Available online at www.sciencedirect.com
ScienceDirect
Trans. Nonferrous Met. Soc. China 23(2013) 92−97
Transactions of Nonferrous Metals Society of China
www.tnmsc.cn

Effect of electrodes and thermal insulators on grain refinement by electric current pulse

Zhen-xing YIN, Dong LIANG, Yu-e CHEN, Yu-feng CHENG, Qi-jie ZHAI

Shanghai Key Laboratory of Modern Metallurgy & Materials Processing, Shanghai University, Shanghai 200072, China

Received 17 November 2011; accepted 20 April 2012

Abstract: The application of electric current pulse (ECP) to a solidification process refers to the immersion of electrodes into the liquid metal and the employment of thermal insulators on the upper surface of metal. In order to ascertain the effects of these two factors on the structure refinement by the ECP technique, three groups of experiments were performed with different types of electrodes or various thermal insulators. By the comparison between solidification structures under different conditions, it is followed that the electrode and the thermal insulator have an obvious influence on the grain refinement under an applied ECP, and further analysis demonstrates that the thermal conditions of the liquid surface play a vital role in the modification of solidification structure. Also, the results support the viewpoint that most of the equiaxed grains originate from the liquid surface subjected to an ECP.

Key words: solidification structure; grain refinement; electric current pulse; electrode; thermal insulator

1 Introduction

It is well known that refining cast structure can improve the mechanical properties of metals and alloys. A variety of grain refinement technologies have been developed to obtain this purpose. The use of electromagnetic fields to refine cast structure has attracted much attention owing to their many merits such as easy-to-control, good adaptability and high effectiveness. A number of researchers [1−8] reported that the application of an electric current pulse (ECP) during the solidification of metals and alloys has an obvious influence on the cast structure, especially on the grain refinement. Meanwhile, several refinement mechanisms [9−12] were proposed in an attempt to clarify the formation of equiaxed zone under the influence of ECP. WANG et al [13] proved that direct current could suppress the growth of the dendrite in Sn−Bi alloy and promote the nucleation ahead of solid/liquid interface by means of in-situ observation. CHANG et al [14] analyzed the current-induced change in undercooling on the basis of interface stability theory. Although these works are beneficial to understanding of the structural refinement, the origin of equiaxed grains under the ECP is still ambiguous. Through delicate experiments and distinct observations, LI et al [15,16] definitely pointed out that when the ECP is introduced to the liquid metal with parallel electrodes at the top of ingot, a large number of free grains will be induced by the ECP mostly near the upper surface, drifting away from the top to the bottom, eventually leading to structural refinement. This mechanism is termed as “crystal rain”. The liquid upper surface is therefore of great importance for the ECP technique because it is the right place where most of equiaxed crystals are created and detached. Furthermore, it can be speculated that all the factors that are able to markedly alter the thermal condition of the upper surface may affect the grain refinement of an applied ECP. However, little literature focused on this problem.

For a foundry process, the application of an ECP during the solidification requires the immersion of the electrodes into the liquid metal and the employment of thermal insulators on the top of the ingot, which maybe influence the grain refinement of ECP by altering the thermal conditions of upper surface of the ingot. In order to clarify the roles of the electrode and the insulator in the grain refinement, in this work, three groups of experiments were conducted and the results were

Foundation item: Project (2009AA03Z110) supported by the National High Technology Research and Development Program of China; Project (2011CB012902) supported by the National Basic Research Program of China
Corresponding author: Qi-jie ZHAI; Tel: +86-21-56331218; E-mail: qjzhai@shu.edu.cn
DOI: 10.1016/S1003-6326(13)62434-6

discussed in terms of the crystal rain mechanism.

2 Experimental

The experimental setup used in this work consists of a pulse generator with the capacitance of 1500 μF, an electric resistance furnace, and an oscilloscope, as shown in Fig. 1. The major parameters of an ECP include the peak value of electric current and the discharging frequency of pulse. A typical pulse waveform is illustrated in Fig. 2. A whole treatment period can be subdivided into two stages, i.e., discharging stage (I) and charging stage (II).

As the starting material, commercial pure aluminum (99.7%, mass fraction) was melted in the resistance furnace and heated to 1023 K (90 K of superheating). After deslagging, the liquid aluminum was poured into the prepared mold, then the ECP was immediately applied to the melt with the parallel electrodes, as shown in Fig. 1(a). The molds used in the former two groups of experiments were permanent molds that were all preheated to 473 K before casting. Sand molds were used in the third group of experiments with an inner dimension of *d*60 mm×140 mm, being held at ambient temperature before casting.

The as-received specimens were sectioned along the center line, ground and finally etched by a mixed acid (V(HCl):V(HNO_3):V(Water)=24:27:39) to reveal the solidification structure. The linear intercept method was used to measure the grain size and the area fraction of equiaxed zone was evaluated with the aid of Image Pro Plus 6.0.

3 Results

3.1 Effect of electrodes on grain refinement under ECP

When an electric pulse is imposed to the melt through the electrodes, Joule heating effect due to the electric resistance is significant for the thermal condition of upper surface of the melt. Moreover, the Joule heat can vary with changes in dimensions, materials and/or structure of the electrode. Based on the considerations, two groups of experiments were conducted.

The macrostructures of the ingots in the first group are shown in Fig. 3, and two types of transverse sections of electrodes were employed, namely 15 mm×2 mm for Fig. 3(b) and 30 mm×2 mm for Fig. 3(c). In the case of small section of electrode (Fig. 3(b)), the effect of ECP on the structural refinement is almost negligible. When the section of the electrode increases from 15 mm×2 mm to 30 mm×2 mm, the solidification structure is modified markedly, as shown in Fig. 3(c). The columnar crystals become smaller with an average width of 1 mm, meanwhile increase rapidly in number. There is an equiaxed zone at the bottom with an average size of 0.9 mm and an area fraction of 19.1%. In addition, two lines of fine equiaxed grains, appearing beside the needle-like columnar crystals, as shown in Fig. 3(c), can be supposed to be frozen free crystals drifting from the upper surface.

Three different kinds of electrodes, including the carbon steel electrode, the water-cooled carbon steel electrode, and the pure copper electrode, were employed in the second group. The cross-section of all these electrodes is 30 mm×2 mm. The cast structures are shown in Fig. 4. The differences between solidification structures of these four ingots indicate that the electrode has an obvious influence on the structural refinement under the same parameters of pulse. In the case of Fig. 4(b), the solidification structure can be divided into three different zones, including a chilling zone close to the mold wall, a center zone with coarse equiaxed grain, and a bottom zone with fine equiaxed grains. Especially in the bottom zone, the average size of equiaxed grains reaches about 0.8 mm, and the area fraction of the equiaxed zone is 21.2%. Also, two lines of fine equiaxed grains can be observed close to the two sides of the cross-section, as denoted by arrows in Fig. 4(b), which might also be frozen during the sedimentation of free

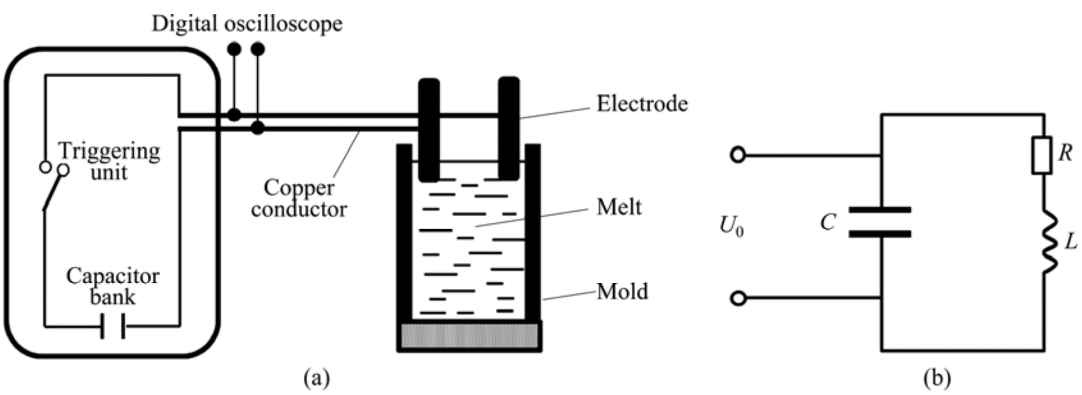

Fig. 1 Schematic diagram of experimental setup (a) and equivalent electric circuit (b) of ECP treatment

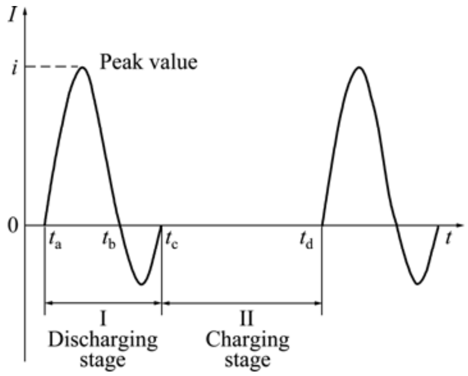

Fig. 2 Illustration of waveform of pulse and one whole treatment period (one whole treatment period can be subdivided into two stages, i.e., discharging stage t_a−t_c and charging stage t_c−t_d, and the latter is about 100 times of the former)

crystals from the top. However, other two ingots exhibit that solidification structures are degenerated seriously compared with the ingot shown in Fig. 4(b), as shown in Figs. 4(c) and (d). That is, the copper electrode or water-cooled electrode leads to a decrease in the equiaxed zone and an increase in the grain size.

3.2 Effect of thermal insulator on grain refinement under ECP

For a foundry process, the riser of the ingot, namely the upper surface, is usually covered with the thermal insulator. Meanwhile, vast Joule heat can be produced near the upper surface of the melt due to the passage of the electric pulse. The third group of experiments was carried out to examine these effects. Experimental

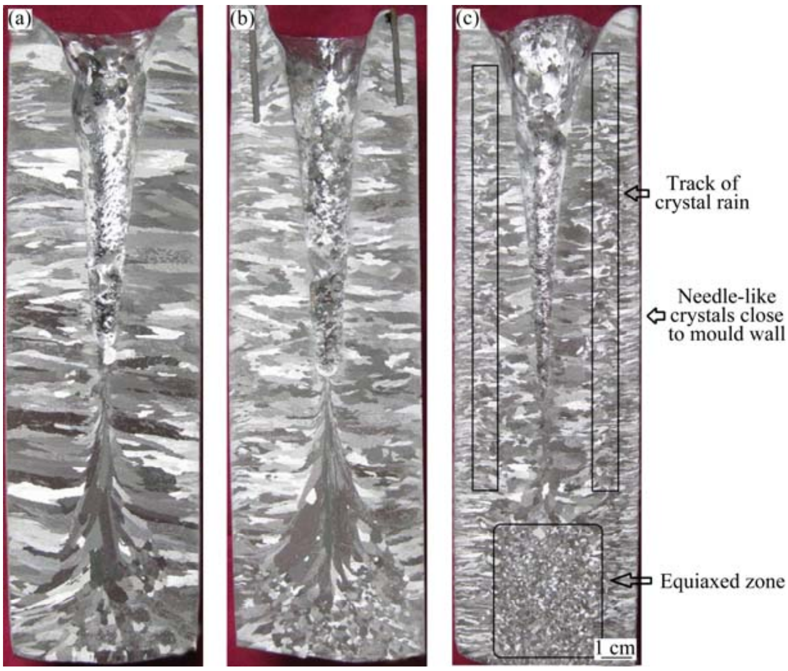

Fig. 3 Macrostructures of ingots (210 mm in height) without or with ECP ($100k_I$ A, $100h_I$ Hz, k_I is current coefficient, h_I is frequency coefficient): (a) Without ECP; (b) With ECP and 15 mm×2 mm electrodes; (c) With ECP and 30 mm×2 mm electrodes

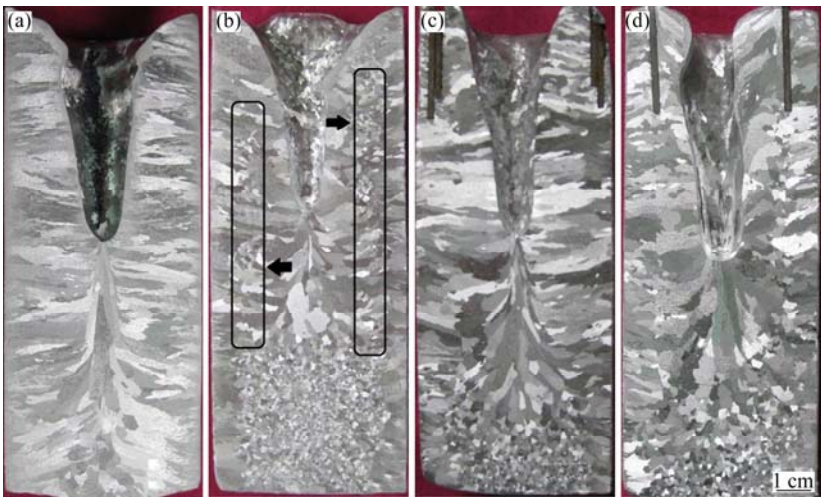

Fig. 4 Macrostructures of ingots (140 mm in height) without or with ECP ($175k_I$ A, $50h_I$ Hz): (a) Without ECP; (b) With ECP and carbon steel electrode; (c) With ECP and water-cooled carbon steel electrode; (d) With ECP and pure copper electrode

conditions are shown in Table 1 and the as-cast structures are shown in Fig. 5.

In contrast to overall coarse grains (Fig. 5(a)) in the absence of the ECP, the area fraction of the equiaxed zone reaches about 47% and the average size of equiaxed grains is about 1 mm in the presence of the ECP, as shown in Fig. 5(b). The results sufficiently demonstrate the grain refinement effect of the ECP. In order to retard the solidification of the melt, various thermal insulators were exerted on the upper surface of the ingots. The fly ash just reduced the thermal conduction while the rice-shell not only did that but also supplied much heat via oxidation during the solidification. Therefore, the insulation effect of the rice-shell is better than that of the fly ash. By comparing solidification structures of Figs. 5(b), (c) and (d), it can be seen that the equiaxed zones and the shrinkage cavities become smaller in the sequence of ingots 5(b), (c) and (d), meanwhile the center grains are coarsened. The same situation also occurs under the ECP of relatively low parameters, as shown in Figs. 5(e) and (f).

Table 1 Experimental conditions and area fraction of equiaxed zone

Ingot in Fig. 5	Thermal insulator	Peak current/A	Frequency/Hz	Area fraction of equiaxed zone/%
5(a)	No	–	–	–
5(b)	No	$250k_{\mathrm{I}}$	$100h_{\mathrm{I}}$	47
5(c)	Fly ash	$250k_{\mathrm{I}}$	$100h_{\mathrm{I}}$	35
5(d)	Rice-shell	$250k_{\mathrm{I}}$	$100h_{\mathrm{I}}$	26
5(e)	No	$200k_{\mathrm{I}}$	$100h_{\mathrm{I}}$	29
5(f)	Rice-shell	$200k_{\mathrm{I}}$	$100h_{\mathrm{I}}$	14

4 Discussion

As mentioned above, “crystal rain” mechanism [15,16] points out that with parallel electrodes at the top of the ingot, most equiaxed grains originate from the upper surface. A next problem is how the passage of a pulse through the upper surface gives a rise in the multiplication and the detachment of free crystals. Based

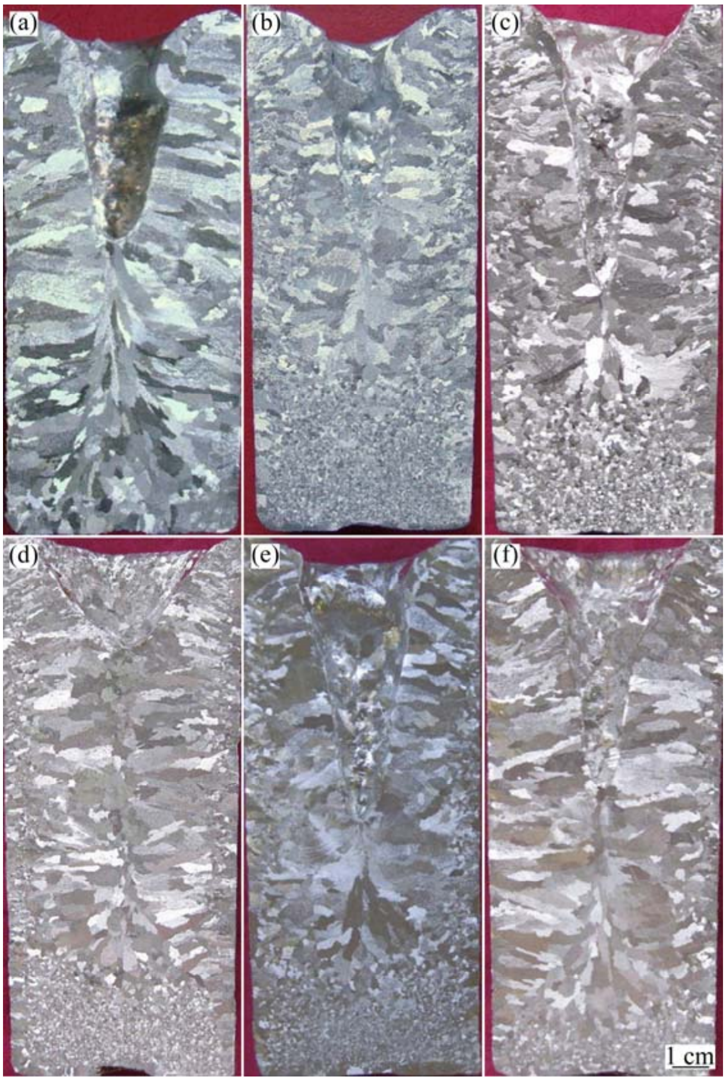

Fig. 5 Macrostructures of ingots without or with ECP and under different surface conditions

on the results and the analyses of published literatures [13,14,17], it can be supposed that when the electric current is passed through a mixture of solid and liquid phases, the crowding effect of current occurs due to the difference between the resistivity of two phases, e.g., in the case of pure aluminum, the resistivity of the liquid metal is about 10 times as large as that of the solid, resulting in the highest current density at the solid/liquid interface [18] and more Joule heat, so the growth of crystals is markedly suppressed owing to the higher temperature gradient ahead of the solid/liquid interface, and more crystals form near the upper surface. Furthermore, since the electric pulse varies greatly with time, a thermal perturbation can be produced on the upper surface, which may allow the existing crystals to partly melt off and separate from the upper surface. Thus, a large number of free crystals can be created and drift away from the top to the bottom under the driving of the fluid flow, the density difference of two phases and the electromagnetic force induced by ECP, eventually these free crystals pile up at the bottom to form an equiaxed zone. As a result, two lines of fine equiaxed grains close to the two sides can be observed, as shown in Figs. 3(c) and 4(b). Note that the works in Refs. [5,15] confirm that the ECP process has no inoculant effect, and some undercooling is also required for the nucleation under the ECP. Hence, the thermal fluctuation is restricted by the required undercooling. If the Joule heat induced by the electric pulse cannot be transferred properly, the upper surface will be heated to reduce the undercooling due to the thermal accumulation, and hence the nucleation of crystal will be depressed. So, the effect of electric pulse on the grain refinement has two aspects: one is to suppress the growth of the crystal and promote the solidified crystals to melt off and separate from the upper surface, the other is to reduce the undercooling and unfavorable to the nucleation due to the accumulation of Joule heat. At the other extreme, if the upper surface solidifies too quickly, the electric pulse will have no effect due to its passage through the solid phase [16]. Therefore, it can be concluded that the thermal condition and solidification process of the upper surface is of great significance for the structural refinement by the ECP.

For the former two groups of experiments, the Joule heat effect of the electric pulse through the electrodes plays a vital role in the refinement of the solidification structure. According to Ohm law $R=\rho L/S$, Joule law $Q=I^2Rt$ [19], the Joule heat is determined by the cross-section of electrode S and the electric resistivity of electrode ρ. When the electrode of small transverse section is employed, vast Joule heat can be produced due to large resistivity and small heat transfer area, resulting in the decrease in the undercooling of the upper surface and suppressing the nucleation. Thus, the equiaxed grain does not appear in this situation, as shown in Fig. 3(b). On the other hand, for Figs. 4(c) and (d), since the pure copper or water-cooled electrode is applied to reducing the Joule heat, leading to a shorter solidification time, the ECP also has no effect on the refinement. Consequently, the thermal condition of the melt surface can determine the effective treatment time of the ECP and hence influence the refinement of the cast structure. Only when appropriate electrodes are employed, such as Figs. 3(c) and 4(d), the resultant thermal condition of the melt surface is favorable for structural refinement.

For the third group, under the higher pulse parameters, a larger equiaxed zone is obtained (Fig. 5(b)); however, when the upper surface is covered with the thermal insulator such as fly ash and rice-shell, the Joule heat cannot transfer properly, resulting in a decrease in the undercooling of upper surface and a degeneration of the structural refinement. Furthermore, since the thermal insulation effect of rice-shell is better than that of fly ash, the refinement effect is worse owing to the smaller undercooling attained. As a result, the equiaxed zone decreases in the sequence of ingots 5(b), 5(c) and 5(d) although the shrinkage cavity decreases in the same order. Even when the peak value of pulse decreases from $250k_{\mathrm{I}}$ A to $200k_{\mathrm{I}}$ A, the thermal insulator also causes the structural refinement to degenerate, as shown in Figs. 5(e) and (f). This proves that the thermal condition of upper surface plays a vital role in the structural refinement of the ECP.

5 Conclusions

1) The electrode has a strong influence on the grain refinement of the ECP due to changes in the thermal condition of the upper surface aroused by the heating or chilling effect of the electrode.

2) The thermal insulator on the upper surface is also of great significance for the grain refinement of the ECP. When the thermal insulator is exerted on the melt surface, the equiaxed zone becomes smaller compared with that of the specimen without thermal insulator.

3) Creating an appropriate thermal condition of the upper surface can optimize the grain refinement of the ECP, which indicates that there is an essential relationship between the mechanism of the grain refinement of the ECP and the thermal conditions of the upper surface. This further proves that major equiaxed grains originate from the upper surface.

References

[1] NAKADA M, SHIOHARA Y, FLEMINGS M C. Modification of solidification structures by pulse electric discharging [J]. ISIJ International, 1990, 30(1): 27−33.

[2] QIN R S, ZHOU B L. Effect of electric current pulses on grain size

in castings [J]. International Journal of Non-equilibrium Processing, 1998, 11(1): 77−86.
[3] MISHRA R S, MUKHERJEE A K. Electric pulse assisted rapid consolidation of ultrafine grained alumina matrix composites [J]. Materials Science and Engineering A, 2000, 287: 178−182.
[4] ZHAO Z L, LIU B, ZHANG H N, ZHANG R, LIU L. Electro-pulse modification of ZL 102 and its compound effect with borax refining [J]. Foundry, 2002, 51(5): 283−285. (in Chinese)
[5] FAN J H, CHEN Y, LI R X, ZHAI Q J. Effect of pulse electric current on solidification structure of austenitic stainless steel [J]. Journal of Iron and Steel Research International, 2004, 11(6): 37−39.
[6] ZHAO Z L, WANG J L, LIU L. Grain refinement by pulse electric discharging and undercooling mechanism [J]. Materials and Manufacturing Processes, 2011, 26: 249−254.
[7] WANG J Z, CHEN Q F, CAO L Y, ZHAO L C. Effect of electropulse modification parameters on ingot macrostructure of Cu−Al−Ni shape memory alloys [J]. Transactions of Nonferrous Metals Society of China, 2002, 12(3): 400−403.
[8] HE S X, WANG J, ZHOU Y H. Effect of high density pulse electric current on the solidification structure of low temperature melt of A356 aluminium alloy [J]. Acta Metallurgica Sinica, 2002, 38(5): 479−482. (in Chinese)
[9] BARNAK J P, SPRECHER A F, CONRAD H. Colony (grain) size reduction in eutectic Pb−Sn castings by electroplusing [J]. Scripta Metallurgica et Materialia, 1995, 32(6): 879−884.
[10] WANG J Z, QI J G, DU H L, ZHANG Z B. Heredity of aluminum melt caused by electric pulse modification (I) [J]. Journal of Iron and Steel Research International, 2007, 14(4): 75−78.
[11] CORIELL S R, MCFADDEN G B, BILLIA B. Electrical pulsing during directional solidification: Analysis of transients by Laplace transform [J]. Journal of Crystal Growth, 2000, 216(1): 495−500.
[12] LIAO X L, ZHAI Q J, LUO J, CHEN W J, GONG Y Y. Refining mechanism of the electric current pulse on the solidification structure of pure aluminum [J]. Acta Materialia, 2007, 55: 3103−3109.
[13] WANG T M, XU J J, XIAO T Q, LI J, LI T J, CAO Z Q. Evolution of dendrite morphology of a binary alloy under an applied electric current: An in-situ observation [J]. Physical Review E, 2010, 81: 042601.
[14] CHANG G W, YUAN J P, WAN G Z D, WU C J, WANG X H, HU H Q. Effect of electric current on stability of solidification interface morphology [J]. Transactions of Nonferrous Metals Society of China 2000, 10(4): 445−448.
[15] LI J, MA J H, GAO Y L, ZHAI Q J. Research on solidification structure refinement of pure aluminum by electric current pulse with parallel electrodes [J]. Materials Science and Engineering A, 2008, 490: 452−456.
[16] LI J. Mechanism and application of metal solidification structure refinement by electric current pulse [D]. Shanghai: Shanghai University, 2009: 39−43. (in Chinese)
[17] ZHAO K H, CHEN M X. New concept physics—electromagnetism [M]. 2nd ed. Beijing: Higher Education Press, 2006: 74−77. (in Chinese)
[18] HUANG J, WANG B F, ZHAO L P, LI J C. Numerical simulation of magnetic field and flow field in aluminum melt with electromagnetic Stirring [J]. Special Casting & Nonferrous Alloys, 2008(2): 119−121. (in Chinese)
[19] POLLACK G L, STUMP D R. Electromagnetism: Photocopied edition [M]. Beijing: Higher Education Press, 2005: 228−239.

电极和保温剂对脉冲电流细化晶粒的影响

尹振兴，梁 冬，陈玉娥，程誉锋，翟启杰

上海大学 上海市现代冶金与材料制备实验室，上海 200072

摘 要：在实际的铸造过程中应用脉冲电流要求电极插入熔体中并在熔体的上面覆盖保温剂。为了探明这两个因素对脉冲电流细化组织的影响，采用不同电极或不同保温剂进行了 3 组实验。通过比较不同条件下得到的凝固组织，发现在脉冲电流作用下电极和保温剂都对凝固组织的细化有明显的影响。进一步的分析表明，液面在凝固组织的细化中起到了关键性的作用，而且结果也支持了在脉冲电流作用下等轴晶主要来自液面的观点。

关键词：凝固组织；晶粒细化；脉冲电流；电极；保温剂

(Edited by Wei-ping CHEN)