

Article

The Role of Electric Current-Associated Free Energy and Forced Convection on Grain Refinement in Pure Aluminum under Electropulsing

Ning Li ^{1,†}, Limin Zhang ^{1,*,†}, Rong Zhang ¹, Pengfei Yin ² , Hongjing Wu ¹, Kaikai Song ³  and Hui Xing ^{1,*,†}

¹ Shaanxi Key Laboratory of Condensed Matter Structures and Properties and Key Laboratory of Space Applied Physics and Chemistry, Ministry of Education, School of Science, Northwestern Polytechnical University, 127 West Youyi Road, Xi'an 710072, China; lining142433@163.com (N.L.); xbw101@mail.nwpu.edu.cn (R.Z.); wuhongjing@nwpu.edu.cn (H.W.)

² College of Science, Sichuan Agricultural University, Ya'an 625014, China; yinpengfei@sicau.edu.cn

³ School of Mechanical, Electrical & Information Engineering, Shandong University (Weihai), 180 Wenhua Xilu, Weihai 264209, China; songkaikai8297@gmail.com

* Correspondence: liminzhang@nwpu.edu.cn (L.Z.); huixing@nwpu.edu.cn (H.X.)

† These authors contributed equally to this work.

Received: 18 October 2019; Accepted: 20 November 2019; Published: 22 November 2019



Abstract: An experimental study with respect to the effect of an alternating electropulsing on grain refinement in pure aluminum was reported. The macrostructural observation with the mold preheated to different temperature and embedded the metal mesh indicated that the change of electric current-associated free energy related with the position of crystal nuclei (ΔG_{em}) and forced convection dominated the generation of fine equiaxed grains (FEG). Under electropulsing with 480 A, ΔG_{em} induced the dissociation of crystal nuclei from the upper interface of the electrode and the melt, leading to the generation of FEG. For a larger current intensity, FEG originated from the dissociation of crystal nuclei on the side wall besides the upper interface due to ΔG_{em} and the forced convection. Furthermore, the model coupling the dissociation of crystal nuclei and dendrite fragmentation due to the forced convection and the dissociation of crystal nuclei due to ΔG_{em} was presented to explain the formation mechanism of FEG in pure aluminum under electropulsing.

Keywords: alternating electropulsing; pure aluminum; solidification; fine equiaxed grains

1. Introduction

It is well known that replacing the large grains in castings by the fine equiaxed grains (FEG) can significantly enhance the mechanical properties of cast alloys. Over the past few decades, the different types of physical fields including the electric field [1–7], ultrasonic vibration [8–12] and electromagnetic field [13–16], have been successfully applied during solidification process of alloys for the improvement of the solidification structure. The application of electropulsing during solidification, as a promising technique for achieving fine grains, has attracted considerable attention. Consequently, extensive experimental and theoretical studies have been performed to explain the formation mechanism of FEG with electropulsing treatment [17–19]. Despite many hypotheses were proposed, no consensus has yet emerged.

To date, it is generally accepted that FEG is mainly achieved during the nucleation stage [3] and electromagnetic convection is essential for its generation in solidified alloys [20–22]. D. Rübiger et al. [3] proposed that the formation of FEG in solidified Al-Si alloy resulted solely from the global convection driven by Lorentz force under electric current. Y.H. Zhang et al. [21] further

confirmed that forced convection could induce the redistribution of temperature and concentration field in the mushy zone and then accelerate dendrite fragmentation based on the improved experiments and numerical simulation. From casting experimental investigation of the relevance of the forced convection to the formation of FEG in pure aluminum under an alternating electropulsing, our previous work [20] pointed out that the forced convection gave rise to the columnar fragmentation and the dissociation of crystal nuclei on the side wall, resulting in the formation of FEG. Additionally, Y. H. Zhang et al. [5] indicated that columnar fragmentation was ascribed to the promotion of solute driven remelting of dendrite arms in low-purity aluminum due to the obvious forced convection. However, the forced convection cannot be used to explain all the structural observations accompanying grain refinement. For example, since the matrix liquid alloy has a higher electrical resistivity compared to the precipitated (Cu, Sn)-rich phase droplets, electropulsing leads to a reduction in the energy barrier to nucleation of the precipitated phase and then promotes the formation of a fine dispersed microstructure in solidified Cu-Bi-Sn immiscible alloys due to electric current-induced free energy minimization [23].

Electric current, passing through a conductor, induces an additional term G_e for the system free energy compared with that without electric current [24,25]. Experiments and theoretical analysis have proved that a significant effect of electric current-associated free energy change (ΔG_e) on the microstructural improvement in materials, such as segregation of inclusions or solute precipitates in alloys [24,26], solid state phase transformations [25], fragmentation of lamellar structure of pearlitic [27] and separation of electrically neutral particles in liquids [28]. Here, it should be noted that there is a striking common feature: the formation of object has different electrical resistivities from that of the matrix. The different configurations of precipitates or inclusions in the matrix affect the distribution of electric current in the materials. Note that the different distribution of electric current corresponds to various system free energy [25,28]. It is still not sure whether electric current-induced free energy has an effect on grain refinement of alloys during solidification process. During casting, crystal nuclei first occur at, or close to, the wall of mold and present the different electrical resistivities from that of liquid alloys. The application of electropulsing may affect the distribution of crystal nuclei in liquid alloy to reduce the system free energy. This provides a possibility to utilize the driving force disturbance from electropulsing to crystal nuclei to refine the grains, while little attention is paid to the role of ΔG_e on crystal nucleus multiplication in grain refinement.

In this paper, the experimental investigation of the role of ΔG_e and the forced convection on the formation of FEG in pure aluminum was carried out. Since the solidification of pure metal is simple due to non-existent solute transport and knowledge about how electropulsing affects the solidification of pure aluminum has been accumulated by experimental and theoretical analysis, pure aluminum is selected to ensure that the formation mechanism of FEG is better clarified during the solidification process. For the work, the alternating electropulsing with frequency of 50 Hz and the effective value of current amplitude of less than 990 A was applied, since the prior work [20,29] indicated that effect of Joule heating and forced convection induced by Lorentz force on grain refinement could be distinguished by experiments with the mold embedded the wire mesh when electropulsing with 990 A was applied. It made it possible to reveal the role of ΔG_e on the formation of FEG based on the differences in electrical resistivities between the solid crystal nuclei and the liquid matrix. With regard to the complicated mechanism of grain refinement with electropulsing, we suggest that the dissociation of heterogeneous nuclei driven by ΔG_e and forced convection are two important factors, and it has important fundamental significance for the development of this technique in practice.

2. Experimental Procedure

The experiments were performed in a solidification apparatus (Northwestern Polytechnical University, Xian, Shaanxi, China) incorporated with a low-voltage alternating electropulsing generator, as schematically shown in Figure 1. The bottom electrode (a rectangle copper block with 65 mm in length, 50 mm in width and 45 mm in thickness) was fixed at the bottom of cylindrical sand mold with a diameter of 34 mm and a length of 110 mm. A copper block with a size of $50 \times 50 \times 45 \text{ mm}^3$,

as the top electrode, was welded to one end of the copper rod with 8 mm in diameter. The used sand mix consists of foundry sand, clay and a little water. In the present work, the high superheating temperature of Al melt, 1173 K, is required to guarantee that the melt can pass through the stainless mesh and eliminate the heterogeneous crystal nuclei formed on the mesh. During casting experiments, pure Al (99.7%) was initially superheated to 1173 K in a graphite crucible using a resistance furnace (Shanghai Shiyuan Electric Furnace Company, Shanghai, China) and kept isothermally for 30 min. Subsequently, the melt was poured into the sand mold, and then the top electrode was immediately dipped into the liquid metal with 1 mm in depth. Here, two types of experiments were performed. In the first one, the electropulsing with a frequency of 50 Hz was applied during solidification process after pouring the melt into the sand mold without preheating. For comparison, a sample without the application of electropulsing was prepared. In the second one, the sand molds were preheated to delay the formation of the solidified shell before the application of electropulsing. The effective values of applied electropulsing current were 480 and 900 A, respectively, and the corresponding current densities were 52.9 and 99.2 A·cm⁻².

An array of K-type thermocouples was used to make temperature measurements in the mold cavity. Here, three thermocouples were placed along the ingot centerline at fixed vertical positions of 19, 55 and 91 mm above the bottom electrode. Temperature readings (at 0.2 s intervals) were recorded by a computer-aid data acquisition system. Cooling curves were immediately measured after pouring the melt. Meanwhile, the alternating electropulsing was applied during solidification.

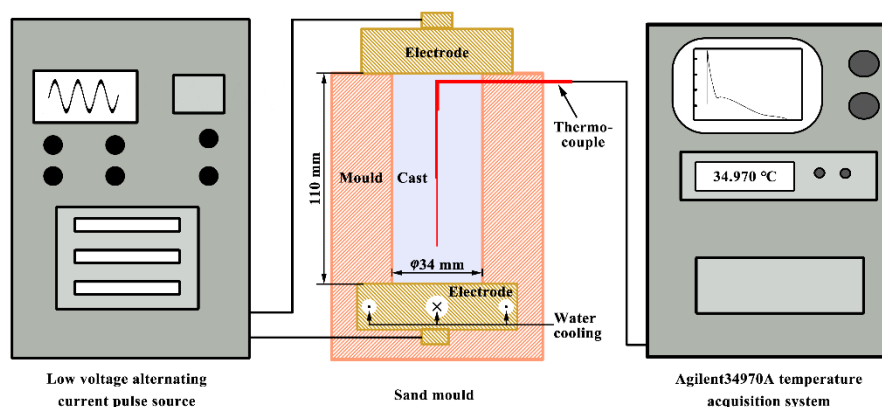


Figure 1. Schematic illustration of the solidification apparatus. Three thermocouples separated by 36 mm gaps were positioned along the axis of mold.

The cylindrical ingots were sectioned along the midplane, mechanically polished using abrasive papers, and etched with an acid solution (3 mL HF, 3 mL HNO₃, 9 mL HCl and 5 mL H₂O) to reveal the macrostructure. Photographs of the macrostructure were taken by a Canon EOS D60 digital camera, and the grain size was measured by a mean linear intercept method.

3. Results and Discussion

3.1. Effect of Electropulsing on the Grain Macrostructure

The as-cast macrostructure of pure aluminum ingot along the longitudinal section without electropulsing is shown in Figure 2. Note that the heat flux is proportional to the metal/mold heat transfer coefficient in the casting process. Since the heat transfer coefficient between the casting surface and the copper surface at the top and bottom of the mold ($\sim 10^3$ W·m⁻²·K⁻¹ [30]) at 933 K far exceeds that between the casting surface and sand mold surface ($\sim 10^2$ W·m⁻²·K⁻¹ [31]), the heat is extracted mainly through the top and bottom copper electrode. As a result, the upward and downward vertical columnar grains expand over most of the ingot, and intersect at the center of ingot indicated by a dotted horizontal white line. In addition, little columnar grains grow radially towards the center of

ingot, and exhibit a gradual increase in the length of the columnar grains from two ends of cylindrical mold to the axial center.

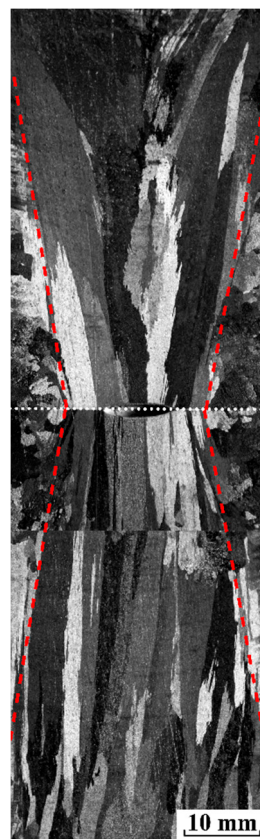


Figure 2. The longitudinal as-cast macrostructure of pure aluminum ingot without electropulsing.

The sequence of macrographs in Figures 3 and 4 displays, under electropulsing with 480 and 900 A, respectively, how the grain macrostructure of pure aluminum changes as a result of gradually increasing the preheating temperature of sand mold. One dotted white curve is superimposed on the macrographs to indicate the FEG zone.

When electropulsing with 480 A is applied to the specimen, using a mold at room temperature, the FEG region appeared in the middle and lower part of the ingot. An increase in the preheated temperature to 373 K (Figure 3b), 473 K (Figure 3c) or 573 K (Figure 3d) gives rise to a downward shift of FEG region, but the area of FEG and the mean grain size are almost unchanged, as illustrated in Figure 5. These results are different from previous observations for the application of electropulsing during the solidification of pure aluminum in which an increase in the amount of FEG as the preheated temperature of mold is increased [2,20]. In their study on an as-cast macrostructure preheating the sand mold, it indicates that more heterogeneous nuclei on the side wall dissociate and produce FEG under adequate electropulsing due to the delay of the solidified shell formation caused by preheating the mold. If we assume this to be operative, the expected macrostructure is in contradiction with the present results. It demonstrates that the dissociation of heterogeneous nuclei formed on the side wall will not happen under electropulsing with 480 A.

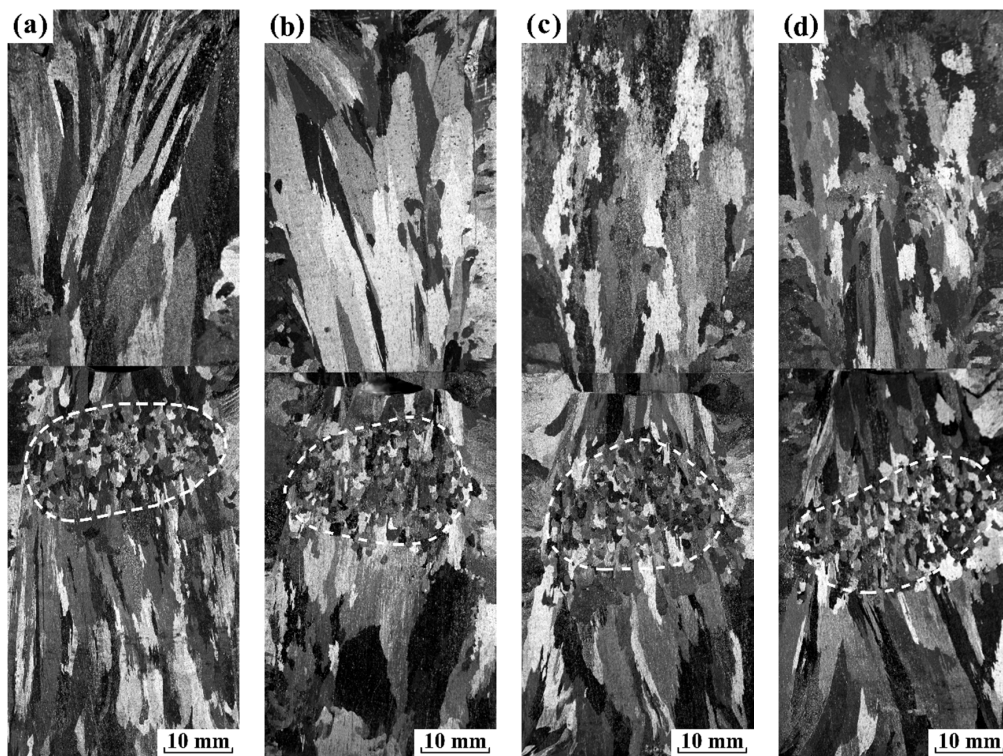


Figure 3. Longitudinal sections of the solidification structure in pure aluminum ingots under electropulsing with 480 A at different temperatures of mold: (a) room temperature; (b) 373 K; (c) 473 K and (d) 573 K.

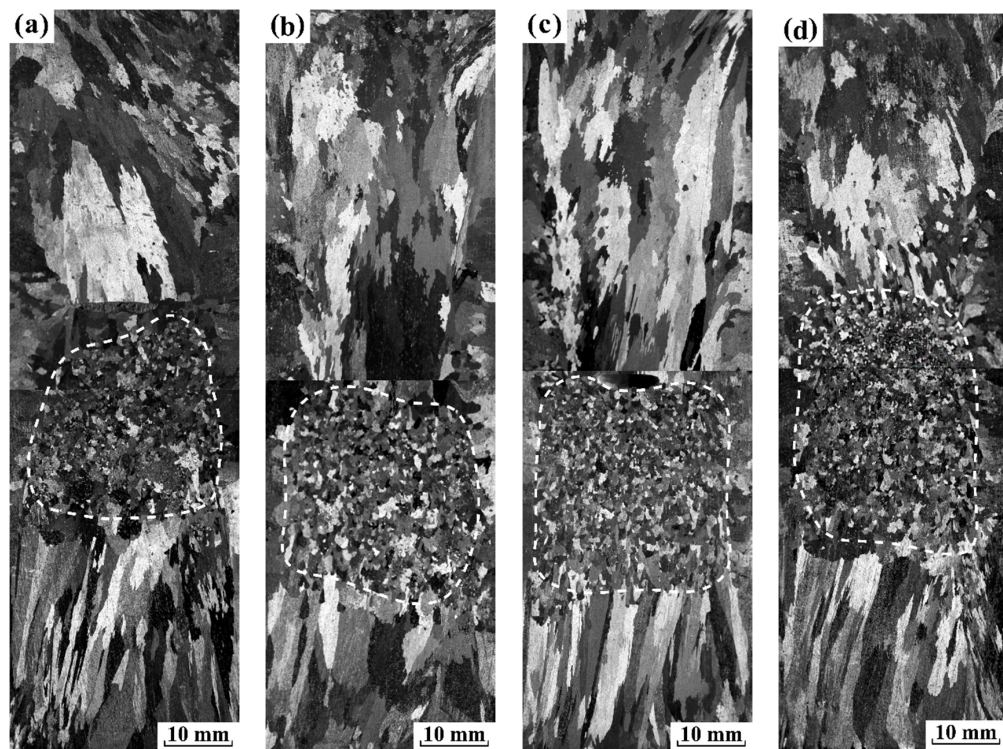


Figure 4. Longitudinal sections of the solidification structure in pure aluminum ingots under electropulsing with 900 A at different temperature of mold: (a) room temperature; (b) 373 K; (c) 473 K and (d) 573 K.

For a further increase in current intensity to 900 A (Figure 4) small equiaxed zone enlarges compared with that at the same preheated temperature of sand mold under electropulsing with 480 A, and the mean size of equiaxed grains becomes even more refined. In addition, the amount of FEG increases with increasing the preheated temperature of mold, as can be seen in Figure 5. As a result, the obviously different behavior to that under electropulsing of 900 A is observed for aluminum ingots under electropulsing with 480 A as the preheated temperature of mold is gradually increased. For grain refinement of pure aluminum under electropulsing, the recent studies [3,20,21] proposed that the dissociation of heterogeneous nuclei and dendrite fragmentation induced by the forced flow give rise to the multiplication of crystal nuclei. Moreover, the dissociation of heterogeneous nuclei on the side wall takes precedence over dendrite fragmentation [20]. Hence, it is speculated that the applied electropulsing with 900 A will inevitably lead to the dissociation of heterogeneous nuclei on the side wall, producing FEG. The other origin of FEG is further addressed in detail later.

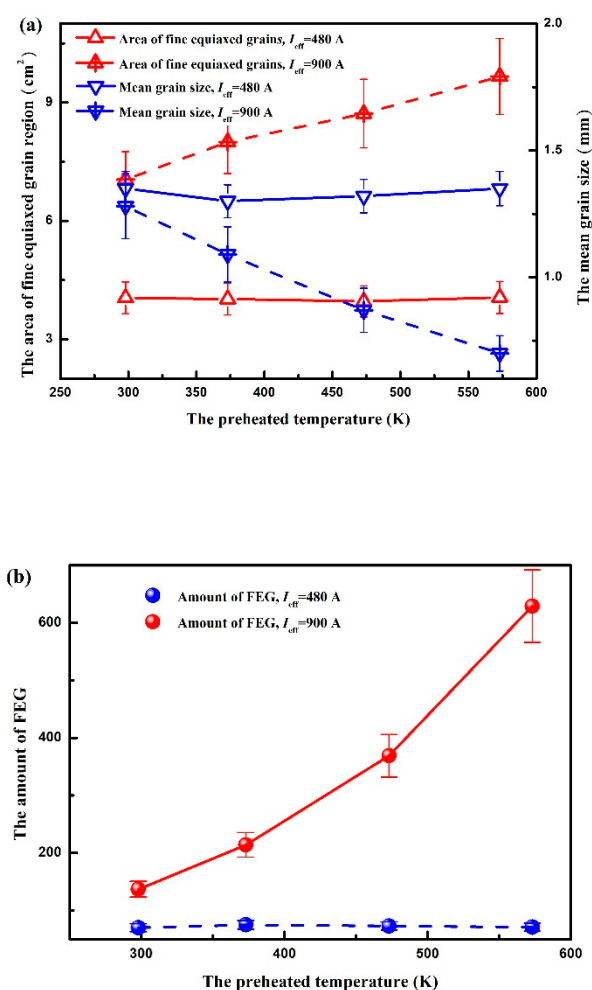


Figure 5. (a) The area of fine equiaxed grain region and the mean grain size in each of the ingots under electropulsing with 480 A and 900 A as a function of the preheating temperature of mold and (b) the amount of FEG in each of ingots under electropulsing with 480 A and 900 A as a function of their corresponding preheating temperature of mold.

3.2. Forced Flow Field and Relevant Thermal Analysis under Electropulsing

This subsection is focused on the forced convection caused by Lorentz force during solidification of pure aluminum under electropulsing mentioned above. Since the electromagnetical convection mainly depends on the intensity of electric current regardless of current pattern [3], the intensity of melt flow driven by electropulsing is nearly the same as that with direct current (DC) when the

effective value of electropulsing, is identical to the DC amplitude. Furthermore, any effect originating from skin effect can be neglected as a result of the applied frequency small enough in our case. Under the condition of this low frequency of 50 Hz, the radial and axial components of the Lorentz force ($F_L = j \times B$) in the melt caused by electropulsing and the induced magnetic field can be written as [32]:

$$F_{Lr} = \sigma(-E_z B_\theta - u_r B_\theta^2), \quad (1)$$

$$F_{Lz} = \sigma(E_r B_\theta - u_z B_\theta^2), \quad (2)$$

where σ is the electrical conductivity of the mixture of solid and liquid phase, E_z and E_r are the axial and radial components of the electric field intensity, u_z and u_r are the axial and radial components of the velocity of melt flow and B_θ is the azimuthal magnetic field. It is noted that Lorentz force is closely related to the velocity of melt flow and the electric potential. Thus, the calculation of F_L has to be coupled with the solution of momentum conservation equations. According to Equations (1) and (2), the electropulsing-driven convection in a cylindrical container is a distinct electro-vortex flow, which induces an axial jet along the direction of current density gradient for the configuration of up-down electrodes. Previous studies proposed that the electro-vortex flow plays a prominent role in the formation of FEG and macrosegregation of alloys [3,22,32,33].

To describe Lorentz force induced by an electropulsing and its driven convection, the S parameter that can be taken as the characteristic non-dimensional parameter is given as [34,35]:

$$S = \frac{\mu_0 I^2}{4\pi^2 \rho \nu^2}, \quad (3)$$

where I is the current intensity, which is the product of the current density j and cross-section area s , ν is the kinematic viscosity, ρ is the density of the liquid metal and μ_0 is the vacuum permeability. Although the value of S can characterize the relative intensity of the flow, it is not feasible to analyze the melt flow regime utilizing the value of S as a criterion. As a consequence, we could not conclude the flow was laminar or turbulent in the present study.

For cooling behaviors of alloys under the condition of electromagnetically driven convection, it is noted that the melt flow makes a distinct reduction of temperature gradient and the turbulent melt flow correspond to the oscillations of the temperature signals [3,36]. Since the influence of the same electropulsing on the temperature field with mold preheated to different temperature was found to be qualitatively identical, it is decided to give the cooling curves obtained with mold at room temperature in the following discussion in order to determine the flow pattern. The cooling curves obtained from temperature measurements at three thermocouples placed along the longitudinal axis of ingots without and with electropulsing are given in Figure 6. Without electropulsing, an obvious axial temperature gradient exists in the aluminum melt from the beginning of directional solidification. However, the effect of electropulsing with 480 A on the axial thermal gradient can be neglected, as displayed in Figure 6b. It suggests that the obvious melt flow does not occur in this case. Therefore, the formation mechanism of FEG in pure aluminum should be different from the previous model [3,19,20,22,37]. When the electropulsing with 900 A is applied, a distinct reduction of the temperature gradient becomes visible implying a thermal homogenization of aluminum melt, and there is no distinct disturbance on cooling curves. It reveals the existence of laminar melt flow spanning the entire melt volume, which is essential for the formation of FEG in alloys [3,19,32,33].

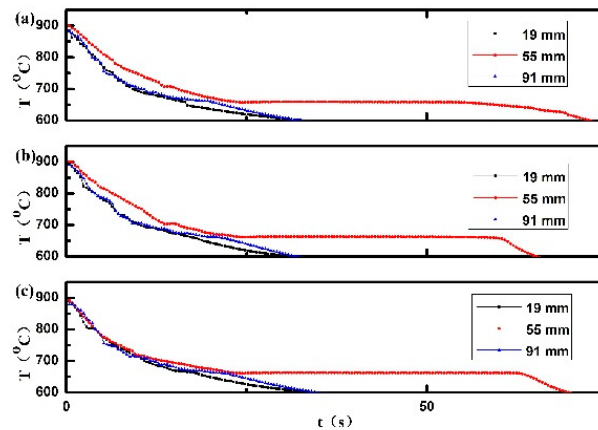


Figure 6. Cooling curves measured during solidification of pure aluminum: (a) without electropulsing; (b) under an alternating electropulsing with 480 A and 50 Hz and (c) under an alternating electropulsing with 900 A and 50 Hz.

3.3. Refinement Mechanism under Electropulsing

3.3.1. Macrostructure in Sand Mold Embedding the Mesh Tube under Electropulsing

As indicated above, the generation of FEG is probably not related to the weak convection caused by Lorentz force in the presence of electropulsing with 480 A. Additionally, the role of Joule heating on grain refinement of pure aluminum is insignificant for the present electropulsing of $\sim 53 \text{ A} \cdot \text{cm}^{-2}$ since effect of Joule heating on the solidification structure in Al-7Si alloy is almost negligible under electropulsing of $\sim 120 \text{ A} \cdot \text{cm}^{-2}$ in Al-7Si alloy [3]. Thus, in the present study the observed formation of FEG is likely to be the consequence of electric current-associated free energy change ΔG_e . When electric current passes through aluminum melt, the distribution of electric current is homogeneous. With reducing temperature, heterogeneous nuclei appear during the nucleation stage. Then the corresponding homogeneous distribution of electric current is modified because crystal nuclei have different electrical resistivity from that of the aluminum melt. Note that various distributions of electric current corresponds to the different G_e , and thus ΔG_e is given as [28,29,38]:

$$\Delta G_e = \frac{\mu}{8\pi} \iint \frac{j_1(r)j_1(r') - j_2(r)j_2(r')}{|r - r'|} d^3r d^3r', \quad (4)$$

where r and r' are two different positions in space, j is the current density and μ is the magnetic permeability. The subscript 1 and 2 represent the states before and after the formation of crystal nuclei. Equation (4) indicates that various distributions of electric current correspond to different ΔG_e . Since both the occurrence of crystal nucleus and the different configuration of crystal nucleus affect the distribution of electric current, ΔG_e is derived by summing the volume and motion terms for electric current-associated free energy change:

$$\Delta G_e = \Delta G_{eV} + \Delta G_{em}. \quad (5)$$

Assuming crystal nucleus of Al phase is a spherical form. If this nucleus occurs at the center of a spherical melt matrix under electropulsing, the general expression of the volume term can be given as [39]:

$$\Delta G_{eV} = k \frac{\sigma_1 - \sigma_2}{2\sigma_1 + \sigma_2} j^2 V, \quad (6)$$

and

$$k = \mu \left[\ln\left(\frac{R}{a}\right) - \frac{65}{48} - \frac{5}{48} \frac{\sigma_1 - \sigma_2}{2\sigma_1 + \sigma_2} \right] R^2, \quad (7)$$

where σ_1 and σ_2 are electric conductivities of the melt and crystal nuclei, respectively, R is the radius of the spherical melt-matrix, k is a positive geometry factor and a is the critical radius of crystal nucleus of Al phase, V is the volume of crystal nucleus. According to Equation (7), ΔG_{eV} will be negative due to $\sigma_1 < \sigma_2$. It means that the application of electropulsing reduces the change of system Gibbs free energy change and thus will help to promote heterogeneous nucleation in theory. However, effect of electropulsing on the nucleation rate is obviously insignificant in the nucleation stage of pure aluminum [20], i.e., ΔG_{eV} is much smaller than the change of Gibbs free energy between the liquid and solid phase.

If the spherical nucleus appears off-center, the crystal nucleus is driven from the initial position towards the center or surface of ingot by a force F arising from electropulsing. It can be expressed as [40]:

$$F = -\frac{3}{2} \frac{\sigma_1 - \sigma_2}{2\sigma_1 + \sigma_2} \zeta j \times BV, \quad (8)$$

where ζ is the geometry factor and B is the magnetic intensity. The force F points from the outer surface to the center of ingot and in the perpendicular direction of electric current due to $\sigma_1 < \sigma_2$. Assuming crystal nucleus moves from one position d to the center of ingot, ΔG_e will tend to decrease. The corresponding decrease in value is equal to the work of the above force:

$$\Delta G_{em} = -\frac{3}{2} \frac{\sigma_1 - \sigma_2}{2\sigma_1 + \sigma_2} \mu_0 \zeta j^2 d^2 V, \quad (9)$$

where d is the perpendicular distance from the position of crystal nucleus in space to the central axis of ingot. In our case, heterogeneous nucleation first occurs at the side wall and the interface of the electrodes and the melt. Since electropulsing is applied during the solidification process, the system free energy is related to the distribution of crystal nuclei during nucleation stage. As a result, crystal nuclei will move towards the central axis of ingot to minimize the Gibbs free energy of system. It is assumed that ΔG_{em} makes crystal nuclei appear at the side wall and the interface of the electrodes and the melt dissociate and drift in the melt, leading to the generation of FEG in ingot.

In order to check the validity of this assumption, it is of critical important that confirming where FEG originate from. Subsequently, a series of experiments with the mold embedded the mesh plate and tube of 06Cr25Ni20 stainless were designed. It is noted that the mesh plate and tube can separate crystal nuclei transfer [2,20,41], and suppress convection inside and outside the tube. Hence, the comparison of the solidification structure at the different regions is performed to clarify the role of ΔG_{em} on grain refinement in pure aluminum.

As shown in Figure 7, the experiment study on confirming the origin of FEG were designed under electropulsing with 480 A. For the first experiment, the applied sand mold consisted of a mesh plate located in the middle of mold and two mesh tubes embedded along axis. For another one, a mesh plate was only placed in the middle of mold. Moreover, both the solidification condition and the applied electropulsing parameters were the same as those of experiment 3 (a). Figure 7 illustrates the corresponding longitudinal sections of the solidification structure of pure aluminum ingots. It is observed that the fine equiaxed zone existed ahead of the vertical downward columnar grains above the mesh plate regardless of inside or outside the tube, as shown in Figure 7a. In contrast, the dominant columnar grains with vertical upward direction appeared below the mesh plate. In Figure 7b, the macrostructure above and below the mesh plate was similar to the corresponding one in Figure 7a. It is noted that current effects could be divided into similar halves along the mesh plate in our case. However, the direction of grain growth above and below the mesh plate reversed during the solidification process. From the macrostructural comparison between the vertical upward and downward directional solidification of Al subjected to electropulsing, there was no doubt that FEG above the mesh plate originated from the dissociation of heterogeneous nuclei on the upper interface of the electrode and the melt. These results were very similar to those presented by Li et al. [19] in which the nuclei detachment is ascribed to the visible shock wave driven by the periodic Lorentz force.

As has been reported before, the mesh tube has an effect on restricting the melt flow inside and outside the tube [20]. If the weak forced flow dominates the formation of FEG in our case, the total amount of FEG inside and outside the tube should obviously decrease, which is in contradiction with the macrostructural observation. It suggests that the weak forced flow had no effect on grain refinement, and thus the dissociation of crystal nuclei on the upper interface was solely attributed to ΔG_{em} .



Figure 7. The solidification macrostructure along the longitudinal section of pure aluminum ingots under electropulsing with 480 A with a mold of 34 mm embedded the metal mesh: (a) a mesh plate located in the middle of mold and two mesh tubes embedded along axis and (b) a mesh plate placed in the middle of mold.

As current intensity increased to 900 A, the origin of FEG refers to a more complex picture including ΔG_{em} on the solidification process and the forced flow. Thus, five experiments were designed to explain the role of ΔG_{em} and the forced flow on grain refinement in pure aluminum. For each experiment, sand mold consists of a mesh plate located in the middle of mold and a mesh tube with different diameters embedded along axis. The diameter of mesh tube was 0, 8, 12, 16 and 24 mm, respectively. Moreover, both the solidification condition and the applied electropulsing parameters were the same as those of experiment 4 (a). The longitudinal as-cast macrostructure of pure aluminum ingots using different molds is displayed in Figure 8. Without the mesh tube, a significant fine equiaxed zone appeared in the front of vertical downward columnar grains above the mesh plate and vertical upward columnar grains below the mesh plate. However, the equiaxed grain above the mesh plate was remarkably finer compared with those below the mesh plate. When a tube with 8 mm in diameter was embedded, the macrostructure changed little above the mesh plate. Below the mesh plate, columnar grains expanded over the entire height of mesh tube inside it, and a fine equiaxed zone occurs outside the tube. An increase in the diameter of mesh tube to 12 mm (Figure 8c), 16 mm (Figure 8d) or 24 mm (Figure 8e) decreased the amount of FEG outside the tube, but the coarse columnar grains still dominated the macrostructure inside the tube. The observed change of fine equiaxed zone below the

mesh plate in the present study was quite similar to those presented by previous work [20]. It implies that the forced convection plays an important role in the generation of FEG. However, it could not be determined to whether ΔG_{em} will give rise to the dissociation of heterogeneous nuclei on the side wall or not.

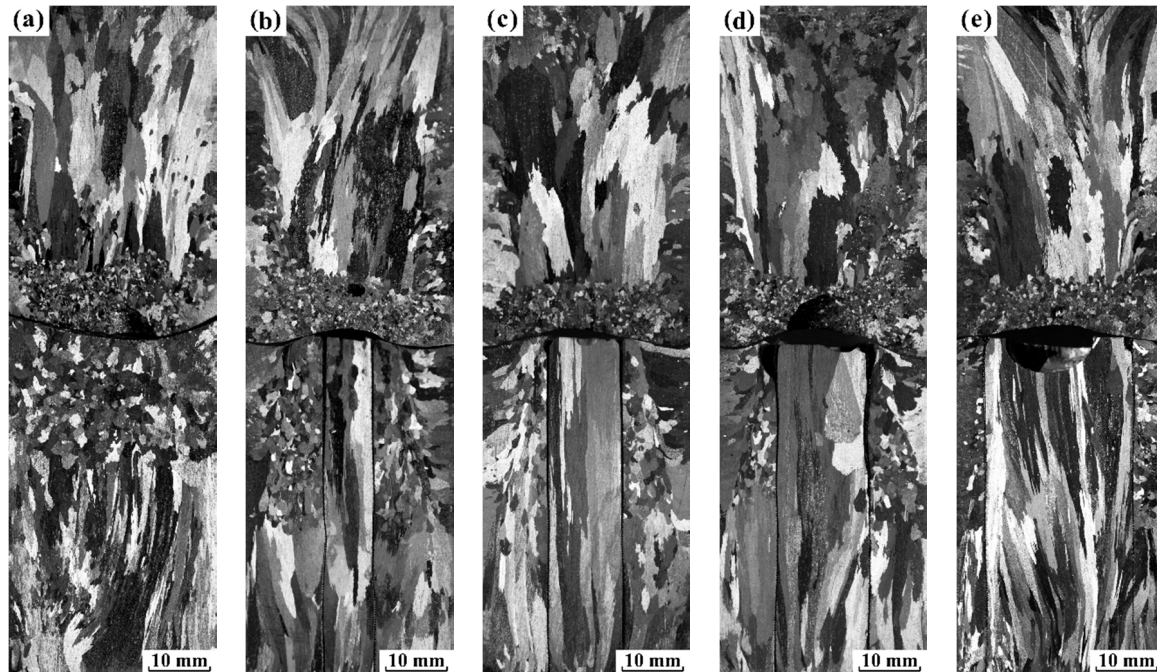


Figure 8. The solidification macrostructure along the longitudinal section of pure aluminum ingots under electropulsing with 900 A with mold of 34 mm embedded a mesh plate located in the middle of mold and a mesh tube of different diameter embedded along axis: (a) without the tube; (b) the tube of 8 mm; (c) the tube of 12 mm; (d) the tube of 16 mm and (e) the tube of 24 mm.

As discussed above, the formation of FEG should attribute to competition and cooperation between ΔG_{em} and the forced melt flow. To clarify this speculation, the amount of FEG in different regions of mold embedded stainless mesh below the mesh plate N was measured with respect to the different melt flow. Figure 9 represents the variation in N as a function of non-dimensional S parameter, which was calculated based on Equation (3). Here, the value $\nu = 1.25 \text{ mPa}\cdot\text{s}$ [42] and $\rho = 2.35 \text{ g}\cdot\text{cm}^{-3}$ [43] were employed. It can be seen that N gradually increased with the S parameter outside the tube under the same electropulsing of $99.2 \text{ A}\cdot\text{cm}^{-2}$, and there was no the formation of FEG inside the tube in the present experiments. When the calculated S parameter was approximately 1.77 FEG occurred outside the tube. In contrast, there was no the formation of FEG beyond this value under electropulsing of $52.9 \text{ A}\cdot\text{cm}^{-2}$. Since the driving force ΔG_{em} was proportional to current density, ΔG_{em} under electropulsing of $52.9 \text{ A}\cdot\text{cm}^{-2}$ was probably insufficient to drive the dissociation of heterogeneous crystal nuclei at the lateral wall (Figure 7b). Meanwhile, the present melt flow had little effect on the formation of FEG. This speculation explains the experimental data under electropulsing of $52.9 \text{ A}\cdot\text{cm}^{-2}$ very well. It is noted that even if the intensity of forced convection was much smaller than in the preceding case, FEG still appeared near the mold wall under electropulsing of $99.2 \text{ A}\cdot\text{cm}^{-2}$ (Figure 8e). This indicates that the higher driving force ΔG_{em} gave rise to the dissociation of crystal nuclei on the side wall, leading to the generation of FEG. For a further increase in the intensity of the melt flow (Figure 8a–d) the additional FEG arising from the dissociation of crystal nuclei on the side wall was induced by the stronger convection.

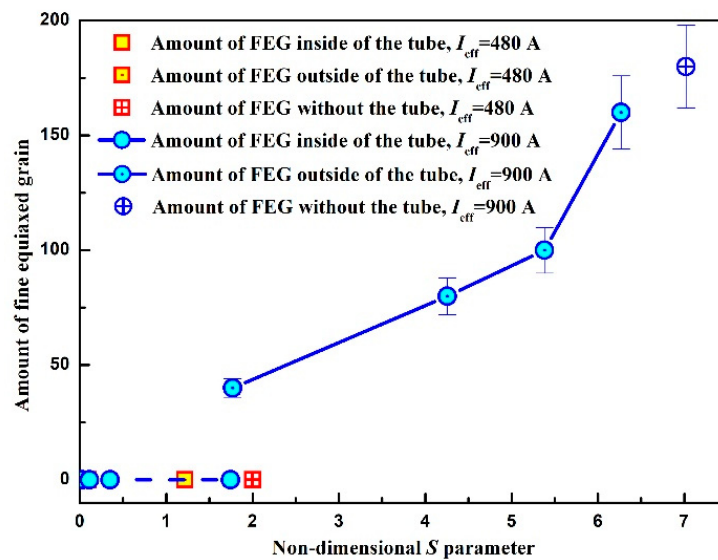


Figure 9. Relationship between N in different regions of mold embedded stainless mesh below the mesh plate and non-dimensional S parameter.

As described in the introduction, the strong forced flow can induce dendrite fragmentation, leading to the formation of FEG [20,21]. However, the present analysis cannot identify whether dendrite fragmentation is distinctly induced by the application of electropulsing with 900 A. In order to explain it, a casting experiment with an improved mold was designed. As shown in Figure 10a, the mold was separated into two parts by a horizontally placed mesh: the sand mold with 34 mm in diameter and 55 mm in height above the mesh, the ceramic mold with 34 mm in internal diameter, 55 mm in height and a 3 mm in thickness below the mesh. The outer surface of the ceramic mold was twined by heated Cr20Ni80 resistance wire to prevent the radial heat flux through the mold. Here, the temperature of the side wall was preheated to 933 K, both the solidification condition and the applied electropulsing parameters were same as those of experiment 4 (a). The longitudinal as-cast macrostructure of pure aluminum ingots using an improved mold is displayed in Figure 10b. As a result, a significant fine equiaxed zone appears in the front of vertical downward columnar grains above the mesh plate. However, the coarse columnar grains take up the entire ceramic mold. Below the mesh plate, crystal nuclei appeared at the bottom electrode and developed into columnar structure during the solidification process. If FEG generates in this case, its dominant origin should be dendrite fragmentation caused by the melt flow. From macrostructural observation, it could confirm that the application of electropulsing with 900 A could not lead to the significant fragmentation of columnar grains.

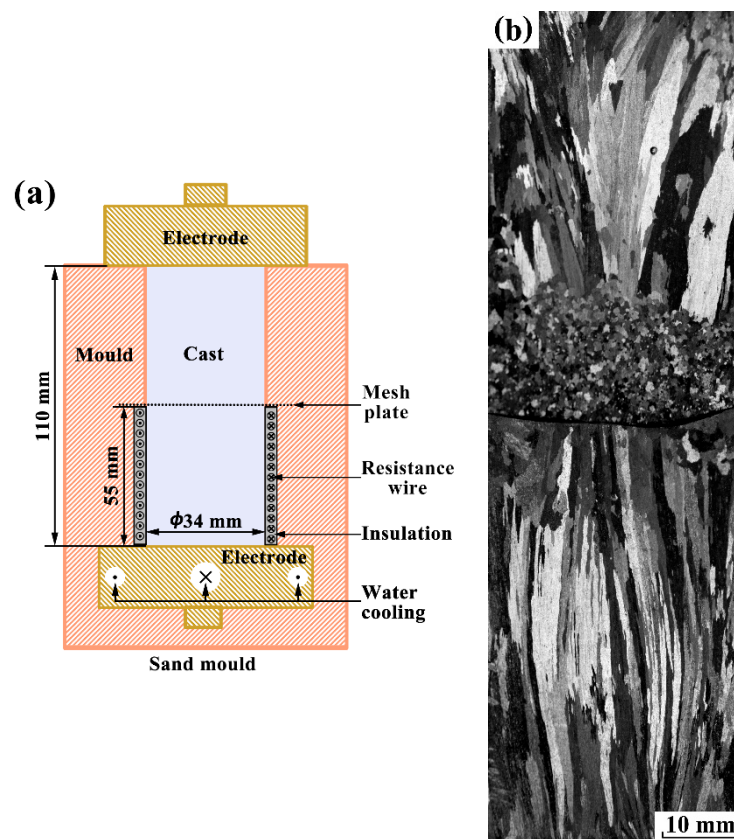


Figure 10. (a) Schematic sketch of the improved mold and (b) longitudinal sections of the solidification structure in pure aluminum ingots under electropulsing with 900 A with the improved mold.

3.3.2. Formation of FEG

In view of the above investigation, the following conclusions were demonstrated.

- (1) The amount of FEG remains almost unchanged as the preheated temperature of mold increased. It confirmed that the dissociation of heterogeneous nuclei formed on the side wall did not occur under electropulsing with 480 A (Figure 3).
- (2) A partial FEG result from the dissociation of heterogeneous nuclei formed on the lateral wall when electropulsing with 900 A was applied (Figure 4).
- (3) The obvious melt flow did not occur under electropulsing with 480 A. However, the application of electropulsing with 900 A could lead to a laminar melt flow spanning the entire melt volume (Figure 6).
- (4) Since the forced melt flow was not taken into account under electropulsing with 480 A, the formation of FEG was attributed to the dissociation of heterogeneous nuclei on the upper interface driven by ΔG_{em} .
- (5) The origin of FEG consisted of the dissociation of heterogeneous nuclei on the side wall and the upper interface due to ΔG_{em} and the forced melt flow under electropulsing with 900 A.

From these findings, the following scenario was considered for the formation process of FEG during solidification of pure aluminum with the application of present electropulsing, as schematically illustrated in Figure 11. Under electropulsing with 480 A, heterogeneous nuclei firstly appeared on the side wall and the interface of the electrodes and the melt. The applied electropulsing made heterogeneous nuclei fall off from the upper interface until the solidification shell forms due to ΔG_{em} , as shown in Figure 11ai. Then, the dissociated crystal nuclei moved towards the bottom of the mold due to its gravity and the grains in the solidification shell, which could grow parallel and opposite

to the heat flow direction, would develop into a columnar structure (Figure 11aii). Subsequent, columnar-to-equiaxed transition occurred, if the number and size of the equiaxed grains ahead of the upward and downward columnar grain front accumulated to their limit, i.e., the columnar growth was blocked and an equiaxed zone formed, as shown in Figure 11aiii. Similar results have been reported in earlier studies [44,45]. The formation process of FEG under electropulsing with 900 A is schematically illustrated in Figure 11bi–iii. Compared with the proposed process under electropulsing with 480 A, the main distinction was that FEG grew from the dissociated crystal nuclei, which resulted from the lateral wall and the upper interface of the electrode and the melt due to ΔG_{em} and the forced convection under electropulsing with 900 A, as shown in Figure 11bi.

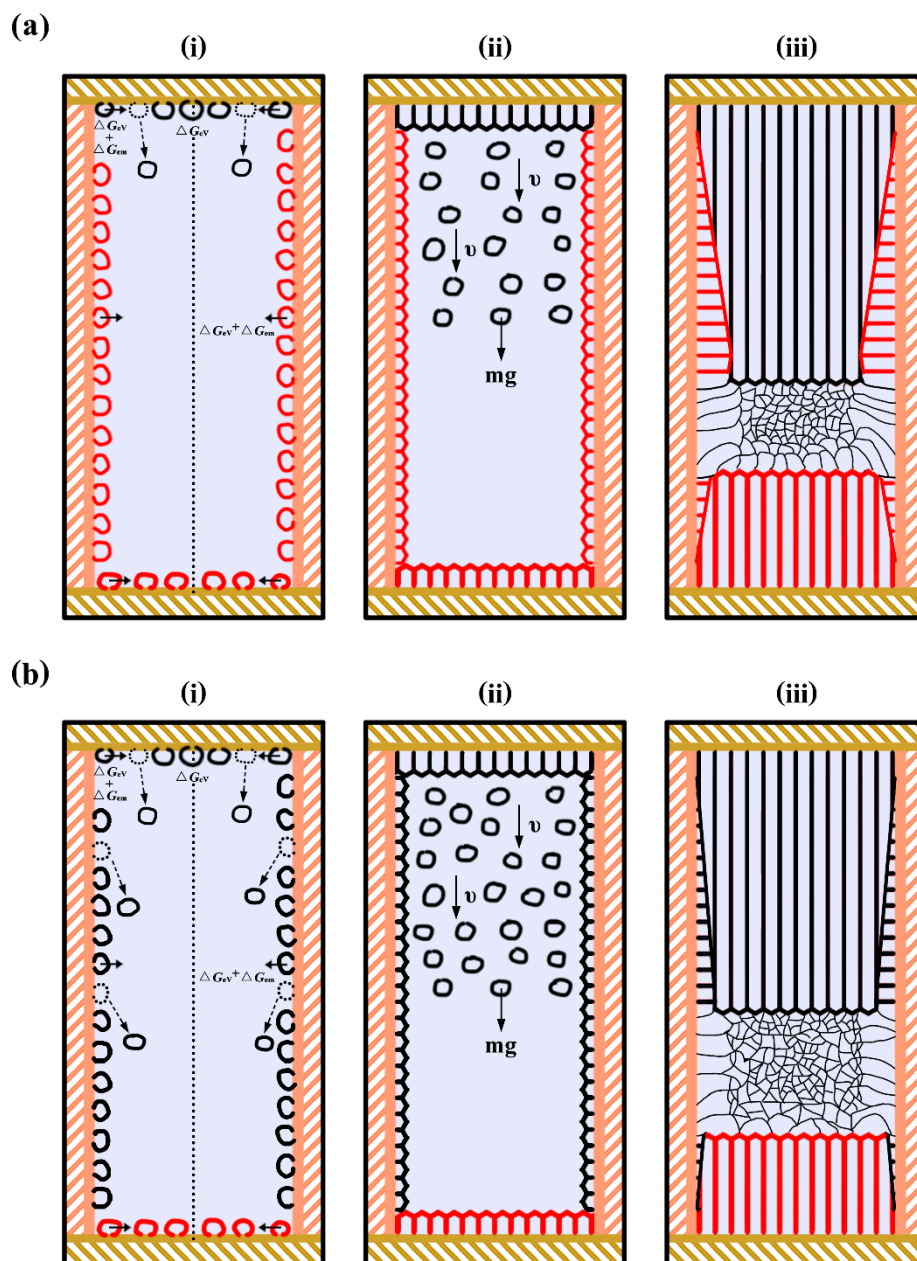


Figure 11. Schematic illustration of the formation process of FEG observed in the present study. The solidification proceeds from (i) to (iii). (a) under electropulsing with 480 A and (b) under electropulsing with 900 A.

As indicated above, the driving force ΔG_{em} was one of the important factors of inducing the dissociation of crystal nuclei in the present study. However, the dissociation of crystal nuclei caused by ΔG_{em} on the upper interface was easier than that on the lateral wall, probably due to the difference in the direction of F imposed to the crystal nuclei attachment surface. As has been reported before, the direction of F is axial symmetrical and from the center of ingot to the lateral surface when the electrical conductivity of inclusions is less than that of the matrix [28]. Since the electrical conductivity of crystal nuclei ($\sim 10^5 \text{ S}\cdot\text{cm}^{-1}$ at 873 K [46]) was more than that ($\sim 4 \times 10^4 \text{ S}\cdot\text{cm}^{-1}$ at 973 K [46]) of the molten aluminum, F with the reversed direction was produced in our case. In other words, the direction of F pointed towards the central axis and was parallel to the upper interface of the electrode and the melt when the force was imposed to crystal nuclei on the substrate of the upper interface. In contrast, the direction was perpendicular to the surface of mold for the force applied to crystal nuclei on the substrate of lateral wall. From the macrostructural observation, it confirmed that the required shear stress on the upper interface of the electrode and crystal nuclei for the dissociation of crystal nuclei was less compared to the normal stress on the contact surface of the lateral wall and crystal nuclei.

Under electropulsing with 900 A, the proposed process for the dissociation of crystal nuclei on the side wall was the result of ΔG_{em} in cooperation with the forced convection, which was distinctly different from the contribution of magnetic pressure pulses presented by Liao et al. [3]. It is noted that the non-dimensional S parameter utilized to characterize the intensity of forced convection was proportional to $(jR^2)^2$, and the maximum value of ΔG_{em} varied as $(jR)^2$. Based on these different functional dependencies, we could conclude the following: (i) there was a threshold value of current density, below which the forced melt flow would make the dissociation of crystal nuclei and thus led to grain refinement before ΔG_{em} had any effect on crystal nuclei on the upper surface. The effect of ΔG_{em} on grain refinement will have a priority if current density exceeds the threshold value. This analysis was strongly supported by the macrostructural observation shown in Figure 7; (ii) another higher threshold value of current density existed. ΔG_{em} firstly dominated the dissociation of crystal nuclei on the upper interface and the side wall before the forced convection played a role in forming the refined grains if current density exceeded this value; (iii) if current density and the size of the mold were large enough, both ΔG_{em} and the forced convection played important roles in the dissociation of crystal nuclei on the side wall for grain refinement, which were in agreement with the analysis for the solidification structure illustrated in Figure 8a–d. Therefore, the model of the combination of ΔG_{em} and the forced melt flow for grain refinement explained the macrostructural difference below the mesh plate shown in Figures 7 and 8, and strongly supported the validity of the proposed scenario in Figure 11.

It is noted that the scenario of the FEG formation process proposed here essentially relied on ΔG_{em} and the forced melt flow. As described in the introduction, previous studies [5,20,21] demonstrated that dendrite fragmentation induced by the strong forced convection was one of the main origins of FEG. Therefore, the mechanism of grain refinement considered comprehensively factors in terms of the dissociation of crystal nuclei due to ΔG_{em} , and the dissociation of crystal nuclei and dendrite fragmentation due to the forced melt flow. Figure 12 shows the schematic summary of the origin of FEG in pure aluminum under electropulsing. j and R were the main variables that determined the origin of FEG in a normal casting. The dash curves represent a constancy of the origin of FEG induced by ΔG_{em} (no effect, the dissociation of crystal nuclei on the upper interface, and on the side wall). Additionally, the various values (solid curves running from upper left to lower right) represent a constancy of the origin of FEG induced by the forced melt flow (no effect, the dissociation of crystal nuclei and dendrites fragmentation). The different regions segmented by curves correspond to the different origins of FEG. Thus, the role of ΔG_{em} and the forced melt flow on the origin of FEG in pure aluminum under electropulsing could be determined. The circle and square symbol were superimposed on the graph to indicate the origin of FEG under electropulsing of 52.9 and $99.2 \text{ A}\cdot\text{cm}^{-2}$, respectively.

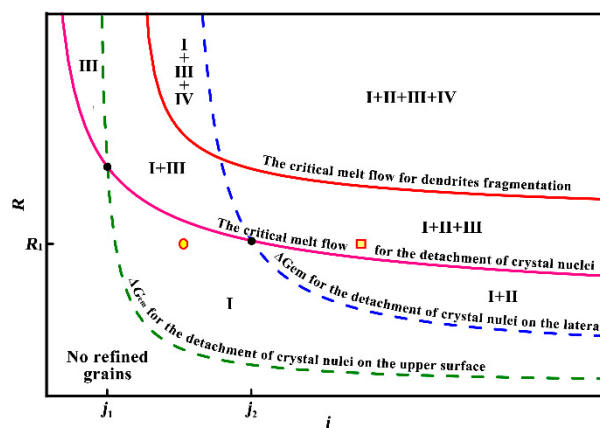


Figure 12. Schematic summary of the origin of FEG in pure aluminum under electropulsing. For the origin of FEG, I, III and IV represent the dissociation of crystal nuclei on the upper interface of the electrode and the melt due to ΔG_{em} , the dissociation of crystal nuclei on the side wall due to ΔG_{em} , the dissociation of crystal nuclei due to melt flow and dendrite fragmentation, respectively.

3.3.3. The Solidification Macrostructure under Different Conditions

According to the analysis above, the formation of FEG observed in these samples were attributed to ΔG_{em} and the forced convection driven by Lorentz force. Furthermore, which current effects play the dominant roles for grain refinement of pure aluminum depends on the current density and the size of the mold. To check the validity of the proposed process, the concluded results that have not been demonstrated should be verified experimentally by controlling ΔG_{em} and the forced melt flow.

As indicated by Figure 12, the forced convection might give rise to the dissociation of crystal nuclei besides the dissociation of crystal nuclei on the upper interface due to ΔG_{em} under electropulsing of $52.9 \text{ A}\cdot\text{cm}^{-2}$ as increasing the radius of ingots, which was proved by the two following casting experiments. Here, the applied current intensity was 700 A and the size of the sand mold was changed compared with that in the casting experiments under electropulsing (Figure 1). For the first experiment, the cylindrical sand mold with 40 mm in diameter was selected and a mesh plate was located in the middle of mold in this case. For another one, the sand mold consisted of a mesh plate embedded along the cross section and a mesh tube with 20 mm in diameter embedded along axis. The corresponding solidification macrostructure is shown in Figure 13. Obviously, a significant fine equiaxed zone appeared ahead of the downward columnar front above the mesh plate and upward columnar front below the mesh plate, respectively. However, the equiaxed grains above the mesh plate were obviously finer compared with those below the mesh plate. When a mesh tube was embedded, coarse columnar grains took up the entire mesh tube inside and outside it below the mesh plate. Based on Equation (3), the non-dimensional S parameter without the mesh tube was calculated to be 4.25, which exceeded the required value (<4.26) for the dissociation of crystal nuclei on the side wall and was below the critical value (>7.02) for dendrite fragmentation due to melt flow. It suggests that FEG below the mesh plate resulted from the dissociation of crystal nuclei on the side wall due to the forced convection. The S parameter was also calculated to be 0.27 and 2.39 inside and outside the tube, respectively. These values were below the critical value for the dissociation of crystal nuclei on the side wall due to forced convection. From the macrostructural observation (Figure 13), it confirmed that ΔG_{em} had no effect on the formation of FEG below the mesh plate, and the forced convection solely led to the formation of FEG originated from the dissociation of crystal nuclei on the side wall in this case, which was identical with the theoretical analysis. Therefore, the role of ΔG_{em} and the forced convection on grain refinement in pure aluminum mainly depended on the size of the mold and current density.



Figure 13. The longitudinal solidification macrostructure in pure aluminum ingots under electropulsing of 700 A with mold of 40 mm in diameter embedded the metal mesh: (a) a mesh plate located in the middle of mold and (b) a mesh plate embedded along the cross section and a mesh tube embedded along axis.

4. Conclusions

In the present study, the effect of an alternating electropulsing on grain refinement in pure aluminum was investigated by the designed casting experiments. Although the application of electropulsing can induce various kinds of current effects including Joule heating, ΔG_{em} and the forced convection are particularly worth considering for grain refinement. Correspondingly, the origin of FEG is different from the conventional assumption for the dissociation of crystal nuclei on the side wall and dendrite fragmentation induced by the forced convection. The important findings could be drawn as follows.

- (1) During solidification in the present casting, crystal nuclei firstly generated at the upper interface of the electrodes and the melt and the side wall. The driven force ΔG_{em} was the first to have an effect on grain refinement due to the dissociation of crystal nuclei on the upper interface if $j > j_1$, which ΔG_{em} or the forced convection had a priority to the dissociation of crystal nuclei at the side wall depended on j_2 in Figure 12.
- (2) There was also a critical value for the forced melt flow, above which the dissociation of crystal nuclei at the upper interface of the electrodes and the melt and the side wall was induced and developed into FEG. With the increase of the forced melt flow, dendrites fragmentation during crystal growth occurred and became a significant factor for grain refinement.
- (3) Current density and the size of mold were two main parameters for grain refinement under electropulsing, which determined ΔG_{em} and the forced convection, affected the origin of FEG.

Author Contributions: Conceptualization, L.Z. and R.Z.; methodology, L.Z. and N.L.; validation, N.L. and K.S.; formal analysis, N.L. and K.S.; data curation, P.Y. and H.X.; writing—original draft preparation, N.L. and L.Z.; writing—review and editing, R.Z., P.Y., H.X. and H.W.; visualization, H.W.; supervision, R.Z.; project administration, R.Z.; funding acquisition, L.Z.

Funding: This research was funded by the National Natural Science Foundation of China, grant number 51704242, 51871132 and the Natural Science Foundation of Shaanxi Province in China, grant number 2018JM5094 and the Fundamental Research Funds for the Central Universities, grant number 310201911cx019.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

References

1. Nakada, M.; Shiohara, Y.; Flemings, M.C. Modification of solidification structures by pulse electric discharging. *ISIJ Int.* **1990**, *30*, 27–33. [[CrossRef](#)]
2. Liao, X.; Zhai, Q.; Luo, J.; Chen, W.; Gong, Y. Refining mechanism of the electric current pulse on the solidification structure of pure aluminum. *Acta Mater.* **2007**, *55*, 3103–3109. [[CrossRef](#)]
3. Rübiger, D.; Zhang, Y.; Galindo, V.; Franke, S.; Willers, B.; Eckert, S. The relevance of melt convection to grain refinement in Al-Si alloys solidified under the impact of electric currents. *Acta Mater.* **2014**, *79*, 327–338. [[CrossRef](#)]
4. Li, N.; Zhang, L.; Zhang, R.; Yin, P.; Xing, H.; Wu, H. Research on grain refinement in hypoeutectic Al-Si alloy during solidification under an alternating electric current pulse. *Metals* **2019**, *9*, 571. [[CrossRef](#)]
5. Zhang, Y.H.; Rübiger, D.; Eckert, S. Solidification of pure aluminium affected by a pulsed electrical field and electromagnetic stirring. *J. Mater. Sci.* **2016**, *51*, 2153–2159. [[CrossRef](#)]
6. Wang, T.; Zhu, J.; Kang, H.; Chen, Z.; Fu, Y.; Huang, W.; Xiao, T. In situ synchrotron X-ray imaging on morphological evolution of dendrites in Sn-Bi hypoeutectic alloy under electric currents. *Appl. Phys. A Mater.* **2014**, *117*, 1059–1066. [[CrossRef](#)]
7. Zhang, L.M.; Li, N.; Zhang, R.; Xing, H.; Song, K.; Wang, J.Y. Effect of medium-density direct current on dendrites of directionally solidified Pb-50Sn alloy. *Mater. Sci. Technol.* **2016**, *32*, 1877–1885. [[CrossRef](#)]
8. Atamanenko, T.V.; Eskin, D.G.; Zhang, L.; Katgerman, L. Criteria of grain refinement induced by ultrasonic melt treatment of aluminum alloys containing Zr and Ti. *Met. Mater. Trans. A* **2010**, *41*, 2056–2066. [[CrossRef](#)]
9. Zhai, W.; Hong, Z.Y.; Wen, X.L.; Geng, D.L.; Wei, B. Microstructural characteristics and mechanical properties of peritectic Cu-Sn alloy solidified within ultrasonic field. *Mater. Des.* **2015**, *72*, 43–50. [[CrossRef](#)]
10. Zheng, D.; Chen, R.; Ma, T.; Ding, H.; Su, Y.; Guo, J.; Fu, H. Coupling effects of melt treatment and ultrasonic treatment on solidifying microstructure and mechanical performance of Ti44Al6Nb1Cr alloy. *Met. Mater. Trans. A* **2017**, *49*, 537–549.
11. Wang, F.; Eskin, D.; Mi, J.; Connolley, T.; Lindsay, J.; Mounib, M. A refining mechanism of primary Al₃Ti intermetallic particles by ultrasonic treatment in the liquid state. *Acta Mater.* **2016**, *116*, 354–363. [[CrossRef](#)]
12. Matsuda, K.; Takehara, T.; Yang, M.; Uno, H.; Kubo, T.; Miyano, G.; Yoshida, M. Verification of the mechanism of grain refinement by ultrasonic treatment of aluminum-4 wt pct silicon molten alloy. *Met. Mater. Trans. A* **2016**, *47*, 2509–2516. [[CrossRef](#)]
13. Li, M.; Tamura, T.; Miwa, K. Controlling microstructures of AZ31 magnesium alloys by an electromagnetic vibration technique during solidification: From experimental observation to theoretical understanding. *Acta Mater.* **2007**, *55*, 4635–4643. [[CrossRef](#)]
14. Campanella, T.; Charbon, C.; Rappaz, M. Grain refinement induced by electromagnetic stirring: A dendrite fragmentation criterion. *Met. Mater. Trans. A* **2004**, *35*, 3201–3210. [[CrossRef](#)]
15. Liu, H.; Xuan, W.; Xie, X.; Li, C.; Wang, J.; Yu, J.; Li, X.; Zhong, Y.; Ren, Z. Columnar-to-equiaxed transition and equiaxed grain alignment in directionally solidified Ni₃Al alloy under an axial magnetic field. *Met. Mater. Trans. A* **2017**, *48*, 4193–4203. [[CrossRef](#)]
16. Wrobel, T. Structure of pure aluminum after endogenous and exogenous inoculation. *Adv. Cryst. Process.* **2012**, 531–556. [[CrossRef](#)]
17. Conrad, H. Influence of an electric or magnetic field on the liquid-solid transformation in materials and on the microstructure of the solid. *Mater. Sci. Eng. A* **2000**, *287*, 205–212. [[CrossRef](#)]

18. Jianming, L.; Shengli, L.; Jin, L.; Hantong, L. Modification of solidification structure by pulse electric discharging. *Scr. Met. Mater.* **1994**, *31*, 1691–1694. [\[CrossRef\]](#)
19. Li, J.; Ma, J.; Gao, Y.; Zhai, Q. Research on solidification structure refinement of pure aluminum by electric current pulse with parallel electrodes. *Mater. Sci. Eng. A* **2008**, *490*, 452–456. [\[CrossRef\]](#)
20. Zhang, L.; Liu, H.; Li, N.; Wang, J.; Zhang, R.; Xing, H.; Song, K. The relevance of forced melt flow to grain refinement in pure aluminum under a low-frequency alternating current pulse. *J. Mater. Res.* **2016**, *31*, 396–404. [\[CrossRef\]](#)
21. Zhang, Y.H.; Xu, Y.Y.; Ye, C.Y.; Sheng, C.; Sun, J.; Wang, G.; Miao, X.C.; Song, C.J.; Zhai, Q.J. Relevance of electrical current distribution to the forced flow and grain refinement in solidified Al-Si hypoeutectic alloy. *Sci. Rep. UK* **2018**, *8*, 3242. [\[CrossRef\]](#) [\[PubMed\]](#)
22. Liotti, E.; Lui, A.; Vincent, R.; Kumar, S.; Guo, Z.; Connolley, T.; Dolbnya, I.P.; Hart, M.; Arnberg, L.; Mathiesen, R.H.; et al. A synchrotron X-ray radiography study of dendrite fragmentation induced by a pulsed electromagnetic field in an Al-15Cu alloy. *Acta Mater.* **2014**, *70*, 228–239. [\[CrossRef\]](#)
23. Jiang, H.; He, J.; Zhao, J. Influence of electric current pulses on the solidification of Cu-Bi-Sn immiscible alloys. *Sci. Rep. UK* **2015**, *5*, 12680.
24. Wang, X.L.; Guo, J.D.; Wang, Y.M.; Wu, X.Y.; Wang, B.Q. Segregation of lead in Cu-Zn alloy under electric current pulses. *Appl. Phys. Lett.* **2006**, *89*, 061910. [\[CrossRef\]](#)
25. Qin, R.S.; Samuel, E.I.; Bhowmik, A. Electropulse-induced cementite nanoparticle formation in deformed pearlitic steels. *J. Mater. Sci.* **2011**, *46*, 2838–2842. [\[CrossRef\]](#)
26. Zhang, X.F.; Qin, R.S. Segregation of copper in an Fe-Cu alloy under pulsed electric current. *Phil. Mag. Lett.* **2015**, *95*, 367–375. [\[CrossRef\]](#)
27. Rahnama, A.; Qin, R.S. Electropulse-induced microstructural evolution in a ferritic-pearlitic 0.14% C steel. *Scr. Mater.* **2015**, *96*, 17–20. [\[CrossRef\]](#)
28. Zhang, X.; Qin, R. Electric current-driven migration of electrically neutral particles in liquids. *Appl. Phys. Lett.* **2014**, *104*, 114106. [\[CrossRef\]](#)
29. Zhang, L.; Zhang, R.; Chen, W.; Wu, Y.; Li, N. Effect of a novel low-voltage alternating current pulse on solidification structure of Al-7Si-0.52Mg alloy. *Adv. Mater. Res.* **2012**, *482–484*, 1431–1435. [\[CrossRef\]](#)
30. Du, F.; Zhang, P.; Xu, Z.; Zhao, L. Law of heat transfer and simulation of temperature field for aluminum ingot solidification. *Chin. J. Nonferrous Met.* **2007**, *17*, 1750–1754.
31. Xu, H.; Hou, H.; Zhao, Y.; Chen, Z. Determination of boundary heat-transfer coefficient for casting solidification simulation. *Chin. J. Nonferrous Met.* **2003**, *13*, 881–886.
32. Nikrityuk, P.A.; Eckert, K.; Grundmann, R.; Yang, Y.S. An impact of a low voltage steady electrical current on the solidification of a binary metal alloy: A numerical study. *Steel Res. Int.* **2007**, *78*, 402–408. [\[CrossRef\]](#)
33. Zhang, Y.; Miao, X.; Shen, Z.; Han, Q.; Song, C.; Zhai, Q. Macro segregation formation mechanism of the primary silicon phase in directionally solidified Al-Si hypereutectic alloys under the impact of electric currents. *Acta Mater.* **2015**, *97*, 357–366. [\[CrossRef\]](#)
34. Bojarevičs, V.; Freibergs, J.A.; Shilova, E.I.; Shcherbinin, E.V. *Electrically Induced Vortical Flows*; Kluwer Academic Publishers: Dordrecht, The Netherlands, 1989.
35. Millere, R.P.; Sharamkin, V.I.; Shcherbinin, E.V. Effect of a longitudinal magnetic field on electrically driven rotational flow in a cylindrical vessel. *Magnetohydrodynamics* **1980**, *16*, 66–69.
36. Willers, B.; Eckert, S.; Michel, U.; Haase, I.; Zouhar, G. The columnar-to-equiaxed transition in Pb-Sn alloys affected by electromagnetically driven convection. *Mater. Sci. Eng. A* **2005**, *402*, 55–65. [\[CrossRef\]](#)
37. Dolinsky, Y.; Elperin, T. Thermodynamics of phase transitions in current-carrying conductors. *Phys. Rev. B* **1993**, *47*, 14778–14785. [\[CrossRef\]](#) [\[PubMed\]](#)
38. Zhang, X.F.; Lu, W.J.; Qin, R.S. Removal of MnS inclusions in molten steel using electropulsing. *Scr. Mater.* **2013**, *69*, 453–456. [\[CrossRef\]](#)
39. Wang, X.L.; Wang, Y.B.; Wang, Y.M.; Wang, B.Q.; Guo, J.D. Oriented nanotwins induced by electric current pulses in Cu-Zn alloy. *Appl. Phys. Lett.* **2007**, *91*, 163112. [\[CrossRef\]](#)
40. Leenov, D.; Kolin, A. Theory of electromagnetophoresis. I. Magnetohydrodynamic forces experienced by spherical and symmetrically oriented cylindrical particles. *J. Chem. Phys.* **1954**, *22*, 683–688. [\[CrossRef\]](#)
41. Gong, Y.; Luo, J.; Jing, J.; Xia, Z.; Zhai, Q. Structure refinement of pure aluminum by pulse magneto-oscillation. *Mater. Sci. Eng. A* **2008**, *497*, 147–152. [\[CrossRef\]](#)

42. Sun, M.; Geng, H.; Bian, X.; Liu, Y. Abnormal changes in aluminum viscosity and its relationship with the microstructure of melts. *Acta Met. Sin.* **2000**, *36*, 1134–1138.
43. Sklyarchuk, V.; Plevachuk, Y.; Yakymovych, A.; Eckert, S.; Gerbeth, G.; Eigenfeld, K. Structure sensitive properties of liquid Al-Si alloys. *Int. J. Thermophys.* **2009**, *30*, 1400–1410. [[CrossRef](#)]
44. Hunt, J.D. Steady state columnar and equiaxed growth of dendrites and eutectic. *Mater. Sci. Eng.* **1984**, *65*, 75–83. [[CrossRef](#)]
45. Martorano, M.A.; Beckermann, C.; Gandin, C.A. A solutal interaction mechanism for the columnar-to-equiaxed transition in alloy solidification. *Met. Mater. Trans. A* **2003**, *34*, 1657–1674. [[CrossRef](#)]
46. Brandt, R.; Neuer, G. Electrical resistivity and thermal conductivity of pure aluminum and aluminum alloys up to and above the melting temperature. *Int. J. Thermophys.* **2007**, *28*, 1429–1446. [[CrossRef](#)]



© 2019 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).