In Situ and Real-Time Observation of the Solidification Process of Al–20 mass% Cu Alloy by Synchrotron X-ray Radiography

Maodong Kang1, Haiyan Gao1, Da Shu1,* Jun Wang1,2,*, Faguo Li1, Yanan Fu3, Lishibao Ling1 and Baode Sun1,2

1Shanghai Key Lab of Advanced High-temperature Materials and Precision Forming, Shanghai Jiao Tong University, Shanghai 200-240, P. R. China
2The State Key Laboratory of Metal Matrix Composites, Shanghai Jiao Tong University, Shanghai 200-240, P. R. China
3Shanghai Synchrotron Radiation Facility, Shanghai Institute of Applied Physics, Shanghai 215-600, P. R. China

The mechanical behavior of the casting is greatly controlled by the solidification microstructures.1,2) In general, the final microstructures of the casting are the result of the competition between the growth of an outer columnar (oriented properties) and an inner equiaxed (isotropic properties) grains.3) Traditionally, most previous studies of the microstructure have relied on the post-solidification, optically transparent materials and computer simulation. These research tools have supplied valuable insight toward understanding alloy solidification. However, the quenching method does not provide the microstructure evolution in situ but gives only a ‘frozen’ image of the solid microstructure. Although transparent organic materials have been successfully used to study directional solidification, their important thermal and physical properties are not completely identical to metallic materials.3,5) Based on the simplified physical model and phenomenological theory, the microstructure simulation is hard to completely reflect the solidification process. Besides, the solidification microstructures belong to the field of self-organization in out-of-equilibrium systems.5) Recently, with the increased availability of 3rd generation synchrotron (high monochromatic, coherence and brilliance), in situ and real time X-ray absorption radiography has been used to investigate the dynamics of the solidification.7–10) The purpose of this paper is to present our recent research, including the competitive dendrite growth, dendrite fragmentation and microporosity formation.

1. Introduction

2. Experimental Methods

The experiments were carried out on ID13W1 beam line at the Shanghai Synchrotron Radiation Facility (SSRF). Solidifications were performed inside a homemade resistance Bridgman furnace. The furnace can be used for directional solidification and equiaxed solidification. In directional solidification, the pulling velocities were ranging from 25 to 200 µm/s in a temperature gradient of 30 K/cm. In equiaxed solidification, the specimen was fixed at the furnace and the solidification was triggered by the so called power down method. Figure 1 shows the schematic diagram of the experimental set-up and specimen. Rectangular windows of the furnace (20 × 40 mm²) was set perpendicular to the incident monochromatic X-ray beam. For equiaxed observation, different parts of the specimen were fixed opposite to the opened window. The nominal alloy composition was Al–20 mass%Cu alloy. In order to obtain sufficient transmission, the thickness of rectangular specimens (30 × 40 mm²) were reduced down to 300 µm. Each specimen was sandwiched with two high purity Al2O3 plates. After holding at 750°C for 30 min to homogenize the solute distribution in the melt, the solidification experiments were performed.
The distance between specimen and detector was about 81 cm, and radiographs were recorded using a fast readout-low noise Charge-coupled Device (CCD) camera with nominal spatial and temporal resolution 7.4 µm and 300 ms, respectively. Based on the alloy composition and the sample thickness, the beam energy of 25 keV was chosen by a double Si (111) monochromatic crystal. In all recorded images, there was a weak contrast between Al-enriched dendrites and surrounding Cu-enriched liquid. Besides, some defects originating from the Al2O3 plates or the X-ray beam lower the recorded images quality. However, the image quality can be improved by the image processing,11,12) it was used to solve those problems by dividing the images taken at time \( t \) (Fig. 2(a)) with a reference image recorded at initial time \( t_0 \) (Fig. 2(b)) when the specimen was totally homogeneous liquid. Therefore, the images quality were improved without any defects, as shown in Fig. 2(c), where the Al-enriched solid appears in white and Cu-enriched liquid appears in dark grey.

3. Results and Discussion

Just as the earlier solidification studies,13) the successive images can display the solid–liquid interface dynamically. The competitive dendritic growth, fragmentation, micro-porosity formation were directly observed. The details are shown in the following.

3.1 Competitive dendritic growth

Based on the analysis of dendrite tip undercooling, the classical dendrite selection mechanism14) points that the favorably oriented dendrites (preferred crystalline orientation parallel to the thermal gradient direction) lead and block their misaligned neighbors by new dendrite arms developing. However, Fig. 3 shows the misaligned dendrite (B: angle 14.6°) was blocking the better aligned dendrite (A: angle 12.7°) at the grain boundary at withdrawal rate of 200 µm/s. Dendrite B moved slightly toward the left side, and dendrite A gradually degenerated before totally blocked by dendrite B, similar experimental results (post-solidification observation) were obtained in Nickel-based superalloy by Y. Z. Zhou.15) Obviously, the traditional mechanism can’t suit the unusual overgrowth phenomenon in the case of converging dendrites. In this case, the solute interaction plays a significant role in overgrowth in addition to dendrite tip undercooling. When converging neighbor dendrites (A, B) get close to each other, their solute fields will overlap. The solute interaction retards the growth of boundary dendrites and induces a lag of these dendrites relative to their immediate neighbors (B).16) The solute interaction may cause the favorably oriented boundary dendrites to move toward its favorably oriented neighbor. Besides, thermo-solute convection17) ahead of the columnar dendrites may promote unusual overgrowth.

3.2 Fragmentation phenomenon during solidification

A phenomenon that was often observed both during columnar and equiaxed dendrite growth was the formation of new grains by dendrite fragmentation.18,19) However, the details of the fragmentation were insufficiently understood and controlled. Figure 4 shows how dendrite arms detach at withdrawal rate of 25 µm/s, the primary dendrites from left to right were named A, B and C, respectively. Fragmentation occurred when dendrite branches (named D) were detached from the main primary trunks of mother dendrite (named B) by local re-melting due to local solute pile-up in the mushy zone during the dendrite ripening. In general, inter-dendritic liquid flow would impose a momentum onto dendrite D. Because the newly formed dendrite D has a lower density than the copper-rich liquid, it moved upward away from the mother dendrite B by buoyancy forces and continued to grow up in the supersaturated inter-dendritic liquid; After a few seconds, dendrite D reaching some critical size, it was too large to move freely due to the confinement of Al2O3.
container. Then, dendrite D was fixed ahead of the columnar front and blocked the advancing mother dendrite B with dendrite E after rotating to a steady position.

In Fig. 5, the columnar arm length of dendrite A, B and C associated with the time sequence of Fig. 4 was presented. The growth behavior of the three dendrites was different, especially for the growth velocity (initial slope). A plateau was distinctly visible after 15 s. It means that the growth of dendrite B was hindered and finally stopped because of the dendrite interaction (solute poisoning). It can also be seen that there was no mechanical impingement of the dendrite B with dendrites D, E. Besides, the decreased growth velocity (initial slope) of dendrite A is due to the growth front dendrite E, similar with dendrite B.

3.3 CET

In directional solidification, the alloy grows in a regular columnar way depending on the temperature gradient at the solid–liquid interface, the pulling velocity and the anisotropy strength. Traditionally, it is believed that the CET could occur when the columnar dendrites are blocked by equiaxed dendrites which nucleate in the constitutionally undercooled liquid. Using nucleation strategy and phase field simulation, the realistic-looking CET transition has been produced without considering melt convection. However, melt convection can significantly influence the CET position by carrying away newly formed grains. Different from the previous research, two kinds of blocking dendrites exist in the melt, including the dendrite fragmentation (dendrite D) and the nucleated dendrite (dendrite E) as shown in Fig. 5. The present works not only confirm the predominance of the nucleated dendrites in the melt but also point out the fragmentation phenomenon in CET also plays an important role in CET process, which improves the characterization and understanding of the mechanism.

3.4 Free solidification microstructure

The specimen could solidify freely just as the casting production in the foundry by the power down method. As the previous mentioned, Cu-enriched liquid will sediment with gravity either in or out of the mush zone, depending on the columnar dendrite growth direction. As shown in Fig. 4, with the growth direction of columnar dendrites along anti-gravity (DDAG), the Cu-enriched liquid flows into the mush zone and result in local solute pile-up. With the growth direction of columnar dendrite along gravity (DDG), the situation is reversed; there is no local solute pile-up and the Cu-enriched liquid flows out of the mush zone as well as transports recalcience heat to the bulk liquid. Figure 6 shows that continuous dendrites were floating toward the front of columnar dendrites and blocking the growth of columnar dendrites at the condition of DDG. It is interesting that there are three zones existing in the field of view, including columnar dendrites (I), small floating dendrites (II) and bulky dendrites (III). The small floating dendrites detached from the bulky dendrites or nucleated dendrites in the liquid, which have not enough time to grow up before touching the columnar dendrites and the mother bulky dendrites, which may stop feeding and result in surface microporosity in the columnar dendrites. Eventually, the distances of each zone depended on the solidification condition.

3.5 Microporosity formation

In a solidifying casting, feeding occurs by the flow of either liquid or solid material in an attempt to compensate for the contraction. Dendrite gridding formation at the last stage of solidification, the pressure in the liquid falls and causes an increasing pressure difference between the inside and outside of the casting, when some critical pressure is reached, the microporosity can appear and the remaining contraction is fed by microporosity growth as shown in Fig. 7. For future experiments, the spatial resolution and amplification should be increased to observe the dynamic process of microporosity formation.

4. Conclusions

In this study, the experiments on Al–20 mass%Cu carried out at SSRF were presented. The critical phenomenon of competitive dendritic growth, CET transition and microporosity formation has been analyzed by X-ray synchrotron radiography. From the observation, the misaligned dendrites were able to block the better aligned dendrites at the grain boundary due to the solute interaction. Two kinds of dendrites can block the growth of columnar dendrites and...
the gravity plays a significant role in the final microstructure. Besides, the microporosity formation was observed in the inter-dendrite at the last stage of solidification.

Acknowledgments

These works were supported by the National Basic Research Program of China (Grant No. 2012CB619505), Science and Technology Commission of Shanghai (Grant No. 11521100703) and the National Natural Science Foundation of China (Grant No. 51001074). The authors are pleased to acknowledge the assistance of the beam line BL13W1 staff members in SSRF (China).

REFERENCES