



# In-situ fabrication of graded material with the application of a horizontal magnetic field during directional solidification

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## ABSTRACT

The present paper proposes a method of graded materials preparation under a low horizontal magnetic field during directional solidification. The external horizontal magnetic field was applied to the mushy zone of the directionally solidified Al-10 wt% Cu and Al-18 wt% Si samples, which was perpendicular to the growth direction. Experimental results demonstrate that the applied horizontal magnetic field produces a gradient size distribution of the primary  $\alpha$ (Al) grains in Al-10 wt% Cu alloys and a gradient fraction distribution of primary Si particles in Al-18 wt% Si alloys. 3D numerical simulations indicate that the formation of graded structure should be attributed to the migration of primary phase driven by the thermoelectric magnetic force (TEMF). The formation of segregation channel should be attributed to the TEM convection (TEMC) in the mushy zone. Further, in situ synchrotron X-ray radiography observations of the initial transient of directional solidification show that the primary  $\alpha$ (Al) phases were migrated approximately along the direction perpendicular to the magnetic field. These results suggest that the TEM effects can greatly change the growth of primary grains and the solidification structure, and the application of an external horizontal magnetic field would be beneficial to fabricate the graded material during directional solidification.

## 1. Introduction

The conception of functionally graded composite materials was first reported in 1972 [1]. The property gradient in the material is caused by a position-dependent chemical composition, microstructure or atomic order [2]. These continuous changes result in physical-property gradients with an absence of any mechanically weak junctions or interfaces. A gradual change in physical properties can be tailored to adapt materials to different applications and service environments. Therefore, in practice, these materials can be used where materials are required to perform different functions in different regions. This makes functionally graded composite materials preferable in many applications. Many practical techniques have been developed to prepare functionally graded materials, including laser cladding [3], centrifugal casting [4], powder metallurgy [5], vapor deposition [6] and three dimensional printing [7]. However, due to the traditional techniques are relatively expensive or complicated, they are still finite. Therefore, the researchers are desperately searching for new ways to fabricate the

functionally graded materials.

In the last decades, the application of an external high magnetic field (of the order of 10 T) to fabrication of graded materials has been investigated. Wang and Liu et al. [8,9] applied a high gradient magnetic field on the Mn–Sb alloys. Experimental results displayed that the bulk layered synthetic Mn–Sb composites were fabricated directly. Further, a metal-ceramics graded material has been fabricated by Sun et al. [10] by applying a strong magnetic field with a high gradient. Gao et al. [11] fabricated a  $Tb_{0.27}Dy_{0.73}Fe_{1.95}$  alloy with a magnetostrictive gradient by solidification under a high magnetic-field gradient. Above results indicated that the phase segregation was induced by an Archimedes magnetic force applied to the magnetic phases by a high gradient magnetic field.

As we know, it is not easy to obtain such a high magnetic field (of the order of 10 T) in industrial manufacture. Thus, the applied low horizontal magnetic field (less than 1 T) has attracted more attention and its effect on growth of metallic alloys during directional solidification has been studied extensively. It was determined that the applied

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low horizontal magnetic field during directional solidification of materials can significantly reduce the thermosolutal flow. During directional solidification in the cellular/dendritic regime, some interesting results were observed [12,13]. As an example, Tewari et al. [12] found that the 0.45 T horizontal magnetic field caused severe distortion in the cellular array morphology and the formation of freckles on the plane perpendicular to the magnetic field when a hypoeutectic Pb–Sn alloy was solidified vertically at very low growth speed. Alboussie et al. [13] also found that a 0.6 T horizontal magnetic field caused the formation of freckles in the directionally solidified Bi-60 wt% Sn alloys. They suggested that this kind of flow was induced by the interaction between the thermoelectric (TE) effects and the magnetic field.

Further, Lehmann et al. [14] gave some experimental results to prove the so-called thermoelectric magnetic convection (TEMC). Recently, the influence of a horizontal magnetic field on the growth of metallic alloys during directional solidification has been systematically reported [15–18]. The applied horizontal magnetic field modified the solid/liquid interface morphology [15,16], the cellular/dendritic growth observably [17] and the distribution of primary phase [18], due to the TEMC.

In the present study, we demonstrate that graded materials can be obtained via control of primary phase movement by the thermoelectric magnetic force (TEMF) during directional solidification and validate the method in two aluminum alloys (i.e. Al-10 wt% Cu and Al-18 wt% Si alloys) with different physical parameters of two primary phases. Further, the simplified geometry models (i.e. sphere; prolate spheroid) were applied for the numerical simulation. The TEM effects imposed on sphere geometry have been studied numerically in order to reveal the underlying mechanism that governs the migration and gradient distribution of primary phases.

## 2. Experimental

Al-10 wt% Cu and Al-18 wt% Si alloys were prepared by the high pure Al (4 N), Cu (3 N) and Si (4 N) by using a vacuum induction furnace. The molten alloy was electromagnetic stirred for 1 h, then casted to the rod-like sample with diameter of 3.5 mm and length of 150 mm by using suction casting technique.

Pre-cast Aluminum alloy samples (about 200 mm long) contained in high purity corundum crucibles (3.5-mm inner diameter) were remelted in vacuum and directionally solidified in a modified Bridgman apparatus by pull down the crucibles at various speeds with respect to the stationary furnace assembly. A schematic of the Bridgman apparatus under a horizontal magnetic field can be found in Ref. [15]. It was composed of a Bridgman apparatus, a growth velocity and temperature controller and a direct current electromagnet with a maximum value of 1.0 T. The temperature of Bridgman apparatus was controlled by a K-type thermocouple. The samples were etched by Keller etching solution and then examined by optical microscopy. Crystallographic characteristics were analyzed using scanning electron microscopes (SEM; HITACHI SU70) equipped with the EDAX's OIM EBSD Analysis System.

## 3. Experimental Results

Fig. 1 displays the longitudinal structures below the solid/liquid interface of the directionally solidified Al-10 wt% Cu and Al-18 wt% Si alloys without and with a 0.5 T magnetic field. It can be observed that the applied magnetic field changed the morphology and distribution of primary phase dramatically. For the Al-10 wt% Cu alloys, the microstructure shows the typical equiaxed structure without magnetic field. The applied magnetic field caused the size of equiaxed grains in the right region smaller than the one in the left region for the growth speed of 10  $\mu\text{m/s}$  (see Fig. 1a<sub>2</sub>). For a higher growth speed of 30  $\mu\text{m/s}$ , the effect of magnetic field on the primary phase becomes weak. For the Al-18 wt% Si alloys, the black primary Si particles were distributed uniformly in the homogeneous Al/Si eutectic matrix without the magnetic

field. When a 0.5 T horizontal magnetic field was applied, more Si particles were distributed at the right part than the one at the left part of the sample. However, a periodic structure consisting of large segregation channels appear. The channels contain almost silicon in the Al–Si alloys.

Fig. 2 presents the EBSD images for the samples in directionally solidified Al-10 wt% Cu alloys at the growth speed of 20  $\mu\text{m/s}$  and various magnetic fields. The horizontal magnetic field caused the formation of a graded material with the grain size decreasing from the left side of crucible to the right side. It is shown in Fig. 3a that the coarse primary Si phase was distributed in the Al/Si eutectic zone at position 1. Some Al-rich structures (see red dashed circle) were distributed in the Al/Si eutectic matrix at position 2 (Fig. 3b), while typical Al/Si eutectic structure at position 3 (Fig. 3c) and the Si-rich region at position 4 were found (Fig. 3d).

To measure the average primary  $\alpha(\text{Al})$  grain size, the longitudinal plane was divided vertically into 6 rectangles equally, and the linear intercept method was used to determine the average grain size in each part of the sample. That is to say, the length and width of the each rectangle are about 8.5 mm and 0.58 mm, respectively. The average primary  $\alpha(\text{Al})$  grain size of each sample in Fig. 2 was summarized in Fig. 4a. It clearly shows that the average grain size is about 500  $\mu\text{m}$  with the absence of magnetic field. With the applied magnetic field, the average grain size decreased gradually from the left side of crucible to the right side. Moreover, the average grain size in the left side of crucible increases with the increase of magnetic field intensity and the average grain size in the right side of crucible decreases with the increase of magnetic field intensity. The fraction of primary Si in Al/Si eutectics for each part of samples in Fig. 1d was calculated by using free software ImageJ and the results were given in Fig. 4b. The primary Si was distributed uniformly in the sample and the fraction is about 15% with the absence of magnetic field. However, when a horizontal magnetic field is applied, the fraction of primary Si reaches 30% in the right side of crucible and drops to 7.5% in the left side of crucible.

## 4. Theoretical Analysis and Discussion

Above experimental results indicate that the applied horizontal magnetic field causes two main effects: one is the gradually distributed primary phase along the horizontal direction; and the other is the formation of tilted segregated channels.

### 4.1. Deflection Mechanism

In the case of equiaxed solidification of Al–Cu alloy, experiments clearly show that the horizontal deflection of the grains leads to variation of the grain size along the horizontal direction and causes the formation of graded materials. The equiaxed primary grains are distributed gradually in the specimen. It may be attributed to thermoelectric effects which will be briefly described hereafter.

During directional solidification, due to the thermal gradient along solid/liquid interface together with the different thermo-physical properties, the Thomson-Seebeck effect will be produced, which results in TE currents flowing through both the solid and the melt. When a magnetic field is applied during directional solidification, two TEM effects can be induced: the TEMF acting directly on the primary grain [19,20] and the TEMC in the melt nearby [14,21,22] leading to hydrodynamic strains on the grain surface. The combination of the two previous TEM effects leads to a segregation of the primary grain in the vertical direction.

For a fixed grain, the basic equations governing the TEMC phenomena are as follows:

$$\vec{j} = -\sigma \vec{\nabla} E - \sigma S \vec{\nabla} T + \sigma \vec{u} \times \vec{B} \quad (1)$$

where  $\vec{j}$  is the electric current density,  $E$  is the electric scalar potential,

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