Research Article

Effects of High-Density Electric Current Pulse on the Undercooling of Fe-B Eutectic Alloy Melt

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The solidification microstructure of Fe-B eutectic alloy under high undercooling and high-density electric current pulse (ECP) was investigated with the technique of molten glass slag purification combined with cyclical superheating and the ECP treatment. The effects of high-density ECP on the undercooling of Fe-B eutectic alloy melt were analyzed by the DSC method. The analysis results showed that the solidification microstructure of Fe-B eutectic alloy under ECP was similar to that obtained by the high undercooling technique. The undercooling obtained under two experimental conditions was basically the same, proving that the high undercooling of the metallic melt could be realized by the ECP.

1. Introduction

High undercooling refers to a phenomenon that crystallization or solidification does not occur when a liquid metal is cooled below the liquids. Eutectic alloy is widely used as the casting alloy, and great progresses have been made in the studies in undercooling of eutectic alloy. Many experimental phenomena, such as refinement of solidification structure, reduction of the segregation, improvement of the distribution of impurity, the formation of metastable phase and amorphous phase, the growth transformation of eutectic alloy, and soluting trapping [1–5], had been found. Undercooling is an important parameter in the solidification process of metals and metal melts.

As a new type of green solidification control technology, the ECP, which conducted in alloys through electrodes directly contacting the melt, can improve the heterogeneous nucleation rate of liquid metal or semi-liquid metal, promote the solute redistribution, and refine the solidification microstructure [6–9]. Many research studies have been carried out to investigate the effect of ECPs on the solidification of pure metals [10], eutectic alloys [11–13], and solid solution alloys [14] in the past decades. The mechanisms proposed to understand the

ECP-induced grain refinement include the heterogeneous nucleation mechanism that the nucleation rate is promoted owing to the raised undercooling [7, 15, 16], the skin effect, the dendrite fragmentation mechanism caused by Joule heating [17], Lorenz force [11, 15], and the crystal rain [10].

The effect of ECP on the undercooling of the melt had been confirmed by many scholars [7, 16, 18]. Previous studies on the undercooling of ECP were performed under the condition of large voltage [19, 20], but the undercooling of ECP under low voltage was not studied. In the study, with Fe₈₃B₁₇ eutectic alloy as the research object, by means of molten glass slag purification and cyclical superheating, according to the high-density ECP method, the influences of the undercooling on solidification microstructure of metallic melt were studied. The undercooling variation of alloy melt was detected by DSC method and explained theoretically. The ECP as a novel solidification technology can be applied in engineering practice.

2. Experimental Methods

2.1. Selection of Alloy. The Fe-B alloy is a kind of good soft magnetic material possessing excellent glass forming ability

FIGURE 1: Schematic drawing of experimental setup.

FIGURE 2: Schematic sketch of the crucible and the electrodes.

in iron-rich end and can realize the greater undercooling [21]. With the eutectic alloy $\text{Fe}_{83}\text{B}_{17}$ as the research object, the solidification evolutions and the undercooling were discussed respectively under the conditions of high undercooling condition and high-density ECP.

2.2. Preparation of Undercooled FeB Alloy. The alloy was melted and purified utilizing the high frequency induction heating device (Fe, B purity was above 99.99%). The output power was 35 kW, and the working frequency was 20~50 kHz. The diameter of induction heating copper coil was 6 mm. The molten glass slag purification and cyclical superheating were used to purify molten metals with B_2O_3 purification. In the experiment, the temperature of alloy melt measured by infrared thermometer was output to the connected computer through the active RS485 converter, and the measured data were stored.

2.3. Preparation of FeB Alloy under High-Density ECP. Figure 1 is the schematic drawing of experimental setup. The experimental apparatus consist of the customized pulse power

supply, vacuum system, high frequency induction heating device, and temperature measuring system. Figure 2 is schematic sketch of the crucible and the electrodes. The pulse power provides a frequency from 0 to 50 Hz, and the electric voltage ranges from 0 to 100 V. First, the master alloy was prepared according to the proportion of eutectic components. The $\text{Fe}_{83}\text{B}_{17}$ master alloy was cut into the specimens with the size of 15 mm × 10 mm × 15 mm, put into a customized boron nitride crucible with a cylindrical cavity, and covered with B_2O_3 . The boron nitride conductive electrode (chemical composition: BN + TiB_2 + AlN), connected to the crucible with the Mo electrodes, was used in the ECP treatment with thermal conductivity 100 W/mK to reducing the heterogeneous nucleation. The samples were melted in the crucible and heated to 1500°C in N_2 atmosphere. The ECP treatment was performed when the end face of boron nitride electrode contacted the molten metal horizontally according to the parameters of pulse voltage 20 V, pulse frequency 30 Hz, pulse width 20 μs , peak current 600 A, and pulse current duration of 30 s. The cooling rate used was 10 K/min⁻¹, which was held approximately constant throughout the solidification. After cooling, the 10 mm piece test sample was cut from solidified

FIGURE 3: Solidification structure of the sample under the high undercooling conditions and ECP treatment. (a)–(c) Solidification structure of the sample under the high undercooling. (d)–(f) Solidification structure of the sample treated with the ECP.

specimen and then polished for metallographic examination. The etching reagent used to reveal the macrostructure was nitric acid. The microstructures were observed by SEM along the center line of the specimen.

3. Results and Discussion

Figures 3(a)–3(c) show the hypereutectic microstructure of $\text{Fe}_{83}\text{B}_{17}$ eutectic alloy under high undercooling conditions. According to the Fe-B equilibrium phase diagram, X-ray powder diffraction spectrum (Figure 4) and the previous studies indicated the primary phase of $\alpha\text{-Fe}$ (shown as dark grey phase) and the irregular eutectic of $\alpha\text{-Fe} + \beta(\text{Fe}_2\text{B})$ (shown as light grey phase) forming the noncontinuous network distributed in the grain boundary between $\alpha\text{-Fe}$. Figures 3(d)–3(f) show the microstructures of the intermediate alloy treated

with the ECP. Similar grain sizes and microstructures were obtained by ECP compared with the ones by high undercooled technology. As shown in Figures 3(c) and 3(f), the volume fraction of the eutectic phase $\alpha\text{-Fe} + \beta(\text{Fe}_2\text{B})$ is less decreased. Figure 5 shows that the lamellar regular eutectic structure of original FeB sample prepared without undercooling and ECP is different from the hypereutectic microstructure of the sample under the high undercooling conditions and ECP treatment.

It has been proposed that the maximum undercooling can be raised to accelerate the heterogeneous nucleation rate. Solidification behavior under high undercooling is an extremely nonequilibrium solidification behavior, which produces the primary phase and the eutectic phase. It was proved by many experiments that the binary eutectic alloy with a facet became an irregular eutectic microstructure when the undercooling reached or exceeded a critical value [22, 23]. With the heating

FIGURE 4: X-ray powder diffraction spectrum of $\text{Fe}_{83}\text{B}_{17}$ eutectic alloy.

FIGURE 5: Solidification structure of $\text{Fe}_{87}\text{B}_{13}$ sample prepared without undercooling and ECP treatment.

and cooling rate of 10 K/min^{-1} , the DSC curve of undercooled melt $\text{Fe}_{83}\text{B}_{17}$ (Figure 6) shows that the undercooling degree of melt alloy is 95 K. In the solidification process, there are two recalescence peaks because the latent heat release rate of phase change is much higher than heat dissipation. The solidification process is divided into two stages: the precipitation of $\alpha\text{-Fe}$ as the primary phase and the eutectic reaction of $L \rightarrow \alpha\text{-Fe} + \beta(\text{Fe}_2\text{B})$ of the residual liquid phase. The two recalescence peaks on the temperature curve correspond to the solidification of two phases. In Figure 3(c), the α -phase in $\alpha\text{-Fe} + \beta(\text{Fe}_2\text{B})$ between the grain boundaries grows and is attached to the primary phase $\alpha\text{-Fe}$ into one because the solute trapping caused by nonuniform solute atom diffusion leads to the increase of the content of $\alpha\text{-Fe}$ in the residual liquid phase and the inhomogeneity of the eutectic structure.

Compared with the solidification process under undercooling conditions, the morphology affected by ECP is similar but the structure is more dense and uniform. In the

FIGURE 6: DTA traces for $\text{Fe}_{83}\text{B}_{17}$ heating (curve a) and cooling (curve b) runs were performed at a rate of $10 \text{ K}\cdot\text{min}^{-1}$.

FIGURE 7: Typical cooling curves of $\text{Fe}_{83}\text{B}_{17}$ in nonequilibrium solidification conditions.

metal melt, $\alpha\text{-Fe}$ primary phase recalesces rapidly after nucleation and growth. Due to the thermal shock of recalescence, the dendrite of $\alpha\text{-Fe}$ fusing and equiaxed grains were formed. The above-mentioned dendritic fragments were ripened and surrounded by eutectic phase $\alpha\text{-Fe} + \beta(\text{Fe}_2\text{B})$ precipitated in residual melts, thus forming the irregular eutectic. The cooling curve (Figure 7) shows two recalescence peaks. The undercooling of alloy melt under high-density ECP is 105 K ($\Delta T = T_L - T_N$, $T_L = 1173^\circ\text{C}$, $T_N = 1068^\circ\text{C}$), which was basically the same to that obtained under high undercooling conditions.

4. Mechanism Explanation

The above results showed that similar solidification microstructures were obtained under high-density ECP and high undercooling technique. The cooling curve indicated that the same undercooling could be obtained by high-density pulse current and high undercooling technique. In this paper, it is proved that the melt undercooling can be

improved by high-density ECP. The mechanism is interpreted as follows:

- (1) According to the atomic cluster theory proposed by Wang [24], the ECP as a kind of energy access cracked large clusters of atoms into smaller clusters. Therefore, the number of small clusters in the same volume of melts increased to strengthen the surface energy of clusters. As a result, higher undercooling was required to provide the nucleation energy for increasing the size of the smaller clusters to the critical nucleation radius. Therefore, the ECP increased the molten metal undercooling. The clusters' sizes decreased, and a larger number of clusters were required for meeting the critical nucleation radius to gain more grains. In this way, the alloy was refined.
- (2) The dendrite fragmentation was another origin for grain refinement under the application of ECP. It is possible that the forced flow can trigger the remelting of high-order dendrite arms to detach from the dendrite trunk due to the forced flow-induced solute fluctuation [13]. The detached dendrite arms would be subsequently transported out as new potential nuclei sites under the influence of forced flow. Since the size of dendrite arms is really less than the maternal dendrite, numerous tiny grains mixed with some larger grains are consequently formed.
- (3) The skin effect associated with the current pulses raised the temperature of the molten metal near the mold wall, thereby suppressing the heterogeneous nucleation. Then, the undercooling increased. The skin depth δ of a current pulse is given by

$$\delta = \sqrt{\frac{\rho}{\pi \mu f}}, \quad (1)$$

where ρ is resistivity of the molten alloy; μ is permeability; and f is the electric pulse current wave frequency [7].

- (4) The crystal grew in the crucible, so the performance and quality of the crucible played an important role in the quality of crystal growth. The thermal conductivity, thermal expansion coefficient, wetting angle, surface finish, chemical stability, and purity of the crucible determined the structures and properties of the crystal. The crucible used in this experiment was a kind of high-purity boron nitride material, which was better than the traditional quartz crucible. The crucible used in this experiment showed the characteristics of small thermal expansion coefficient, high thermal conductivity, obvious anisotropy, and large wetting angle and can eliminate more heterogeneous nucleation and obtain large undercooling degree of the melt.

5. Conclusions

- (1) The similar grain sizes and the same microstructures of Fe₈₃B₁₇ alloy were obtained by high-density ECP and

high undercooled technology. The α -Fe is the primary phase, and the irregular eutectic α -Fe + β (Fe₂B) formed the noncontinuous network distributed in the grain boundary between α -Fe. The eutectic phase of α -Fe + β (Fe₂B) became finer and the volume fraction was decreased.

- (2) DSC method indicated that the similar undercooling of alloy melt was obtained under high-density ECP and high undercooling. It was proved that the high undercooling applied on the metallic melt could be realized by the ECP, thus laying the foundation for large-scale production practice.
- (3) The atom cluster cracking, the dendrite fragmentation, the skin effect, and the crucible material are responsible for the melt undercooling increased by high-density ECP.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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