Investigation of refining mechanism in pure Al under pulsed magnetic field

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Abstract
The application of pulsed magnetic field can refine the macrostructure of metal effectively in solidification process. This paper presents experimental and numerical investigations on the refining mechanism of PMF on the solidification structure of pure aluminum. Forced convection induced by PMF was applied to the pure Al to investigate its effects on the grain refinement. Experiment results indicated that the forced convection promoted a thermal homogenization in the melt which is beneficial to the survival and growth of the nucleus and resulted in a significant refinement in the pure Al.

Keywords: Macrostructure; Force convection; Grain refinement; Aluminum

Introduction
Grain size is one of the significant factors that affect the processing and mechanical properties of material [1, 2]. Many methods can be used to refine the grain of alloy. Apart from addition of particles which can act as substrates for heterogeneous nucleation [3, 4], applying ultrasonic wave or electrical current, or magnetic field during solidification process to enhance grain nucleation and control grain growth [5]. A number of mechanisms for the grain refinement or CET under electromagnetic field have been proposed, such the fragmentation of dendrites induced due to the circulation of solute rich inter-dendritic liquid [6], the separate of dendrite fragments from the wall and mushy zone [7]. However, the influence of impurity element on the grain refinement has not clarity up to now.

In the research, the solidification structure evolution of commercial-purity aluminum and high-purity aluminum with a forced convection induced by PMF during the unidirectional solidification process was investigated for an in-depth understanding of the influence both the forced convection and the impurity elements, and the corresponding mechanism was discussed.

Experimental
Fig. 1 shows a schematic view of the experimental setup which consists of a straight solenoid connected with a self-prepared power which includes a capacitor bank of 1000 μF, temperature measurement system, cylindrical graphite mold. All experiments used a pulse frequency of 10 Hz, a half sinusoidal waveform with a peak voltage range between 0 V and 400 V and the internal was 100 V. Commercial-purity (CP) aluminum (99.7 wt. %) and high-purity aluminum (99.997 wt. %) were charged and held in a resistance furnace for 30 min at temperature 100 K higher than the melting point. Furthermore, the pouring temperature for each experiment was fixed 60 K higher than the melting point and the mold temperature was maintained at 973 K before casting. The solidification process was rendered a near isothermal condition by use of thermal insulating blanket on the lateral and bottom surfaces of the mold. The liquid melt was poured into graphite mold with an inner diameter of 45 mm and height of 100 mm which was placed on a water-cooled copper mold for 30 s, and then transferred into the PMF immediately. The K-type thermocouples were arranged vertically along the axis of the specimen to monitor the distribution of temperature during the solidification process.

The specimens were sectioned longitudinally along the mid-plane and then grounded on SiC papers and polished to remove any scratches for metallographic analysis. The etching reagent used to reveal the macrostructure was a solution of 75 ml HCl, 25 ml HNO₃ and 5 ml HF. A CanoScan (LiDE120) was employed to obtain the photographs of the macrostructure. Quantitative measurements of the grain size were determined by the mean linear intercept method using Nano Measurer software package.
Result and Discussion

Fig. 2 is an image sequence map to show the macrostructure of the commercial-purity aluminum solidified on the water-cooled copper mold for 30 s before the PMF treatment with different excitation voltages. It can be seen that compared with the solidification structure in the absence of PMF at the bottom of the specimen, a columnar-to-equiaxed transition occurs in the upper portion of the specimen and the growth of the columnar dendrites is suppressed. By combination of Fig. 2 (a)-(d), it should be mentioned that the application of PMF during the solidification process can generate the isotropic fine equiaxed grains and the average grains size decreases with increasing the excitation voltages.

Fig. 2 The macrostructure of commercial-purity aluminum solidified on the water-cooled copper for 30 s before PMF treatment with different excitation voltages: (a) 100 V. (b) 200 V. (c) 300 V. (d) 400 V

The representative solidification macrostructure with same processing conditions are shown in Fig. 3. As can be seen from Fig. 3, a significant discrepancy exists in the solidification structure between the commercial-purity aluminum and high-purity aluminum. Fig.3 a shows a dominance of columnar grains growing opposite to the direction of heat flow and there are no fine equiaxed grains along the longitudinal section of the specimen under the PMF which the excitation voltage is 100V. With increasing the excitation voltage, fine equiaxed begin to appear in the upper portion of the specimen but its grains size is larger than the commercial-purity aluminum at the same solidification conditions.

In the convention casting process for the case in the absence of the PMF, the heat dissipation of the liquid melt near the wall of the graphite mold is fast during the solidification. Upon initiation of cooling, a certain undercooling is establishes in the liquid, and the grains begin to nucleate first at the wall where the temperature is the lowest. The already nucleated grains continue grow opposite the direction of heat flow. However, when the PMF is imposed to the liquid, a Lorentz force will be generated by the coupling between the induction current and magnetic field which caused a forced convection in the liquid. Hence, a large number already nucleated grains originate from fragmentation both at the walls and the front of the columnar crystal will advec ted into the bulk melt. The schematic sketch of refinement mechanism under PMF can be illustrated as shown in Fig. 4.
In order to exactly illustrate the refinement mechanism of pure aluminum under PMF, the distribution of two major impurity elements Fe and Si between the commercial-purity and the high-purity was experimentally determined by electron probe microanalysis (EPMA).

As can be seen in Fig. 5, compared with high-purity aluminum, the distribution of Fe element on the grain boundary is successive. According to the Al-Fe binary alloy diagrams, the intermetallic compound Al$_3$Fe formed at the temperature of 928 K. Due to the magnetic oscillation caused by the PMF, the Al$_3$Fe with low melting point will be deformed and bent, which will destroy the solidified shell. The fragmentation of solidified shell at the wall are advected into bulk melt by the convection flow.

Fig. 3 The macrostructure of high-purity aluminum solidified on the water-cooled copper for 30 s before PMF treatment with different excitation voltages: (a) 100 V, (b) 200 V, (c) 300 V, (d) 400 V

Fig. 4 The schematic sketch of refinement mechanism under PMF: (a) The distribution of flow field under PMF (b) The schematic sketch of nucleus multiplication

Fig. 5 The distribution of impurity elements in commercial-purity aluminum and high-purity aluminum: (a)-(c) Commercial-purity aluminum. (d)-(f) High-purity aluminum.
Submission
In the present study, the refinement mechanism of both commercial-purity aluminum and high-purity aluminum was investigated. The results show that for the case where the PMF was applied, a columnar-to-equiaxed transition occurs in the upper of the melt. Furthermore, compared with the high-purity aluminum, due to the distribution of Fe element on the grain boundary, the solidification structure of commercial-purity is smaller. It is indicated the impurity elements play an important role in the refinement of pure aluminum.

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References
1. M.A. Easton, M. Qian, A. Prasad, D.H. Stjohn, Current Opinion in Solid State & Materials Science, 20 (2016) 13-24.
2. N. Wang, Z. Wang, K.T. Aust, U. Erb, Acta Metallurgica Et Materialia, 43 (1995) 519-528.
3. Y. Xu, D. Casari, Q. Du, R.H. Mathiesen, L. Arnberg, Y. Li, Acta Materialia, 140 (2017).
4. Y. Xu, D. Casari, R.H. Mathiesen, Y. Li, Acta Materialia, 149 (2018) 312-325.
5. D. Räbiger, Y. Zhang, V. Galindo, S. Franke, B. Willers, S. Eckert, Acta Materialia, 79 (2014) 327-338.
6. E. Liotti, A. Lui, R. Vincent, S. Kumar, Z. Guo, T. Connolley, I.P. Dolbnya, M. Hart, L. Arnberg, R.H. Mathiesen, Acta Materialia, 70 (2014) 228-239.
7. X. Liao, Q. Zhai, J. Luo, W. Chen, Y. Gong, Acta Materialia, 55 (2007) 3103-3109.