Materials Science and Engineering A 530 (2011) 271–276

Contents lists available at SciVerse ScienceDirect

Materials Science and Engineering A

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

In situ observation of columnar-to-equiaxed transition in directional solidification using synchrotron X-radiation imaging technique

Qing Dong[a], Jiao Zhang[a,*], Jinfang Dong[a], Honglan Xie[b], Zhijun Li[b], Yongbing Dai[a], Yang Liu[a], Baode Sun[a]

[a] The State Key Laboratory of Metal Matrix Composites, Shanghai Jiao Tong University, Shanghai 200240, PR China
[b] Shanghai Synchrotron Radiation Facility, Shanghai Institute of Applied Physics, Shanghai 215600, PR China

ARTICLE INFO

Article history:
Received 2 July 2011
Received in revised form 26 August 2011
Accepted 20 September 2011
Available online 29 September 2011

Keywords:
Microanalysis
Aluminium alloys
Casting
Grain growth
Interfaces

ABSTRACT

A columnar-to-equiaxed transition (CET) was induced by controlling the cooling rate in the directional solidification of an Al-15 wt.% Cu alloy. The morphological evolution of the mush–liquid interface in CET was tracked and recorded by X-ray imaging. The dendrite growth and detachment, solute distribution at the growth front and excessive solute enrichment in the liquid pockets were clearly observed. Based on the observations, it was found that there is an obvious increase in melt density in the liquid pocket due to the excessive solute enrichment. The density difference between the melt and the solid is an important reason for dendrite detachment and fragment movement, which is considered as the key to the inducement of CET. Finally, the CET process and the growth of equiaxed dendrites at the mush–liquid interface were described from the viewpoint of dendrite detachment.

1. Introduction

Most liquid–solid transitions in the casting process can be considered as processes far from thermodynamic equilibrium. The pattern formation in crystal growth can be influenced by many factors such as cooling rate and temperature gradient, so metals show morphological diversity during solidification. This diversity not only brings a wide selection to design the microstructure to improve the properties of materials, but also leaves a barrier that is hard to overcome in the production of materials with a high degree of structural uniformity, for example, the casting of superalloy single-crystal turbine blades [1,2]. Microstructures are at the center of materials science and engineering. The solidification interface has an important effect on the formation and evolution of structures [3]. Therefore, material science always focuses on the interface and the research methods related to it. A lot of work, including experimental and theoretical endeavors, has been done to deepen the fundamental understanding of the solidification interface.

One difficulty in the imaging study of the solidification interface is that metals, most with high melting points, are opaque to visible light. The other is that the solidification interface is hard to be continuously tracked and recorded. Therefore, transparent organic alloys, rapid quenching technique, mathematical and physical models and computer-based simulation techniques were commonly used to study the interface [4–11]. However, in many cases, there is a great difference between the model and the reality. Real-time observations of the solidification interface during the solidification of metals are still necessary. Fortunately, at present, the high-brilliance synchrotron X-radiation imaging technique based on phase-contrast has shown promise for in situ observations of evolving solidification microstructures [12–16].

In directional solidification, the columnar-to-equiaxed transition (CET) is an important morphological transition which is not yet completely understood [10,13,17–22]. A viable transport of the free crystals from the mush to the columnar front is considered as the key to the inducement of CET [13,17]. However, this argument still needs to be confirmed by convincing evidences and detailed analysis. In addition, there is a controversy on the mechanism of dendrite detachment. Both mechanical breakdown and remelting of dendrite arms have been considered to cause fragmentation [13,14]. In the present work, synchrotron X-ray imaging at Shanghai Synchrotron Radiation Facility (SSRF, China) has enabled us to carry out in situ and real-time observations of the CET process at the mush–liquid interface during the directional solidification of a hypoeutectic Al–Cu alloy. Based on the observation results, the reasons for dendrite detachment were analyzed using a mechanical model. The CET process and the growth of equiaxed dendrites at the mush–liquid interface were described from the viewpoint of dendrite detachment and fragment movement.

* Corresponding author. Tel.: +86 21 54747387; fax: +86 21 3420 2951.
E-mail address: zj119@sjtu.edu.cn (J. Zhang).

0921-5093/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.msea.2011.09.085

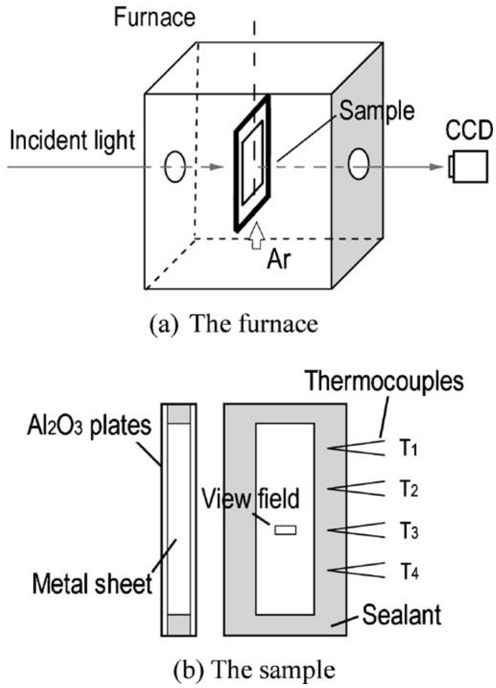

Fig. 1. Schematic of synchrotron radiation imaging experiment. (a) The furnace and (b) the sample.

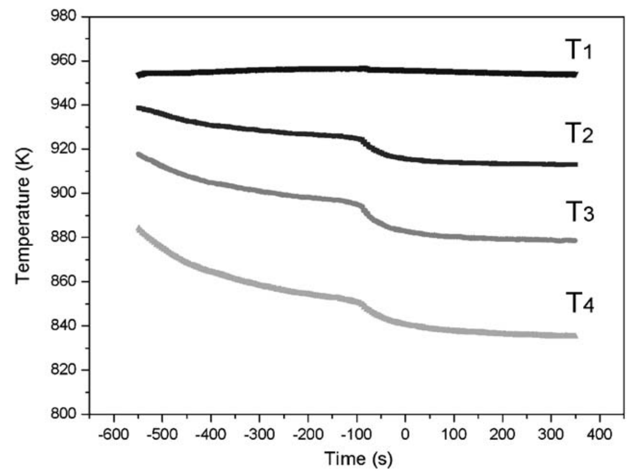

Fig. 2. Real-time temperature curve during directional solidification.

2. Materials and methods

The solidification was carried out inside a resistance furnace with two opposite windows for the incident X-ray beam to pass through the sample. Fig. 1(a) shows the schematic diagram of the experimental setup. An Al-15 wt.% Cu alloy was prepared from pure elements (99.999 wt.% purity) in a graphite crucible. Rectangular slices ($60 \times 15\,mm^2$ and 700 μm thick) of the alloy were sandwiched between two 120-μm-thick Al_2O_3 plates, as shown in Fig. 1(b). The edge of the samples was sealed with a mixture of silica sol and Al_2O_3 powders. Four thermocouples were located at 8-mm intervals along the length of the sample to measure the temperature at different positions.

The experiments were performed at beam line BL13W1 at SSRF under experiment code 10sr0265. The sample was fixed upright in the furnace and heated to 953 K, about 60 K higher than the melting point of the alloy. After the sample held at 953 K for 60 min to homogenize the melt, the directional solidification experiment was carried out. The sample was cooled by a gas cooler at the bottom of it. The cooling rate was kept at 0.028 K/s and the temperature gradient at the location was G = 5.0 K/mm. An instantaneous increase in argon flow rate was employed to induce CET and a cooling rate of 0.250 K/s was reached and lasted for about 30 s, as shown in Fig. 2. An incident monochromatic X-ray energy of 20 keV was used for the X-ray imaging. The full image field of view corresponded to $6.8 \times 3.4\,mm^2$.

A mechanical analysis was carried out to evaluate the effect of density difference between the melt and the solid on the detachment of dendrite arms. The model is borrowed from the column model presented by Billia et al. [14].

3. Results and discussion

3.1. In situ observations

According to the phase diagram of the binary Al–Cu system [23], the alloy with a composition of Al-15 wt.% Cu is shown a typical hypoeutectic structure during equilibrium solidification, the proeutectic α-Al phase plus lamellar Al–Al_2Cu eutectic. The α-Al phase usually shows dendritic morphology. Because the eutectic temperature (821.4 K) of the alloy is much lower than that (∼893 K) of the liquidus, there is a thick mushy zone (∼15 mm in thickness here) during the directional solidification. However, the growth of the dendrites and their morphology evolution mainly happen in the solution diffusion layer, which is at the top of the mushy zone (∼1–2 mm in thickness here) and corresponds to the constitutional supercooling zone.

3.1.1. Dendrite growth and detachment

The whole process of CET in the directional solidification of the Al-15 wt.% Cu alloy was shown in Fig. 3. The moment right before CET occurs was set at t = 0.0 s [Fig. 3(a)]. It can be seen that columnar dendrites with the primary dendrite arm grew in a regular manner. Slight indications of growth instability, i.e., the variation in the growth direction, were observed at the tips of the dendrites. Besides, a free dendrite (named A) detached from the dendrite network was found in a liquid pocket at the right side of the figure. The growth instability of the columnar dendrites became obvious from Fig. 3(b). The dendrites at the growth front had a transitional pattern between the columnar and equiaxed dendrites. They grew up rapidly in different directions. Some dendrite tips followed the original growth direction of the columnar dendrites, while others did not. Meanwhile, the growth of the columnar dendrites was inhibited. Secondary dendrite arms of Dendrite A could be clearly identified from Fig. 3(c). The growth rate of the dendrite tips was about 50 μm/s. It was also noted that the free dendrite tends to move upwards away from its original location. Besides, this dendrite was also got rotated in the melt. As shown in Fig. 3 (b) and (d), it got rotated by 32.2° clockwise within 10 s, indicating that there is a strong melt flow at the growth front of the dendrites. Tiny needle-like crystal fragments, named B, C and D respectively, were observed in Fig. 3 (d, e, and g). Their movement and growth processes could be tracked. Considering that spontaneous nucleation is extremely difficult at shallow undercoolings and the melt has a high purity almost with no heterogeneous crystal nucleus, it is inferred that they evolved from the detached dendrite arms, especially the detached secondary or tertiary arms. From Fig. 3(h), Fragment B was well ahead of the tip of a big dendrite, and its movement stopped. As a result, the growth of the big dendrite below it was limited. A fracture process of a big dendrite branch (named E) was recorded in Fig. 3(j). Two indistinct images at the left side of the figure corresponded to the same detached dendrite branch. This indicated that the fracture of the dendrite branch was a sudden