Manuscript received 12 December 2012, in revised form 22 January 2013]

© The Chinese Society for Metals and Springer-Verlag Berlin Heidelberg

The effect of spiral magnetic field (SMF) on the solidification process of Sn-20 wt.%Pb hypoeutectic alloy was studied in this work. The temperature distribution and the macrosegregation together with the eutectic lamellar spacing during solidification were measured under different magnetic fields. The results indicate that the compositions in the top and bottom portions of the sample are nearly the same and the segregation phenomenon is basically eliminated by SMF at an exciting current of 125 A. The morphology of the primary phase transforms from coarse dendrite to rosette or ellipsoidal equiaxed crystal and the spacing of the eutectic lamellar increases from 1.6 to 3.9 μm when the exciting current increases from 0 to 125 A. Moreover, if the exciting current is high enough, the forced fluid flow caused by SMF will enhance the process of transition from lamellar eutectic structure to anomalous rod-like one.

KEY WORDS: Spiral magnetic field; Forced convection; Eutectic lamellar spacing; Sn-Pb alloy; Composition segregation

1. Introduction

The electromagnetic stirring has various advantages, such as refining the solidification structure, improving surface quality of the ingot as well as reducing the macrosegregation during the solidification process of the liquid materials^[1]. The fundamental study of the moving electromagnetic field has been carried out since the early 20th century. Johnson *et al.*^[2,3] studied the influence of electromagnetic stirring on solidification structures of pure Sn and Sn-Pb alloys. The quantity of nuclei increased when the size of the equiaxed grain was decreased due to the dendrite fragmentation under the electromagnetic stirring. The forced convection driven by rotating magnetic field (RMF) accelerated the solute diffusion, decreased the temperature gradient, promoted the transition of the primary phase from coarse to equiaxed dendrite. As

a result, it reduced the macrosegregation and refined the grain size^[4,5]. The effect of inner traveling magnetic field (TMF) on the solidification microstructure of hollow tubular billet by continuous casting for Sn-3.5 wt.%Pb alloy was studied by Zhang *et al.*^[6]. The results revealed that TMF can not only arouse the circulation flow in the vertical section, but also reduce the temperature gradient of the solidification front and the cooling rate of molten metal. Moreover, TMF can refine the grains and facilitate the transition of solidification structure from the columnar grains to the equiaxed ones. The investigation of Eckert *et al.*^[7] indicated that the subsequent pulses of RMF and TMF during the solidification process could control the microstructure of Al-Si alloy and lessen the segregation defects effectively.

Many studies have been reported for the influence of RMF and TMF on the metal solidification in recent decades and the main role of RMF and TMF is refining grains and reducing the segregation, respectively^[8–10]. The spiral magnetic field (SMF), composed of RMF and TMF, is expected to overcome the known deficiencies of the conventional stir-

[†] Corresponding author. Prof., Ph.D.; Tel.: +86 411 84706183; Fax: +86 411 84706183; E-mail address: zxgwj@dlut.edu.cn (Xingguo ZHANG)

ring and improve the solidification structures more effectively. However, up to now only few experimental results have been reported on this aspect, because the solidification process of the alloy under SMF is more complex. The mechanisms of the grain refining, solute distribution and fluid flowing under SMF condition are not fully clear^[11,12]. So, it is important to clarify the solidification mechanism of the alloy under SMF condition. This work studied the segregation improvement effect and solidification structure transformation process of Sn-20 wt.%Pb alloy under SMF systematically. The research work includes three aspects: (1) to analyze the influence of the SMF on the temperature distribution of Sn-20 wt.%Pb alloy during the solidification process; (2) to study the effects of different stirring intensity on the microstructures and composition distribution of Sn-20 wt.%Pb alloy; (3) to discuss the influence of SMF on eutectic lamellar spacing of Sn-20 wt.%Pb alloy.

2. Experimental

2.1 Experimental apparatus and procedure

The Sn-20 wt.%Pb hypoeutectic alloy was chosen for solidification experiments, which was prepared with Pb (purity is 99.95%) and Sn (purity is 99.9%). A stainless steel mold with a length of 180 mm, the internal diameter of 103 mm and the wall thickness of 5 mm was used. The pouring temperature was 250 °C and the mold was preheated to 150 °C.

The cooling curves of solidification process without magnetic field and with SMF were measured by the NiCr-NiSi thermocouple with an outer diameter of 1 mm, installed along the ingot axis at a position $z=65$ mm from the bottom of the ingot. The temperature values were recorded by a multi-meter GL220 paperless recorder with 10-channel differential multiplexer card.

A new type of multifunctional spiral magnetic field stirrer was adopted to stir Sn-20 wt.%Pb alloy melt during solidification. The new stirrer can induce rotating, traveling and spiral magnetic stirring by changing winding. The schematic of the experimental apparatus is shown in Fig. 1. The frequency of SMF was fixed at 10 Hz and the magnetic intensity could be adjusted by changing exciting current (I).

2.2 Metallographic analysis

Metallographic specimens were cut from cross-section along the axial direction of the ingot from top to bottom, ground, polished and then etched in the solution of 2 mL HNO_3 +98 mL $\text{C}_2\text{H}_5\text{OH}$. The microstructures, compositions and eutectic lamellar spacing were observed and analyzed by multifunction optical microscopy (OM, MEF-3), X-ray fluorescence spectrometer (XRF, CSS-2205) and scanning electron

Fig. 1 Scheme of the experimental apparatus (1-thermocouple; 2-insulating ceramic shielding; 3-alloy melt; 4-wooden block; 5-mold; 6-electromagnetic field generator)

Fig. 2 Cooling curves of Sn-20 wt.%Pb alloy during solidification under different exciting currents (I)

microscopy (SEM, JSM-5600LV), respectively. The software package “Image-Pro Plus” was used for measurement of grain size.

3. Results and Discussion

3.1 Influence of SMF on the solidification process

Fig. 2 shows the solidification curves of center position of the Sn-20 wt.%Pb alloy with the exciting current $I=0, 50, 75, 100$ and 125 A. A steep slope change can be observed during the initial stage of the solidification as a result of the rapid heat transfer between the metal melt and mold. When the temperature is close to liquidus temperature ($T_L=205$ °C), the curve shows a slight dropping which indicates the beginning of the solidification process and the precipitating of primary phase $\beta(\text{Sn})$. Then the recalescence appearance can be seen in the cooling curves because of the release of the latent heat. When the melt temperature reduces to 184 °C, the eutectic transformation

Fig. 3 Microstructures of the Sn-20 wt.%Pb alloy without and with SMF: (a) $I=0$ A; (b) $I=50$ A; (c) $I=75$ A; (d) $I=100$ A; (e) $I=125$ A

$L \rightarrow \alpha + \beta$ occurs and the cooling curves show a distinct eutectic plateau. With the application of the SMF, the heat transfer at the solid-liquid interface increases due to the forced convection, this leads to a steeper slope of the temperature curves. And it also shows distinct fluctuations of the temperature signals arising from the turbulent melt flow for the case with electromagnetic stirring. However, with a further increase of the exciting current, the influence of electromagnetic stirring is gradually weakened because of the increased induction heating effect, which is caused by the eddy current formed in the alloy melt under SMF.

Moreover, it can be seen from the cooling curves that the cooling rates of the alloy with SMF before eutectic process are higher than that without magnetic field, and the eutectic reaction time has not changed significantly without and with magnetic field at an exciting current of 50 A, which indicates that the rise of the heat transfer before eutectic process is mainly caused by stirring. With increasing the exciting current, the induction heat will significantly affect the solidification process, and the cooling rate slows down relatively and the eutectic reaction time becomes longer than that without magnetic field.

3.2 Influence of SMF on the solidification microstructures and composition distribution

3.2.1 Morphology transition of the primary phase $\beta(\text{Sn})$

Fig. 3 shows the microstructures of Sn-20 wt.%Pb alloys at the center position of the ingot solidified without and with different SMF. Without electromagnetic stirring, the Sn-20 wt.%Pb alloy exhibits typical dendrite structure, and the primary phase $\beta(\text{Sn})$ grows into coarse dendrite arms with a size of about $216 \mu\text{m}$, as shown in Fig. 3(a). When the exciting current is 50 A, a rosette-type equiaxed structure is observed in the Sn-20 wt.%Pb alloy, as shown in

Fig. 3(b). The grain size becomes smaller, but some dendrite structures still can be observed. With the exciting current of 75 A, the dendrite arms are broken into rosette-type equiaxed dendrite and ellipsoidal grains (Fig. 3(c)), and the grains become finer with an average size of about $146 \mu\text{m}$. It can be found in Fig. 3(d) that the solidification microstructures of the alloy cooled under the exciting current of 100 A consist of the ellipsoidal grains. Fig. 3(e) shows the morphology of the structures when exciting current is 125 A, which reveals that mainly rounded spheroid grains are distributed uniformly in the matrix, but the size of grains increases to $186 \mu\text{m}$. The results indicate that under the SMF stirring with the increase of exciting current the primary phase $\beta(\text{Sn})$ shows a morphology transition: coarse dendrites $\rightarrow$ rosette equiaxed grains $\rightarrow$ ellipsoidal dendrites. The effect of the SMF on the microstructure is remarkable.

Flemings^[13] and Spencer *et al.*^[14] studied the rheological behavior of Sn-15 wt.%Pb alloy in the solidification range, and pointed qualitatively that the morphology evolved along the spectrum from initial dendrites to spheroid grains. For Sn-20 wt.%Pb alloy in this work, the evolution extent increases with increasing shear rate and amount of solidification and with decreasing cooling rate, as shown in Fig. 4. With the increase of SMF exciting current, the fluid flow becomes stronger. The grown crystal is fractured when the sum of force made by the electromagnetic and the fluid viscosity forces exceeds the fracture strength of dendrites. Moreover, the forced fluid flow gives rise to violent temperature fluctuation in the solid-liquid interface and accelerates the flow and redistribution of the intergranular solute. Then the roots of the secondary dendrite arms are remelted, followed by the formation of large quantities of spherical grains. More spherical grains occur in the microstructures as a result of the collision and friction between particles.

Fig. 4 Schematic illustration of evolution of structure during solidification with vigorous agitation: (a) initial dendritic fragment; (b) dendritic growth; (c) rosette; (d) ripened rosette; (e) spheroid

Fig. 5 Average primary arm length (λ) of Sn-20 wt.%Pb alloy vs. exciting current (I)

The relationship between the grain sizes and exciting currents is shown in Fig. 5. It can be seen that relatively lower exciting currents are more effective than the higher ones on the grain refinement of the alloy. When the exciting current is higher, the grains are more uniform and rounded but coarser. Therefore, magnetic intensity has a critical value in grain refining.

Both stirring force and heating effect contribute to the alloy melt during the electromagnetic stirring. It is believed that the stirring action plays a very important role in the refinement of solidification structure under lower exciting current. The melt flow caused by stirring makes dendrite arms remelt or fall off. Those falling crystals have not enough time to remelt due to low temperature gradient in the melt, so they will play a heterogeneous nucleation role in achieving microstructure refinement. However, under the higher exciting current, the effect of the induction heat on solidification process is greater than the stirring action, which leads to the increase of average temperature of the melt, the reduction of undercooling ΔT and cooling rate and finally results in the coarsening of the precipitation. SMF causes the change in undercooling ΔT , which directly affects the nucleation rate, derived as

$$N = c \cdot \exp\left(\frac{-\Delta G_A}{KT}\right) \cdot \exp\left(\frac{-\Delta G^*}{KT}\right) \quad (1)$$

where N is the nucleation rate, c is proportional

Fig. 6 Difference of Sn content between the top and bottom of the ingot (ΔW_{Sn}) of Sn-20 wt.%Pb alloy solidified under different exciting currents

constant, ΔG_A is atom diffusion activation energy, ΔG^* is crystal nucleus energy, K is Boltzmann constant and T is absolute temperature. Eq. (1) shows that the crystal nucleus energy ΔG^* is inversely proportional to the undercooling ΔT . The nucleation rate decreases with the reducing of undercooling, which definitely results in grains coarsening. Moreover, with the improvement of collision probability between particles, the grains aggregate more easily and the size of aggregation increases^[15].

3.2.2 Influence of SMF on the composition segregation

In order to study quantitatively the influence of SMF on the primary phase $\beta(\text{Sn})$, the differences of Sn content between the top and bottom of the ingot were measured by XRF, and the results are shown in Fig. 6. It can be seen from Fig. 6 that the difference value of the primary phases $\beta(\text{Sn})$ content along the axial direction of the ingot is about 12.5 wt.% without SMF. While under the SMF, the difference value between the top and bottom of samples gradually reduces with the increase of the exciting current. When $I=125$ A, the difference value of Sn content decreases to 0.6 wt.%, so the composition segregation is basically eliminated.

Under the conventional condition, the primary phases $\beta(\text{Sn})$ tend to accumulate at the top of sample owing to the density difference. The electromagnetic

Fig. 7 Scheme of mechanical analysis of Sn-20 wt.%Pb alloy melt under different conditions: (a) without SMF; (b) under SMF (f -buoyancy; G -gravity; F_1 - resultant force under the conventional condition; f_x -electromagnetic force along the tangent direction; f_z -electromagnetic force along the axial direction; F_2 - resultant force under the SMF)

Fig. 8 Eutectic microstructures of Sn-20 wt.%Pb alloy without and with SMF: (a) $I=0$ A; (b) $I=50$ A; (c) $I=75$ A; (d) $I=100$ A; (e) $I=125$ A (λ_e is the spacing of eutectic phase; S_α is the width of α phase; S_β is the width of β phase))

force acting on the alloy melt becomes more complex under SMF than conventional magnetic field^[12]. Schematic diagram of the force acting on a unit in the melt is presented in Fig. 7. When Sn-20 wt.%Pb hypoeutectic alloy is melted, the β (Sn) phase per unit volume is subjected to the interaction of the Lorentz force and gravity because of the influence of the imposed magnetic field. The interaction of f_x , f_z and gravity will not only cause the three-dimensional fluid flow in the melt but also improve the homogenization of temperature and solute fields. With the increase of exciting current, the Lorentz force will be strengthened. The resultant force of the Lorentz force and gravity changes and makes the motions in the vertical and horizontal planes strengthen. Thus, the distribution of the components in the melt tends to be homogenized. On the other hand, the cooling rate is higher

under the SMF condition than that without magnetic field, and the homogenized components distribution could be kept during the solidification process more easily. This suggests that the volume fraction gradient of the primary phase β (Sn) at different positions of the alloy reduces by SMF stirring.

3.3 Transition of the eutectic microstructures

Fig. 8 illustrates the eutectic structures of Sn-20 wt.%Pb alloy with different exciting currents. Without SMF, the eutectic microstructure exhibits lamellar morphology (Fig. 8(a)). However, the applying of SMF stirring brings notable changes of eutectic structures morphology. It can be seen from Fig. 8(b) to Fig. 8(e) that the lamellar is broken and rod-like eutectic appears with the increase of exciting current.

Fig. 9 Lamellar spacing of eutectic phases of Sn-20 wt.%Pb alloy *vs.* exciting current

The relationship between the lamellar spacing and exciting current is shown in Fig. 9. The eutectic spacing $\lambda_e = 2(S_\alpha + S_\beta)$ is about 1.6 μm without SMF, and λ_e is about 3.9 μm with an exciting current of 125 A. It is found that spacing coarsens with the increase of exciting current.

It is well known that fluid flow enhances the mass transportation in the solidification process. During the growth of eutectic structures, inter-phase spacing basically depends upon the diffusion of atoms ahead of the interface and the interfacial energy^[16]. It is evident that a stronger external magnetic field leads to more severe forced flow which increases the diffusion distance of atoms. The enhanced mass transport in the front of the solidifying interface produces large eutectic spacing so as to decrease area of the interface or the interfacial energy in solidification process. Moreover, the stirring can promote the liquid flow and reduce the solute concentration in front of the solid-liquid interface, which makes the undercooling ΔT decrease. According to the theoretical model of regular eutectic growth developed by Jackson-Hunt^[17], the relationship between ΔT and λ_e can be described as below: $\lambda_e \Delta T = K$, where K is a constant. The smaller ΔT is, the larger λ_e is inevitably. On the other hand, the Joule effect of the induction current will reduce the solidification rate, which causes an additional enhancement of the structural coarsening. And the higher exciting current is applied, the more Joule's heat is generated in the solidified alloys, which gives rise to the eutectic spacing coarsening.

The increase of magnetic intensity produces more intense melt turbulence and increases inter-phase spacing, and then destroys the cooperative growth way of the eutectic lamellar and even promotes the forming of rod-like grains. The solute redistribution and the temperature gradient change caused by the forced flow of the melt is the main reason for the transition from lamellar to rod-like structure. The rod-like eutectic mostly appears in the lower solidification rate and the low solidification velocity region, and the eutectic structure coarsens by the melt convection^[18].

The solidification curves shown in Fig. 2 also indicated that the eutectic reaction time of Sn-20 wt.%Pb alloy is prolonged under SMF with the increase of exciting current. It is believed that SMF reduces the growth rate of the eutectic structure. When the growth rate slows down, with increase of the melt flow rate, the lamellar spacing increases easily to the maximum lamellar spacing at $\lambda_{e,\text{max}}$ ($\lambda_{e,\text{max}}$ is the maximum limit of the stable existence of the lamellar eutectic). So, if the lamellar spacing is more than $\lambda_{e,\text{max}}$, lamellar structure will become unstable, which will lead to the morphology transition^[19]. So, the eutectic morphology transition depends on the coarsening of the eutectic lamellar spacing, that can be inferred from Fig. 8(b) and Fig. 8(e).

4. Conclusions

In this work, the effect of SMF on the solidification microstructures and composition segregation of Sn-20 wt.%Pb alloy was studied. The following conclusions can be obtained:

- (1) The macrosegregation of Sn-20 wt.%Pb alloy is reduced significantly by SMF stirring. The difference of Sn content between the top and bottom of ingot decreased from 12.5 wt.% to 0.6 wt.% with the increasing exciting current from 0 A to 125 A, and the macrosegregation of Sn-20 wt.%Pb alloy is eliminated basically;
- (2) The strong stirring caused by SMF reduces the undercooling and the temperature gradient during solidification of Sn-20 wt.%Pb alloy, and with the increase of exciting current, the primary phases morphology transformed from dendrite to rosette equiaxed grain, and finally formed the ellipsoidal or spherical crystal structures;
- (3) The eutectic lamellar spacing coarsened from 1.6 to 3.9 μm with increasing the exciting current from 0 A to 125 A during the solidification of Sn-20 wt.%Pb alloy. It also enhances the transition of eutectic structures morphology from lamellar to rod-like eutectic when the stirring strength is high enough.

Acknowledgements

This research was supported by the National Natural Science Foundation of China (No.50875031) and High-Tech Project of Bao Steel (No.10K029ECES).

REFERENCES

- [1] M. Garnier, *ISIJ Int.* **30** (1990) 1.
- [2] W.C. Johnston, G.R. Kotler and W.A. Tiller, *TMS AIME* **227** (1963) 890.
- [3] W.C. Johnston, G.R. Kotler, S. Ohara, H.V. Ashcom and W.A. Tiller, *TMS AIME* **233** (1965) 1856.
- [4] Z. Chen, C.L. Chen, X.Y. Chen, L.M. Hao, Z.Y. Hong and K.N. Wang, *Rare Met. Mater. Eng.* **36** (2007) 2011. (in Chinese)

- [5] Z. Chen, X.L. W and C.L. Chen, J. Alloys compd. **491** (2010) 395.
- [6] Q. Zhang, J.Z. Jin, T.M. Wang, T.J. Li and Q.T. Guo, Chin. J. Nonferrous Met. **17** (2007) 98. (in Chinese)
- [7] S. Eckert, P.A. Nikrityuk, D. Rabiger, B. Willers and K. Eckert, Int. J. Cast Met. Res. **22** (2009) 1.
- [8] A. Noepfel, A. Ciobanas, X.D. Wang, K. Zaidat, N. Mangelinck, O. Budenkova, A. Weiss, G. Zimmermann and Y. Fautrelle, Metall. Mater. Trans. B **41** (2010) 193.
- [9] F.D. Bai, M.H. Sha and T.J. Li, J. Alloys Compd. **509** (2011) 4835.
- [10] Q. Wang, T. Liu, K. Wang, C.J. Wang, K. Nakajima and J. Hel, ISIJ Int. **50** (2010) 1941.
- [11] Q. Zhao, C.F. Fang, X.G. Hou, Y.B. Han, N. Zhang and X.G. Zhang, Spec. Cast. Nonferrous Alloys **31** (2011) 495. (in Chinese)
- [12] N. Zhang, Q. Zhao, C.F. Fang, Y.B. Han, J.S. Guo, X.G. Hou, Q.Y. Xing and X.G. Zhang, Chin. J. Nonferrous Met. **22** (2012) 1731. (in Chinese)
- [13] M.C. Flemings, Metall. Trans. A **22** (1990) 957.
- [14] D.B. Spencer, R. Mehrabian and M.C. Flemings, Metall. Trans. **3** (1972) 1925.
- [15] Z. Chen, C.L. Chen, X.L. Wen, J.H. Zhu and W.S. Gao, Chin. Sci. Bull. **52** (2008) 2575.
- [16] H.F. Shi and W.Q. Zhang, Trans. Nonferrous Met. Soc. China **13** (2003) 1171.
- [17] K.A. Jackson and J.D. Hunt, Trans. Metall. Soc. AIME **236** (1966) 1129.
- [18] W.Q. Zhang, Y.S. Yang and Z.Q. Hu, Acta Metall. Sin. **34** (1998) 1. (in Chinese)
- [19] W. Zhang, H. Fu, Y. Yang and Z.J. Hu, Cryst. Growth **194** (1998) 26.