Acta Materialia 121 (2016) 240–256

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Effect of a high magnetic field on the growth of ternary Al-Cu-Ag alloys during directional solidification

Dafan Du [a], Yves Fautrelle [b], Zhongming Ren [a], Rene Moreau [b], Xi Li [a, b, *]

[a] State Key Laboratory of Advanced Special Steels, Shanghai University, Shanghai, 200072, PR China
[b] SIMAP-EPM-Madylam/G-INP/CNRS, PHELMA, BP 75, 38402, St Martin d'Heres Cedex, France

ARTICLE INFO

Article history:
Received 14 May 2016
Accepted 12 September 2016

Keywords:
Al-Cu-Ag alloy
High magnetic field
Solidification structure
Directional solidification
Electron backscatter diffraction (EBSD)

ABSTRACT

The influence of a high axial magnetic field (up to 6 T) on the solidification structure and crystallography in directionally solidified Al-21.5 wt%Cu-27 wt%Ag and Al-17.6 wt%Cu-42.2 wt%Ag ternary alloys has been systematically studied. Both alloys followed its own typical solidification processes: two-phase univariant eutectic growth ($L \rightarrow \alpha$-$Al+Al_2Cu$) and three-phase invariant eutectic growth ($L \rightarrow \alpha$-$Al+Al_2Cu+Ag_2Al$) for Al-21.5 wt%Cu-27 wt%Ag and Al-17.6 wt%Cu-42.2 wt%Ag, respectively. It is found that the solidification structure and the orientation relationship (OR) between eutectic phases are modified at lower growth speeds ($R \leq 3$ μm/s) under the magnetic field ($B \geq 2$ T). For univariant eutectic growth, the magnetic field refines eutectic cell and promotes the transform from cellular structure to planar structure, whereas for invariant eutectic growth, the magnetic field reduces the eutectic spacing and causes the formation of banded structure at lower growth speeds ($R \leq 0.8$ μm/s). The magnetic field enhances the richness of the Ag solute in the liquid ahead of the quenched liquid/solid interface. In addition, the magnetic field modifies the preferred OR between Al_2Cu and Ag_2Al eutectic phases at lower growth speeds ($R \leq 3$ μm/s) and induces the formation of the OR with higher interface energy. Modifications of the solidification structure and the OR under the magnetic field are attributed to the thermoelectric magnetic (TEM) effects and the changes in the interfacial energy caused by the magnetic field.

1. Introduction

A lot of commercial materials are multicomponent, multiphase alloys, whose properties are determined by the microstructure that develops during solidification and subsequent processing stages. Most research works on the fundamentals of solidification has been primarily developed from pure substances [1], and binary alloys exhibiting single-phase growth [2,3] (solid solution) and/or two-phase growth eutectic [4,5] and peritectic [6,7] class reactions *etc.* However, the solidification microstructure formation in multi-component alloys with three or more components has not been intensively understood, especially for cases where multiphase reactions occur along the solidification path of the alloy. Therefore, the evolution of solidification microstructures in the multicomponent, multiphase alloys has attracted pronounced academic interest. Among of them, the ternary Al-Cu-Ag alloy is an ideal model alloy due to a relatively low eutectic temperature and a roughly equal amount of each solid phase. The first experimental observations of ternary eutectic alloys, including Al-Cu-Ag alloy, were conducted by Cooksey and Hellawell [8] in 1967. McCartney and Hunt [9,10] investigated the Al-Cu-Ag ternary alloys with various compositions and different growth speeds during directional solidification. They proposed the models for the growth of a two phase eutectic as well as for single phase dendrites or cells in three component systems, which were well agreement with the experimental results. Wilde et al. [11] proposed a theoretical approach to describe the two-phase planar and lamellar coupled eutectic growth in the univariant growth of Al-Cu-Ag alloys. Recently, to characterize the morphology of Al-Cu-Ag alloys, experimental studies [12–15] show a wide range patterns, like the so-called "brick-like" structures and the effect of solidification parameters on the inter-lamellar spacing. Phase-field simulations [16–20] were performed to understand how surface energies and volume fractions influenced the microstructure pattern formation during three-dimensional directional solidification, which raised interesting questions regarding the correlation of microstructure

* Corresponding author. State Key Laboratory of Advanced Special Steels, Shanghai University, Shanghai, 200072, PR China.
E-mail address: lx_net@sina.com (X. Li).

http://dx.doi.org/10.1016/j.actamat.2016.09.016
1359-6454/

features to material and/or processing parameters.

With the development of superconducting technique, a high magnetic field has been widely used to improve material properties during material processing, such as diffusion [21,22], texturing [23–25], phase transition thermodynamics [26,27] and phase transformations [28,29]. In the previous works, the influence of the high magnetic field on the dendrite growth [30–33] and eutectic microstructure [34–36] has been systematically reported. However, so far, the effect of a high magnetic field on the solidification microstructure in the ternary alloys has not yet been studied. In the present work, two different compositions Al-Cu-Ag ternary alloys were selected to investigate the effect of the high magnetic field on the solidification behavior. The aim of this work is twofold: on one hand, the effect of a high magnetic field on the solidification structure in the Al-Cu-Ag ternary alloys has been investigated experimentally; on another hand, by studying the influence of the magnetic field on the solidification structure in the Al-Cu-Ag ternary alloys, the understanding on the evolution of the solidification structure in ternary alloys may be extended and deepened.

2. Experimental

Master alloys of composition Al-21.5 wt%Cu-27 wt%Ag and Al-17.6 wt%Cu-42.2 wt%Ag were prepared by induction melting of high purity elemental materials (99.99% or better) under a high purity argon atmosphere. Both alloy compositions were plotted on the reported liquidus projections of the Al-Cu-Ag system as shown in Fig. 1. They follow two different solidification processes: two-phase univariant eutectic growth ($L \rightarrow \alpha$-Al+Al_2Cu) and three-phase invariant eutectic growth ($L \rightarrow \alpha$-Al+Al_2Cu+Ag_2Al) for Al-21.5 wt% Cu-27 wt%Ag and Al-17.6 wt%Cu-42.2 wt%Ag alloys, respectively. Each alloy sample was heated to 900 °C, magnetically stirred for half an hour, poured into a graphite mold to cast sample with a diameter of 3 mm and a length of 300 mm. Then, the cast sample was enveloped in a high-purity corundum tube with an inner diameter of 3 mm and a length of 200 mm for directional solidification experiment. A details of the experimental apparatus has been described in Ref. [37]. The apparatus consisted of a static superconductor magnet, a Bridgman-Stockbarger-type furnace, and a growth speed and temperature controller. The temperature gradient was equal to 20 K/mm, and the magnet could produce an axial static magnetic field with an adjustable intensity up to 6 T. During the experiment, the sample in a corundum crucible was

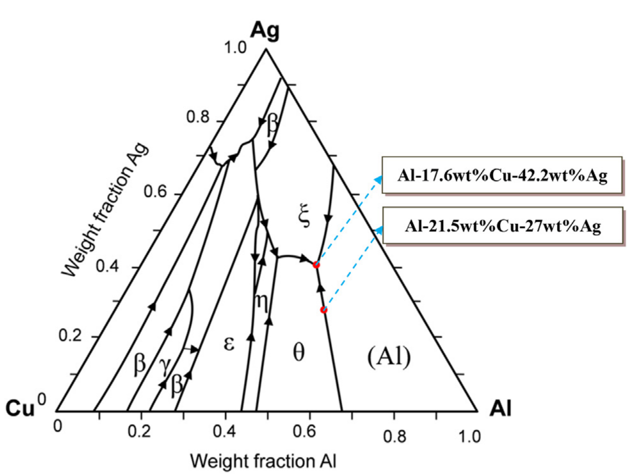

Fig. 1. Liquidus projection of the Al-Cu-Ag system: The investigated alloy compositions have been selected in this study near the univariant eutectic reactions: $L \rightarrow \alpha$-Al + θ-Al_2Cu and invariant eutectic reactions: $L \rightarrow \alpha$-Al +θ-Al_2Cu + ξ-Ag_2Al.

melted and directionally solidified in the Bridgman apparatus by pulling the crucible assembly downwards at a constant growth speed by means of synchronous motor. After 120 mm of steady-state growth, the sample was quenched by rapidly withdrawing the crucible into a water-cooled cylinder containing liquid Ga-In-Sn metal (LMC).

The microstructures of the as-solidified samples were characterized by using an optical microscope (Leica DM 6000M), a scanning electron microscope (Hitachi SU70) equipped with an energy-dispersive X-ray (EDX) spectrometer and electron backscattered diffraction (EBSD). For the EBSD measurements, the samples were ion polished to obtain good Kikuchi patterns. The EBSD data shown in the present study were rotated by 90°around the reference direction (RD) to the transform the data into a more familiar reference frame, with the normal direction (ND) along the solidification direction. Compositions of the bulk samples and phase constituents were analyzed using electron probe microanalysis (EPMA) with an error of 3–5%.

3. Results

3.1. Morphology

Fig. 2 shows the microstructures near the liquid/solid interface in directionally solidified Al-21.5 wt%Cu-27 wt%Ag alloy at various growth speeds with and without a 6 T magnetic field. Typical convex liquid/solid interface is observed in the case of no magnetic field. At a lower growth speed of 0.8 μm/s, the interface consists of regular coupled α-Al and Al_2Cu lamellae. At moderate growth speeds of 3 and 5 μm/s, the interface displays eutectic cellular structure. At a higher growth speed of 10 μm/s, the primary α-Al phase appears and is surrounded by the eutectic. The above results are similar to the ones reported in Refs. [11,38]. When a 6 T magnetic field is applied, the interface becomes flat and the eutectic cells transform into planar coupled eutectic. Moreover, the application of a 6 T magnetic field induces the deformation of the α-Al dendrites as shown in Fig. 2(h). Fig. 3 presents transverse microstructures in directionally solidified Al-21.5 wt%Cu-27 wt%Ag alloys under various growth speeds without and with the magnetic fields. The lamellar/rod mixed structure tends to transform into the rod-like structure with increasing magnetic field intensity. The magnetic field causes the decrease of the eutectic cellular spacing and the disappearance of the eutectic cell boundaries. Fig. 4 shows the longitudinal microstructures near the liquid/solid interface and corresponding transverse microstructures in directionally solidified Al-17.6 wt%Cu-42.2 wt%Ag alloys at a growth speed of 0.8 μm/s without and with a 6 T magnetic field. It can be found that the magnetic field induces the formation of banded structure which consists of a grey Al_2Cu phase and a white Ag_2Al phase (see Fig. 4(b)). In addition, the macro-segregation occurs on the transverse section and various eutectic morphologies can be observed in different regions of the sample when a magnetic field is applied.

Fig. 5 displays typical transverse microstructures in directionally solidified Al-17.6 wt%Cu-42.2 wt%Ag alloys at various growth speeds without and with a 6 T magnetic field. It can be observed that an aligned ladder or brick-like structure forms either without or with the magnetic field. The two intermetallic phases Ag_2Al and Al_2Cu alternate in a row and thus one could image them as one lamella consisting of two sub-lamellae. Note that the grey-scale contrast between Al_2Cu phase (grey) and α-Al (dark) phase decreases and the boundaries disappear at the growth speeds of 3 and 5 μm/s under a 6 T magnetic field.

Fig. 6 (a) and (b) illustrate the coupled eutectic cellular spacing (λ_c) in the Al-21.5 wt%Cu-27 wt%Ag and eutectic spacing (λ_α and λ_θ) in the Al-17.6 wt%Cu-42.2 wt%Ag as a function of the magnetic field