X-ray Microradiography of Dendritic Alloy Microstructures

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Metallic alloy materials are used everywhere. The most common examples include formed metal parts, such as automobile engine blocks, fenders, etc., and commonly used welding materials. Producing them usually involves cooling a high temperature liquid to form a solid phase. Almost universally, the solid phase shows a microcrystalline structure that is branched and dendritic. The dendritic structure evolves continuously during solidification, and size and entanglements of the dendrites play a critical role in the mechanical properties of materials during subsequent processing and in service.

Many theoretical and experimental efforts have focused on understanding dendritic growth and evolution. A number of models exist that try to explain so-called “coarsening mechanisms”, including dendrite remelting and dendrite coalescence. These problems have excited materials scientists and computational scientists because of the rich variety of phenomena exhibited by relatively simple mixtures. Melting and solidification are technologically important because they control the production of almost all common materials, including single- or poly-crystalline materials, polymers, ceramics, clays, and formed or welded metals. High-temperature melts and solidification are also studied to understand geological formations, deposition and reclamation of ores, and the Earth’s core and plate dynamics.

Most studies of alloys are limited to post-process imaging. Typically, cooled solid samples are sliced and sectioned for visual inspection. Some specimens are translucent or transparent, so visible light can be used to record dynamics of dendritic growth. Up until 2004, there was no way to directly image dendritic growth in opaque metal alloy solidification. That year, Li, as part of his thesis research, started working at the Cornell High Energy Synchrotron Source (CHESS) to develop an instrument for real-time observation of dendrite coarsening from dense opaque materials using x-ray synchrotron microradiography [1]. For a model system to study, they chose a eutectic alloy of Sn and Bi. During solidification, the Sn/Bi liquid mixture becomes enriched in bismuth and thus more opaque to x-rays. Using the intense wiggler x-ray source at the A2 station of CHESS, they chose an x-ray energy of 20 keV, midway between the absorption edges of Sn and Bi, to achieve an optimal image contrast. An additional advantage of this alloy is that its low liquidus temperature (~200C) makes it relatively straightforward to engineer a graphite vessel to hold the specimen with x-ray transparent windows.

The radiograph instrument used two imaging detectors. First, for rapid, low resolution “focusing” an x-ray fluorescent screen and a video camera sufficed. Once a dendritic front formed within the field of view, a modified, remotely controlled “point and shoot” 35-mm camera was loaded with high-resolution x-ray film and frame advanced to capture a growth sequence. Graduate student Li recalls how machinists thought he was crazy to take to modifying a $10 camera for the experiment at the synchrotron. In the end, though, the inexpensive camera could record over 150 frames very efficiently. The graphite vessel was outfitted with a flexible temperature control system so specimen temperature and temperature gradients could be controlled to simulate a variety of processing conditions.

In their latest work, this group explored whether the same or similar coarsening mechanisms are also at work in “directional solidification” of Sn-13 weight percent Bi alloys [2]. In a process well known as “zone melting”, short for temperature gradient zone melting (TGZM) (Pfann, 1955), a liquid zone is sandwiched between two solid zones and a thermal gradient imposes a compositional gradient in the liquid (Figure 1). The combination of thermal and compositional gradients causes solidification on the cold side of the liquid and remelting on the hot side. Concurrent solidification and remelting drives the liquid to march up the temperature gradient; dendritic branches to appear to move, coalesce, or disintegrate (Figure 2). With their x-ray microradiographs, they were able to characterize zone fronts moving at up to 26 microns per minute (no upper limit was determined) and measure values to agree well with analytic models describing the kinetics of TGZM. By varying both the temperatures and thermal gradients they saw a wide variety of dendritic morphology and evolution. At the faster cooling rates, the dendrites became finer. Secondary and tertiary dendrite branches could be seen and tracked easily.

This group has now proven that high-resolution, time-resolved data can be collected on dense, opaque metal alloys during solidification and zone melting. With this quality of visualization, materials scientists can now explore what role thermal gradients and zone melting play in the detailed morphology and microsegregation of alloy systems. Higher flux x-ray sources and faster electronic imaging detectors will push these measurements to better spatial resolution and faster time scales.

[1] B. Li, H. D. Brody, and A. Kazimirov, Phys. Rev. E., **70**, 062602 (2004).

[2] B. Li, H. D. Brody, and A. Kazimirov, Metallurgical and Materials Transactions A, **37A**, 1039 (2006).

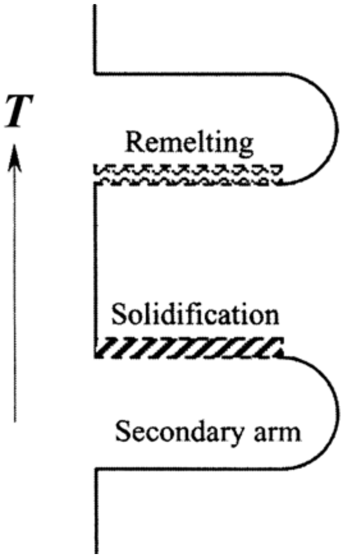

Figure 1: Schematic of dendrite remelting at the high temperature (top) side of the liquid pool and solidification at the lower temperature side.

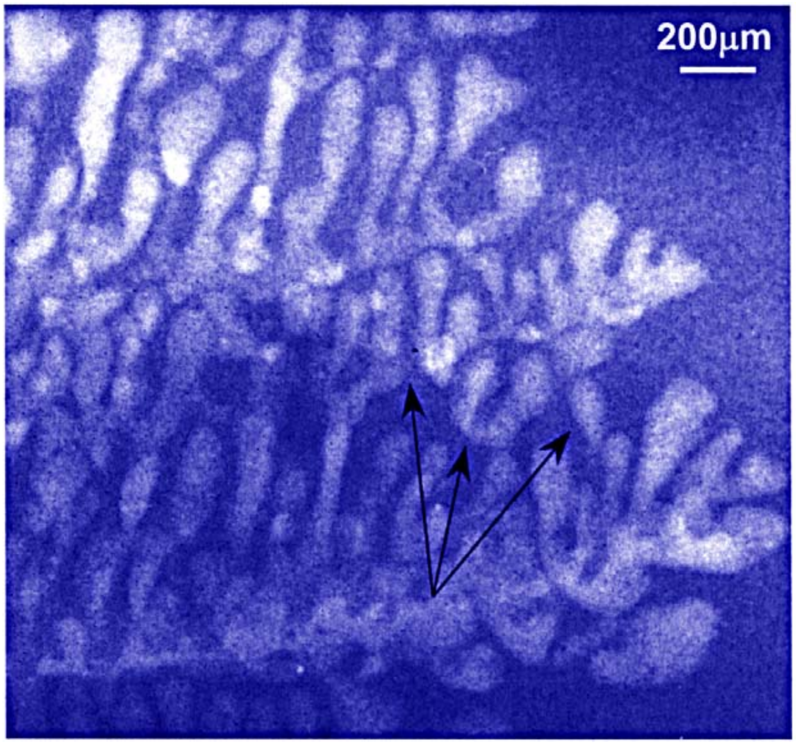

Figure 2: Full field radiographic image of tertiary dendrites growing upwards along the temperature gradient and decomposing (arrows) at their lower temperature ends.