Solidified Structure Control of Metallic Materials by Static High Magnetic Fields

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Recently, the studies on the effects of high magnetic fields on solidification processes have been paid much attention both from the fundamental and applied points of view. With the aid of the enhanced Lorentz force and magnetization effect caused by the remarkably increased magnetic field intensity, several interesting phenomena, such as the control of fluid flow and particle migration in a melt, crystal orientation, and phase alignment, have been obtained. Moreover, the magnetic force induced by the interaction of magnetization and high magnetic field gradient has been evidenced to show significant effects on the microstructure evolution of alloys. In this paper, the recent development of the control of the solidification process by high magnetic fields is reviewed from the view point of uniform magnetic fields and magnetic field gradients.

KEY WORDS: high magnetic field; solidification process; fluid flow; crystal orientation; phase alignment; structural evolution.

1. Introduction

The solidified structures of metals and alloys play a key role for the subsequent heat treatments and even the final performances of these materials. Their control is therefore essential for improving and tailoring the properties of these materials. One possible processing route by which microstructure control can be accomplished is solidification under high magnetic field conditions.

Experimentally, there are mainly two ways can have such control. The first one is through the Lorentz force resulting from the interaction of a uniform magnetic field and an induced or imposed current in electroconducting fluids. It has long been recognized that the Lorentz force can significantly suppress the thermal convection in a melt. In addition, such suppression effect has now been realized on the local flows and the movement of particles in a liquid matrix by increasing the magnetic field intensity. The second way is to use the magnetization. Usually, during the solidification processes materials show a weak response to the applied magnetic field even for ferromagnetic ones, because their magnetic susceptibilities strongly decrease with temperature increasing, especially near their melting temperatures. However, with the aid of the enhanced magnetization effect induced by the field with enough high intensity, several interesting phenomena, such as crystal orientation and phase alignment, have been realized. Recently, the study has been extended to high magnetic field gradient. Strongly enhanced magnetic force induced by the interaction of the magnetization and magnetic field gradient has been evidenced to show significant effects on the microstructural evolution of some alloys.

This paper reviews recent attempts at the control of solidification process of metallic materials using high magnetic fields. However, some emphasis on attempts of our group was put, particularly in the selection of examples which were more readily available.

2. Control of Fluid Flow in Uniform Magnetic Fields

2.1. Suppression Effect

In 1966, on the basis of the theoretical analysis of previous studies, an experimental attempt at elimination of solute banding in indium antimonide crystals by a magnetic field was made. The success of such attempt led to the novel insight that the use of a high uniform magnetic field can significantly suppress the thermo-solutal flow in a conducting melt in terms of Lorentz force. Following this attempt, numerous studies were carried out on understanding the mechanism of such effect and its operating parameters using both the numerical simulation and experimental methods. This understanding led to various techniques to improve the quality of the crystal which has an electroconducting or partially electroconducting nature using a magnetic field.

Recently, the studies have been extended to flow-relative processes such as the movement of particles and the distribution of solute elements in alloys during solidification. The effect of a vertical static magnetic field on the movement of particles in the conducting liquid matrix was investigated...
through the observations of their microstructures. The movement of the particles was remarkably restrained by the Lorentz forces either acting on the particles or on the convection in the liquid matrix. More homogeneous microstructures of some monotectic and eutectic systems were thus obtained. Theoretical analysis was also carried out to elucidate such effect. By considering the fact that Lorentz force still has a vertical component with the imposed magnetic field and exhibits a similar effect to that of viscous resistance, the terminal velocity of the particles was deduced in terms of the Hartman number. Such quantitative description has been used to characterize the solidification processes of Cu–Pb monotectic and Al–Ni eutectic systems in high magnetic fields, during which the migration of the Cu-rich drops and primary Al$_3$Ni particles was strongly affected by the Lorentz force respectively. A high magnetic field was also found to reduce the local flows around the solidifying front caused by solute rejection and thermocapillary effect. As shown in Fig. 1, by applying a high magnetic field to the unidirectional solidification of an Al–10at%In hypermonotectic alloy, an aligned rod structure, which is commonly observed in the eutectic alloys, is produced.

Furthermore, the effect of high magnetic fields on the distribution of solute elements of some alloys was investigated in some literatures. In some Al-rich alloys solidified in high magnetic fields, the concentrations of Cu and Mg solutes in α-Al grains were increased and decreased respectively, while their gravity segregation was both obviously reduced. These changes in the distribution of the solute elements were produced by the Lorentz force and depended on the physical properties of the elements such as density and conductivity. A similar decrease of Cu concentration as well as its uniform micro distribution was also observed in a Fe–3.95%C–0.44Cu% alloy solidified in a high magnetic field. These findings suggest the possibility of controlling the distribution of solute elements at micro-regions and the gravity segregation in alloys by imposing a uniform magnetic field.

2.2. Thermoelectromagnetic Effect

In ingot solidification, the application of a magnetic field brakes the convection of both macro- and micro-sopic in the liquid bulk. However, in directional solidification, the magnetic field may induce additional flow, i.e. thermoelectromagnetic convection (TEMC). In 1989, Gel’Igat and Gorbunov reported that during the growth of InSb crystals using a Czochralski method, the application of a magnetic field could cause a strong distortion of the crystals. They attributed this distortion to the TEMC induced by the interaction between the magnetic field and thermoelectric effect which occurred at the solid–liquid interface.

Following their work, some researchers carried out a series of directional solidification experiments in magnetic fields and numerical simulations to investigate the TEMC effect. The striation- and freckle-type segregations mainly observed in some semiconductor crystals and alloys respectively confirmed the existence of the TEMC at the growth interface and mushy region. For instance, the microstructure of Cu–Ag alloys shows a eutectic border at the lower part of the specimen without magnetic field (Fig. 2(a)), but at the upper part of the specimen with a magnetic field (Fig. 2(b)), indicating that the convection has been inverted due to the TEMC. Recently, Li et al. investigated the influence of a high magnetic field on the morphological instability of the growth interface of binary alloys during directional solidification. The obtained results indicated that the high magnetic field could induce the change in the morphological instability of the growth interface in terms of the TEMC.

3. Crystal Orientation in Uniform Magnetic Field

In 1981, from the structural observations, Mikelson and Karklin reported that in Al–Cu, Cd–Zn and Al–Ni alloys which were solidified in 0.5–1 T magnetic fields with a low cooling rate, the primary crystals of both Al–Cu and Cd–Zn alloys were oriented with their longer axes along the field direction, while the primary Al$_3$Ni platelets of Al–Ni alloy were oriented with their planes perpendicular to the field direction. They then explained such orientation using a rotation to orientation mechanism that a crystal is rotated to an angle by a torque due to the magnetic anisotropy of the crystal to minimize the system energy. Such mechanism has been adopted by following researchers to be the basis of the magnetic orientation theory.

In the present paper, two types of orientations were distinguished: the crystal orientation (or texture) and the phase alignment. The former is the orientation in lattice or in crystallography which is generally determined by diffraction techniques, while the later is the orientation in morphology of macro scale which is generally illustrated by structure observation. In some cases, these two orientations can be correlated, such as fibres in fibre-reinforced ceramics. However, in many other cases, they are not.
instance, in a rolled cubic metal the grains are elongated to form an aligned structure but do not necessarily show crystal orientation. Although the details for evaluating the orientation were not provided, the microstructural changes observed by Mikelson and Karklin can then be classified as phase alignment. In fact, the Al$_3$Ni crystals have been evidenced to be oriented in a magnetic field up to 4.4 T with their c-axes parallel to the imposed magnetic field by other researchers using X-ray diffraction techniques.

In 1991, de Rango et al.\(^{37}\) successfully fabricated oriented YBa$_2$Cu$_3$O$_7$ materials at high temperature by solidification in a 5 T magnetic field and proven the possibility for in-situ producing oriented structure during solidification in a high magnetic field. Following their study, the oriented structures were obtained in a series of alloy systems such as Sm–Co\(^{38,39}\), Bi–Mn\(^{40,41–43}\), Bi–Zn\(^{44}\), Al–Fe\(^{45}\), Mn–Sb\(^{46,47}\) and Al–Ni\(^{23,36,48}\) alloys and some high-temperature superconductor materials\(^{49–51}\) by solidification in a high magnetic field. Figure 3 shows c-axes pole figure measured on a face of a Sm–83.4at%Co solidified in a 5 T magnetic field which is perpendicular to the field direction.\(^{39}\)

The figure reveals that c-axes of SmCo$_5$ grains deviated by no more than 10° from the magnetic field direction. Such X-ray diffraction pole figure was confirmed by the magnetization curves obtained from the same alloy (Fig. 3, insert) which show a c-axes orientation along the field direction. On the basis of these experimental observations, theoretical analyses were carried out to characterize the conditions needed by the magnetic crystal orientation and to discuss its affecting parameters.\(^{52–55}\) The principle of the magnetic crystal orientation has been reviewed by Asai.\(^{56}\)

4. Phase Alignment in Uniform Magnetic Field

As well as the magnetic crystal orientation, macrostructural alignment of phases in materials was also received considerable attention. By applying a high magnetic field to a slow solidification process, the primary phase of an alloy can be aligned either parallel or perpendicular to the magnetic field direction. Such alignment of primary phase has been realized in a series of binary and ternary systems such as Al–Ni,\(^ {23,36,48,57}\) Mn–Bi,\(^ {40–42}\) Mn–Sb,\(^ {46,47,50}\) Al–Fe\(^ {55}\) and Al–Si–Fe\(^ {43}\) alloys. Figure 4 shows the typical microstructures of Al–11mass%Si–2mass%Fe alloy solidified at 0 T and 5 T.\(^ {51}\) The application of the magnetic field is observed to cause the alignment of the intermetallic compound Al–9mass%Si–1mass%Fe phase with its long axis perpendicular to the magnetic field direction (Fig. 4(b)).

Although different mechanisms were proposed to explain the alignment phenomenon, these mechanisms were totally linked to the magnetic crystal orientation, because these aligned phases were usually accompanied by the crystal orientation. For instance, the primary MnSb in a hypoeutectic Mn–Sb alloy solidified from a semi-solid state in a high magnetic field was not only aligned along the magnetic field, but also oriented with its c axis perpendicular to the magnetic field.\(^ {47}\) For some systems consisting of a liquid matrix and foreign particles of large size with shape anisotropy, such as an Al–Si melt with rod-like TiAl$_3$ particles suspending in it,\(^ {50}\) the alignment of these particles in a high magnetic field can be directly attributed to the rotation of them driven by the torque induced by the interaction of their magnetic anisotropy and the imposed magnetic field. For those alloys\(^ {23,36,41,42,45–48,57,50}\) in which the aligned phases (usually the primary phases) formed during solidification processes and their nucleation and growth were strongly affected by the solidification conditions, the phase alignment resulted from the combination of the crystal orientation and other crystal growth-relative factors such as solute distribution, preferred crystallographic direction, heat flow direction, interaction between crystals, etc. Which factor would be dominant was dependent on the alloy system and morphology of the aligned phases. For instance, solute distribution was dominant for the alignment of the primary Al$_3$Ni in Al–Ni alloy\(^ {23}\), preferred crystallographic and heat flow directions were dominant for the alignment of the primary MnSb dendrites in Mn–Sb alloys\(^ {56}\), while magnetic dipole interaction between crystals were dominant for the primary MnSb and MnBi grains in Mn–Sb\(^ {57}\) and Bi–Mn\(^ {49,42}\) alloys, respectively. Depending on the dominant factors, the alignment degree of these primary phases were strongly affected by the alloy composition\(^ {48,53}\) and the time during which the primary phase and liquid matrix coex-
The distribution of alloying elements and their product phases in the melt during solidification processes is very important for the performances of alloys, because it can directly affect the final microstructures. Recently, some attempts have been made at developing novel processes using high magnetic field gradients to control the distribution of alloying elements and their product phases in the melt during solidification processes. When a binary system consisting of a liquid matrix and dispersed particles is subjected to a vertical magnetic field gradient, the particles will be acted upon by a magnetic force in the direction along (paramagnetic) or opposite (diamagnetic) to the field gradient. Although the Lorentz force which can strongly suppress the convection in the melt could eliminate the effect of the convection on the migration of particles, the migration of these particles will be magnetic force-controlled if this magnetic force is high enough. Therefore, the control of the distribution of these particles in the liquid can be realized.

5. Control of the Distribution of Particles and Solute Elements by Magnetic Field Gradients

The distribution of alloying elements and their product phases in the melt during solidification processes is very important for the performances of alloys, because it can directly affect the final microstructures. Recently, some attempts have been made at developing novel processes using high magnetic field gradients to control the distribution of alloying elements and their product phases in the melt during solidification processes. When a binary system consisting of a liquid matrix and dispersed particles is subjected to a vertical magnetic field gradient, the particles will be acted upon by a magnetic force in the direction along (paramagnetic) or opposite (diamagnetic) to the field gradient. Although the Lorentz force which can strongly suppress the convection in the melt could eliminate the effect of the convection on the migration of particles, the migration of these particles will be magnetic force-controlled if this magnetic force is high enough. Therefore, the control of the distribution of these particles in the liquid can be realized.

5.1. Particle Distribution

Some experimental studies on the microstructure evolution caused by the change in the distribution of particle phases in high magnetic field gradients were carried out, such phase can be either foreign or primary ones. Alloys were solidified either from a semi-solid state or from a melting state. Liu et al. and Wang et al. heated a hypo-eutectic Mn–Sb alloy to a semi-solid state to produce a mixture of a liquid matrix and dispersing particles. After holding the mixture for a period of time in a high magnetic field gradient, the distribution of the MnSb particles in the alloy were successfully controlled. In the obtained alloys, the primary MnSb particles were obviously segregated in one side of the alloys. The positions where these particles located depended on the field gradient direction. Furthermore, the volume fraction of the MnSb particles exhibited a continuous decrease or increase, i.e. a graded distribution, along the depth from the lower surface in the alloys. Using a similar experimental procedure but a mixture of Al–Si melt and TiAl₃ particles to be the starting material, the strong segregation of the TiAl₃ particles in the lower part of the alloys in the absence of the magnetic field was substituted by a uniform distribution of these particles in the presence of a negative magnetic field gradient. Other researchers solidified several hypereutectic Al–Si and Bi–Mn, in a high magnetic field gradient from a melting state and also realized the control of the distribution of the primary phases of these alloys. The above-mentioned control was affected by the direction of the magnetic field gradient and the $[BdB/dz]$ value. This process has been proposed as an effective process route for fabricating functionally graded material.

5.2. Solute Element Distribution

Recently, the possibility of directly controlling the distribution of solute elements in alloys during solidification process using high magnetic field gradient was investigated by Wang et al. and Liu et al. Mn–Sb alloy was chosen to investigate because Mn has the largest magnetic susceptibility under melting conditions and can exhibit a strong response to the magnetic field gradient. A series of Mn–Sb alloys were solidified in various magnetic field gradients. It was found that a high magnetic field gradient could control the distribution of solute element in the alloys during solidification processes and therefore resulted in microstructural changes. In the presence of a high magnetic field gradient, for alloys of near eutectic composition, the coexistence of both primary MnSb and Sb phases in one specimen with a continuous change in volume fraction was formed. The positions where these primary phases located depended on the direction of field gradient. While for the alloys of far from eutectic compositions, the primary MnSb was segregated in one side of the specimens with a continuous change in volume fraction. For instance, the microstructures of Mn–89.7wt% Sb alloys solidified in high magnetic field gradients indicate the separation of both primary phases and their graded distributions, as shown in Fig. 5. By comparing the structural changes of alloys with various compositions and the theoretical analysis, the authors argued that the control of the solute element distribution in Mn–Sb alloys was realized through the magnetic buoyancy force. Such force originated from the difference
in the magnetic susceptibility between Mn and Sb (the magnetic susceptibilities of Mn and Sb in the liquid state are 882.62×10⁻⁶ and -1.0048×10⁻⁶, respectively, and could drive Mn clusters to migrate in the melt. The effectiveness of this control depends on the alloy composition, specimen dimension, cooling rate, and the [BdBlez] value. In addition, Takeda et al. investigated the influence of high magnetic field gradient on the ionic concentration in the mixture of Dy(NO₃)₆H₂O and Sm(NO₃)₆H₂O aqueous solution. They found that high magnetic field gradients could induce the migration of paramagnetic ions in the aqueous solution if the solution was held in the field gradients for 336 h. The following theoretical consideration verified the occurrence of such magnetic force-induced migration. Although the mechanism about the change in the solute distribution during solidification processes in high magnetic field gradients still needs to be clarified by future research, the above studies indicate that it is possible to, in situ control the distribution of solute elements in alloys during solidification process and therefore to control their microstructures using high magnetic field gradient.

6. Conclusions

This review shows the progress of the application of high magnetic fields on solidification processes of metallic materials. High magnetic fields have been widely used to control both macro- and micro-sopic fluid flow and flow-relative processes such as the movement of particles and the distribution of solute elements in alloys during solidification in term of Lorentz force, as well as induce thermoelectromagnetic convection (TEMC) in directional solidification on the basis of thermoelectric effects. The enhanced magnetization effect caused by the remarkably increased magnetic field intensity can induce the orientation of crystals with magnetic anisotropy at high temperature. The combination of magnetic crystal orientation with other crystal growth-relative factors such as solute distribution, preferred crystallographic direction, heat flow direction, interaction between crystals causes phase alignment. In addition, high magnetic field gradient is realized to be a powerful tool to control the distribution of particles and solute elements in alloys during solidification processes.

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