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ON NUCLEATION TEMPERATURE OF PURE ALUMINUM IN MAGNETIC FIELDS

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Abstract—Solidification of pure aluminum without and with a magnetic field has been investigated by differential thermal analysis (DTA). DTA curves showed that the nucleation temperature of pure aluminum was decreased as a magnetic field strength increased although its melting process was almost not influenced. The nucleation suppression could be attributed to the increase of the solid-liquid interfacial energy which might originate from more orderly arrangement of atoms on the solid-liquid interface upon applying a magnetic field.

1. INTRODUCTION

High magnetic field (HMF) has been widely applied to materials processing with the development of magnet technology, and a new interdisciplinary science, electromagnetic processing of materials (EMP), has come into being and flourished. Some novel phenomena in HMF has been found in sequence, e.g., levitation [1, 2], orientation [3, 4], change of phase transformation points [5, 6], magnetic interaction [7]. Among those researches, phase transformations of ferromagnetic substances were widely studied [8, 9]. Nevertheless, it is generally believed that a magnetic field with the order of 10 T has no obvious effect on phase transformations of non-magnetic materials in thermodynamics and thus the work of this aspect has not been paid much attention. However, there have been reports showing that the non-magnetic materials processing could be also influenced by a magnetic field via various actions, e.g., magnetic force, magnetic

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energy, during phase transformations whose kinetic or thermodynamic behaviors might be regulated, and further their final structures and properties could be altered. Hence, it is necessary to extend to investigate phase transformations of non-magnetic substances in order to deeply understand the essence of changes in magnetic fields. Solidification of metals and alloys is one of the most common phase transformations in which nucleation plays a crucial role. Unfortunately, the research on the subject in a magnetic field is in much lack except that the solidification temperature of pure bismuth with diamagnetism was observed to increase in a magnetic field [10, 11].

Hence, for simplicity, the present work aims to investigate the effect of a magnetic field on nucleation of pure aluminum with paramagnetism with aid of the differential thermal analysis (DTA)apparatus. The DTA curves indicate that the melting of pure aluminum is almost not affected, but its solidification has been delayed in the present of magnetic fields. Further discussion on nucleation suppression in magnetic fields has been given.

2. EXPERIMENTAL DETAILS

The DTA apparatus was used to detect the solidification behaviors of pure aluminum with and without a magnetic field. Its configuration was described in detail in previous work [12]. In DTA runs, the DTA apparatus was placed in the core of the superconducting magnet (Oxford Instruments), and the pure aluminum (5N) sample with diameter of 4 mm and height of 4 mm in the alumina crucible was kept in the center of magnet, where a magnetic field was homogeneous, and its strength is maximum in the vertical direction. The axial distribution of magnetic fields is shown in Figure 1.

The samples were heated to 750°C at the heating rate of 10°C/min, hold for 20 min and then cooled to the room temperature at the cooling rate of −5°C/min. High pure argon was passed into the furnace in order to reduce the oxidation of the samples. Figure 2 shows DTA curves for a sample cycled in the process of heating and cooling without a magnetic field (the ordinate axes of DTA curves denotes the temperature difference between a sample and a reference, hence, tick labels in the y-axis would be routinely concealed, and only a scale bar is shown, the same hereinafter). Generally, the nucleation temperature T_n gradually decreases with the increase of running cycles. Thus, the cycle tests for every sample were performed three to six times in order to obtain a limited nucleation temperature in specific conditions. DTA curves with the lowest nucleation temperature T_n, namely, maximum undercoolings $\Delta T = (T_m - T_n)$, obtained in different magnetic fields

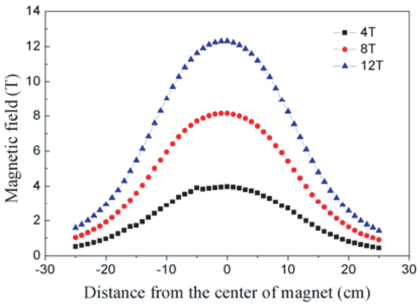

Figure 1. Axial distribution of magnetic fields with maximun strength of 4 T, 8 T, 12 T in the center respectively in the cylindrical core of the magnet.

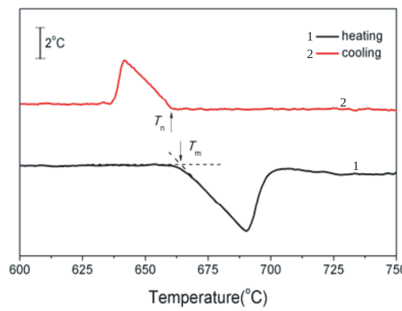

Figure 2. DTA curves for a test cycle of heating and cooling without a magnetic field. T_m and T_n denote the melting point and the nucleation temperature, respectively.

were compared in order to observe the effect of magnetic fields on solidificatoin behaviors of pure aluminum, where T_m is the melting point of pure aluminum attained by tangent extrapolation. It is worth noting that DTA curves have been calibrated by pure metals [11].

3. RESULTS AND DISCUSSION

Figure 3 shows DTA curves for bulk aluminum during the cooling in magnetic fields of 0 T, 4 T, 8 T, 12 T. It is distinctly observed that the nucleation temperature T_n shifts to a lower temperature as the magnetic field increases. The nucleation temperature T_n is about 660.2°C without a magnetic field, which is nearly approaching the equilibrium melting temperature 660.452°C. Nevertheless, the nucleation temperature T_n decreases to 649.7°C, 646.5°C, 638.6°C in the presence of magnetic fields of 4 T, 8 T and 12 T respectively even though the same experimental conditions are ensured except a magnetic field. Therefore, the current results indicate that the nucleation of pure aluminum is suppressed by magnetic fields, moreover, the higher the magnetic field, the more obvious the suppression. It follows that the reduction of nucleation temperature implies that certain factors, e.g., thermodynamic or dynamic parameters, influencing nucleation have been changed by the action of a magnetic field.

As we know, when a nucleus of critical size is formed from the undercooled liquid phase, nucleation into a crystallographic phase is characterized by using an activation energy ΔG^*. According to the classical nucleation theory, the activation energy ΔG^* [13] can be written as following:

$$\Delta G^* = \frac{16\pi\gamma_{sl}^3}{3(\Delta G)^2} \cdot f(\theta) \tag{1}$$

where $f(\theta)$ is a catalytic factor in the case of heterogeneous nucleation, ΔG the difference in free energy between liquid and solid phases and γ_{sl} the interfacial energy. Of course, the nucleation of bulk pure aluminum, without a doubt, was heterogeneous regardless of a magnetic field in the present experiments due to the limit of experiment conditions. However, the conclusion from the DTA experiments are not altered if heterogeneous nucleation is supposed provided that the catalytic factor in Equation (1) has the same value for all phases under consideration. The present discussion, therefore, is based on the assumption of homogeneous nucleation.

It is well known that the free energy difference ΔG is a driving force for nucleation while the nucleation barrier comes from the interfacial energy γ_{sl}. For paramagnetic pure aluminum, the magnetic Gibbs free energy induced by a magnetic field with the order of 100 T is extremely small [14], thus one should not expect that thermodynamic parameters, such as melting point, are influenced in a magnetic field with the order of 10 T. Actually, melting curves in Figure 4 evidently demonstrate that melting temperatures T_m of pure aluminum without and with 12 T magnetic field are equal for the experimental error. Similarly, a magnetic field has no effect on a driving force, namely, ΔG for nucleation as well.

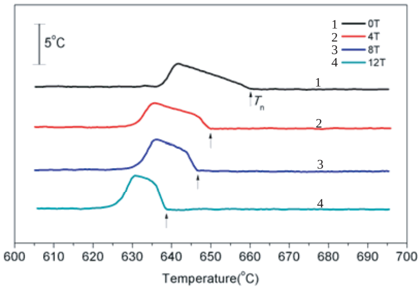

Figure 3. DTA curves for solidification of pure aluminum at the cooling rate of -5°C/min in various magnetic fields.

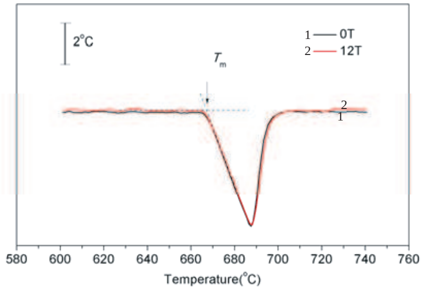

Figure 4. Melting curves of pure aluminum at the heating rate of 10°C/min with and without a 12 T magnetic field.

Recently, Mondal et al. [15] proposed a new model which predicted the temperature and structure dependency of solid-liquid interfacial energy. According to their model, the interfacial energy γ_{sl} gradually increased with undercooling. It reached a maximum value at some intermediate temperature and then decreased on further increase in undercooling. In the present experimental results, the nucleation temperature falls between the intermediate temperature and melting point. It is learned that γ_{sl} increases with a magnetic field strength. Or stated differently, a magnetic field influences the solid-liquid interfacial energy in the process of nucleation.

In fact, we observed the change of surface morphologies of post-treated samples in various conditions, as indicated in Figure 5, and found that the surface shapes vary from bumpy surface without a magnetic field to convex surfaces in magnetic fields, which means that the surface tension of liquid aluminum increases in magnetic fields. According to Vinet's report [16], the ratio of interfacial and surface energy was a dimensionless number, and the change of surface tension implied the variation of the interfacial energy. Consequently, it also provides some indirect evidence that the interfacial energy increases upon applying a magnetic field. Fujimura et al. [17] applied the surface-wave resonance method to measure the surface tension of water in HMF and found that the surface tension increased linearly with the square of the magnetic field. They attributed it to two possible factors: One was that a magnetic field stabilized the hydrogen bonds of water, and the other was that the Lorentz force dampened the surface excitation of ripplons. Whereas pure aluminum is entirely different from water in structure, the similar explanation cannot be applied to the current cases.

One possible explanation for the present results is that a magnetic field modifies the arrangement of atoms on the solid-liquid interface. The schematic illustration of the solid-liquid interface with and without a magnetic field was displayed in Figure 6. The arrangement of liquid and crystal atoms is staggered packing on the interface without a magnetic field and leads to a smaller value of the interfacial energy due to similarity in structure between the liquid and crystal [18]. Nevertheless, the disorder state will be improved in a magnetic field because moving liquid atoms near the interface in a magnetic field will be subjected to the Lorentz force which makes unstable or metastable atoms on the interface migrate to a stable position. Therefore, the solid-liquid interface will be more planar, and correspondingly the interfacial energy will be larger. It is worth noting that we attribute the activation of nucleation for pure bismuth in magnetic fields to the reduction of kinetic barrier of nucleation [11]. From this

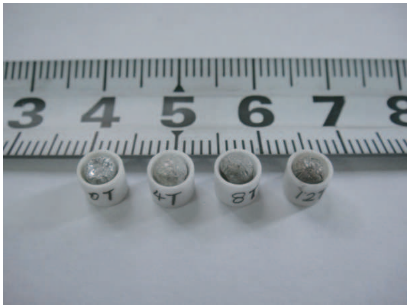

Figure 5. The change of free surfaces of samples in DTA runs in magnetic fields of 0 T, 4 T, 8 T, 12 T marked with signs of 0 T, 4 T, 8 T, 12 T on the sides of crucibles.

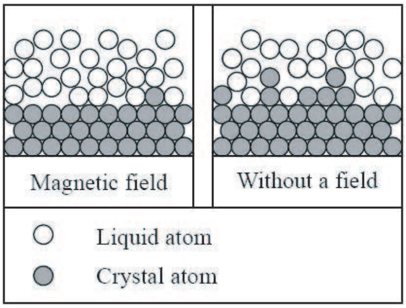

Figure 6. Schematic illustration of the solid-liquid interfaces with and without a magnetic field.

point of view, it is more difficult for liquid Al atoms to attach to a nucleus in the case of disturbance of the Lorentz force, and it leads to the increase of kinetic barrier which embodies in the increase of undercooling. Nevertheless, whether or not the difference of nucleation for paramagnetic pure aluminum and diamagnetic pure bismuth is caused by magnetism difference is unknown. Thus, further theoretic analysis is needed to explain the phenomena.

4. CONCLUSION

The current DTA experiment gave primary results that the nucleation temperature of pure aluminum was decreased as the magnetic field strength increased though the melting temperatures were equal regardless of a magnetic field. The probable reason for nucleation suppression is that the increase of nucleus-liquid interfacial energy is induced by a magnetic field which makes the arrangement of atoms on the interface more orderly than that without a field. Certainly, further experiments and theoretical analysis will be needed in order to clarify nucleation process in a magnetic field.

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