**In situ** study on dendrite growth of metallic alloy by a synchrotron radiation imaging technology

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This study was trying to observe the real-time dendrite growth of Sn-Bi and Sn-Pb binary alloys by a synchrotron radiation imaging technology. The imaging system includes an intense and high brightness synchrotron radiation source, a high-resolution and fast-readout charge coupled device camera, an alloy sample and a Bridgman solidification system. The imaging experiments were done at Beijing Synchrotron Radiation Facility with an updated synchrotron radiation imaging technique, diffraction-enhanced imaging, which was firstly used to study the dendrite growth of metallic alloy. A series of growth behavior and morphology evolution of dendrite have been in situ observed, such as columnar-to-equiaxed transition, dendrite competition, dendrite fragmentation and floating, etc., which can offer the direct proofs to verify or improve the solidification theories of metallic alloy. This research opens a novel window for the study of alloy solidification and enables the unambiguous understanding of solidification processes in optically opaque, metallic alloys.

dendritic growth, synchrotron radiation, solidification, diffraction-enhanced imaging, metal and alloys


1 Introduction

The microstructure has a strong influence on the mechanical properties of castings. The solidification of castings (i.e. nucleation and growth) plays a dominate role in the formation of microstructure. Therefore, many researchers have been using various methods to investigate the grain growth [1]. The microstructure mainly presents a dendritic morphology during solidification. A better understanding of dendrite growth and the dependence on various physico-chemical parameters is of major importance in controlling and forecasting the final microstructure. However, the real-time grain growth during alloy solidification is hardly observed. It is because that the metal and alloys are optically opaque and solidifying in a high temperature condition.

Most of experimental studies on metal solidification have been done based on a static analysis (e.g. microstructure observation after solidification) for the past decades. However, the static analysis can not provide the dynamic information of dendrite growth during solidification. In this sense, the researchers actually have a blind view in studying the dynamics of grain growth. A transient microstructure can be observed through an interrupted solidification by quenching. The quenching has been used in order to remain the transient microstructure of a solidifying casting sample [2–5]. This technique results in a substantial refinement, therefore the observed microstructure is not the original one. The growth information before and after quenching is missing. The method is still limited in analyzing the dynamics of the solidification processes. Certain transparent organic compounds, such as NH\textsubscript{4}Cl-H\textsubscript{2}O solution, have...
been used to simulate the solidification of metals since 1980’s [6]. For they are transparent, growing crystals can be studied with a microscope, but none of these analog systems included an opaque alloying element that could represent the solidification of metallic alloy. Equilibrium processes like planar and cellular growth in metals have been investigated by conventional X-ray microradiography, but the spatiotemporal resolutions are not sufficient for studying dendritic growth [7].

During this decade, the numerical modeling technique has been used to study the dynamics of solidification, and predict the evolutions of various microstructures. The stochastic models and deterministic models, including heterogeneous nucleation, dendrite growth, preferential orientation, and dendrite competition, were established to study the dendrite growth and morphology evolution [8–13]. Since these models are based on a number of assumptions and conjectures, their accuracy needs direct experimental verification.

A cutting-edge experimental technique was recently developed and demonstrated to be successful in studies of alloys [14–19], in which a third-generation monochromatic synchrotron radiation X-ray with high energy, high brightness, and high resolution is used for in situ imaging the solidification processes. However, no relevant researches have been reported in China due to the limitation of quality of synchrotron radiation source.

In this paper, the solidification kinetics of Sn-Bi and Sn-Pb alloys was in situ studied by the synchrotron radiation imaging technology at the first-generation Beijing Synchrotron Radiation Facility (BSRF) in China. For improving the spatiotemporal resolutions of the first-generation source, an updating imaging method, diffraction enhanced imaging (DEI), was applied. We succeeded in acquiring a series of time-sequenced images of dendrite growth including columnar-to-equiaxed transition (CET), dendrite competition, dendrite fragmentation, etc. The solidification phenomena in micro scale were discussed and understood based on the real-time observations.

2 Experimental

The experiments were carried out on the 4W1A beam line at BSRF. The energy can be selected with a range of 4–22 keV. The exit of the beam-line is 43 m away from the light emitting point. The spatial resolution is down to 5 μm in the horizontal direction and 1.8 μm in the vertical direction. A fast readout-low noise charge coupled device (CCD) camera made by Xradia Company was used to collect the images. The image signal can be converted into digital format and stored in a frame memory with a format of 1024×1024 pixels. Its spatial resolution is down to 3.25 μm. The view field is a square of 3.3 mm×3.3 mm.

DEI was used in this experiment. As compared with the conventional imaging of absorption contrast, DEI is a method based on the phase-contrast imaging, by which the internal structure of sample can be imaged by combining one or more contrasts [20]. A quasi-plane wave is generated passing through the sample by the monochromator. Changes in the refractive index in the sample produce disturbance in wave-front of a plane wave. Changes in phase of wave-front relate to X-ray scattering of electron in the sample. Suppose that X-ray transmits along the z direction, and then the phase change can be expressed as

$$\Phi(x, y) = -r_{c}\frac{\lambda}{\rho(x, y, z)}dz,$$

where $\rho(x, y, z)$ is electron density of the point of $(x, y, z)$ located inside the sample, $r_{c}$ represents classical electron radius, $\lambda$ represents the wavelength of X-ray, and the integral formula expresses the integral of the electron density within the propagation path $M$ in the sample.

Suppose that X-ray transmits along the $z$ direction initially, $k$ is wave number, and $\psi_{0}$ is wave function. Since phase change is generated along the $x$ direction, wave function of X-ray can be expressed as

$$\psi(x, y, z) = \psi_{0}(x, y, z)e^{i\Phi(x)}.$$

The wave vector in the X-ray propagation direction is

$$k'(x) = \frac{1}{i\psi(x, z)}\nabla\psi(x, z) = \frac{\partial\psi(x)}{\partial x} + k z.$$

For the phase gradient (first derivative along the $x$ direction) is relatively small, the angle that X-ray deviates from the initial direction is

$$\Delta x = \frac{\partial\Phi(x)}{k}.$$

Consequently, the phase gradient of wave front is the change in the direction of propagation. The information on the internal structure of the sample taken by the change of X-ray transmitting direction is recorded after being magnified by the analyzer [21].

Figure 1 shows the schematic diagram of DEI. The alloy sample is placed between the monochromator crystal and the analyzer crystal, which are made of Si (111). X-ray passes through the alloy sample after monochromatization. There is an additional analyzer crystal which is set between the sample and CCD detector. The analyzer crystal can filter the noisy X-ray to get more clear images. Since the diffraction angle of X-ray from analyzer to CCD detector is limited to the order of $10^{4}$ rad, the signal-to-noise ratio can be enhanced by using DEI.

A Bridgman solidification system was designed in this experiment, as shown in Figure 2. The system includes double furnaces. Each furnace consists of two parallel copper plates and a heat source, which is surrounded by a heat insulation material. The furnaces were used to melt the alloy