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Focused Ion Beam Milling: A Practical Method for Preparing Cast Al-Si Alloy Samples for Transmission Electron Microscopy

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Communications

Focused Ion Beam Milling: A Practical Method for Preparing Cast Al-Si Alloy Samples for Transmission Electron Microscopy

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The aluminum-silicon eutectic reaction has been of great interest ever since Pacz patented aluminum-silicon alloys in 1921.[1] However, despite the many hypotheses proposed over the past 80 years,[2] the genesis of this technologically important transformation and the crystallographic relations between its constituents remain uncertain. This is mainly due to the lack of conclusive evidence provided by experimentation in support of the proposed hypotheses. Particularly, conventional sample preparation techniques such as ion milling and electropolishing are not capable of allowing the user to select specific microstructural areas for examination in the transmission electron microscope (TEM). Often times the researcher must produce many samples before specific microstructural features of interest are coincident with electron transparent regions in the TEM foil. In this article, we introduce the focused ion beam (FIB) milling technique as a viable method for producing good quality TEM samples of cast aluminum-silicon alloys; however, we make no attempt to establish any results pertaining to the physical metallurgy of this system.

Giannuzzi and Stevie[3] recently reviewed the details of the focused ion beam milling technique, and Figure 1 shows a schematic illustration of the procedure employed in this work. Bulk samples of cast Al-7 wt pct Si and Al-7 wt pct Si-0.022 wt pct Sr alloys were prepared from high-purity metals and one of their respective surfaces electropolished. This electropolished surface was then viewed using a scanning electron microscope (SEM) and the desired location for TEM work selected. The FIB unit is a fixture in the SEM allowing the location of ion impingement on the sample to be precisely selected. A tungsten metal strip, about 4-μm thick, was placed over the desired location in the microstructure and the volume of metal surrounding this thin strip ion milled by a focused gallium ion beam. This procedure resulted in a sample of about 100-nm thickness and therefore electron transparent. However, sample thickness can be chosen to result in a range appropriate for TEM. After the FIB sample has been milled from the bulk, an electrostatic probe allows the user to place the sample on a carbon-coated grid for TEM analysis. The FIB milling technique allows the user to select specific aspects of the microstructure before TEM sample preparation commences. This saves considerable time compared to the "hit-or-miss" approach of conventional TEM sample preparation techniques.

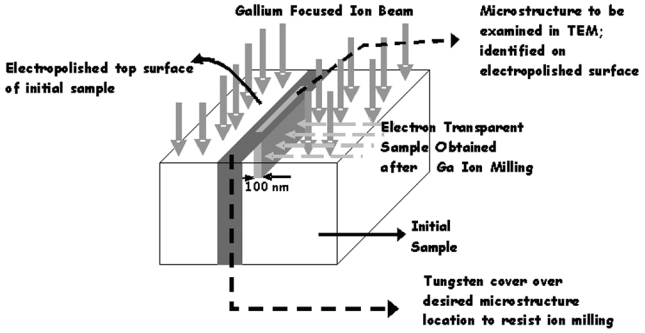

Fig. 1—Schematic illustration of the FIB sample preparation technique. A desired microstructure on the initial electropolished surface of the sample is located and masked by a tungsten strip. The rest of the initial sample bulk around this desired location is milled out by a Ga ion beam resulting in an electron-transparent sample cross section.

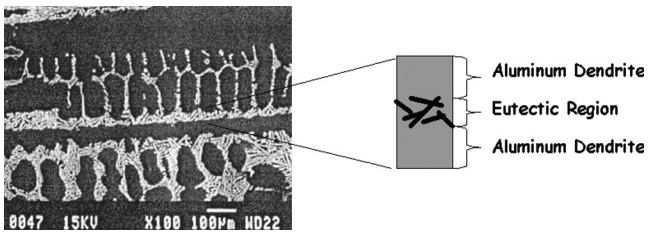

Fig. 2—Microstructure of the Al-7 pct Si sample showing the location (white rectangular box) where the sample for TEM work was produced by FIB milling. The white rectangular box is illustrated in the adjoining schematic.

Figures 2 through 5 illustrate the high quality of cast Al-Si alloy samples produced *via* the FIB milling technique. Figures 2 and 5 are SEM images of Al-7 wt pct Si and Al-7 wt pct Si-0.022 wt pct Sr, showing the locations in the microstructure where samples for TEM analysis were prepared by FIB milling. The TEM samples were prepared such that both an aluminum dendrite and a eutectic region were visible in the same sample. Figures 3(a) and (b) are low-magnification bright-field TEM and high-magnification secondary electron SEM images of the Al-7 wt pct Si sample, respectively. Figure 3 compares the TEM and SEM images of the desired location shown in Figure 2. The incident electron beam is not aligned to a specific zone axis in the sample. Boundaries not visible in the SEM are apparent in the TEM. Figure 4 shows higher magnification TEM images at various silicon and aluminum boundaries. Figure 4 also shows $[100]_{Al}$ convergent beam electron diffraction patterns (CBED) from the two primary aluminum dendrites surrounding the eutectic phase as well as the Al in the eutectic region. This demonstrates FIB as a viable method for creating TEM samples from precisely selected locations. Furthermore, the quality of these samples is sufficient to allow conventional TEM analysis.

Figure 6 shows high-magnification secondary electron SEM images of the FIB milled desired locations in the cast

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(*a*) (*b*)

Fig. 3—Comparison of TEM and SEM micrographs of Al-7 pct Si at the desired sample location: (*a*) low-magnification TEM image of the Al-7 pct Si sample and (*b*) high-magnification SEM image of the Al-7 pct Si sample.

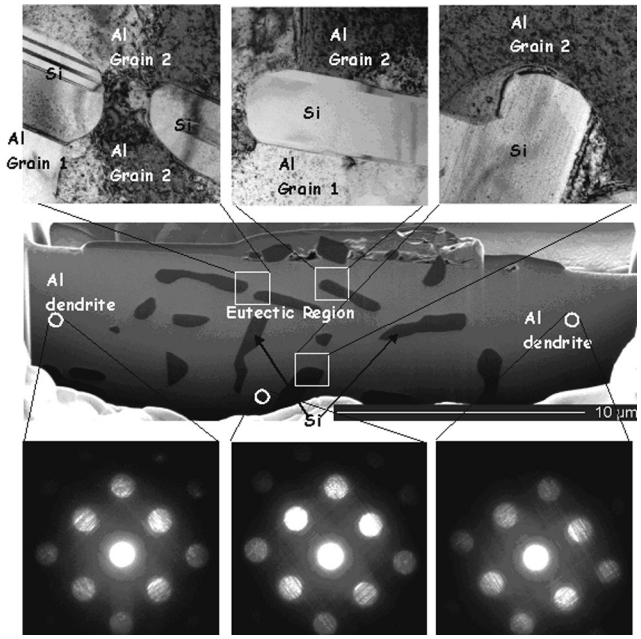


Fig. 4—High-magnification TEM images taken at various boundaries between silicon and aluminum in the eutectic region of the Al-7 pct Si cast alloy sample. The TEM images are taken with an aluminum grain (Al grain 2) in the middle of the eutectic phase oriented to the $[012]_{Al}$ zone axis. Also shown are $[100]_{Al}$ CBED patterns at locations in the two aluminum dendrites and the aluminum in the eutectic region (Al grain 1). The locations of the images and the CBED patterns are shown as white boxes and circles, respectively, in the high-magnification SEM image.

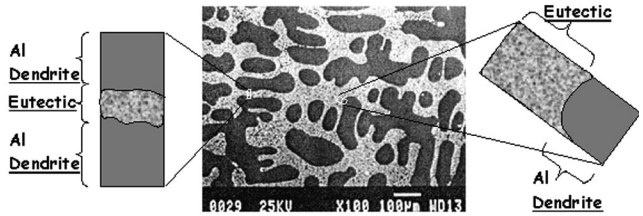
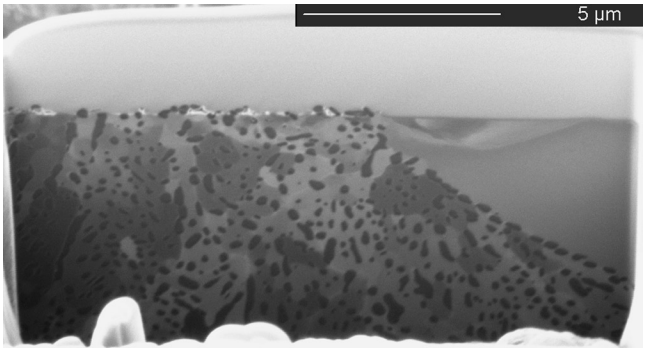
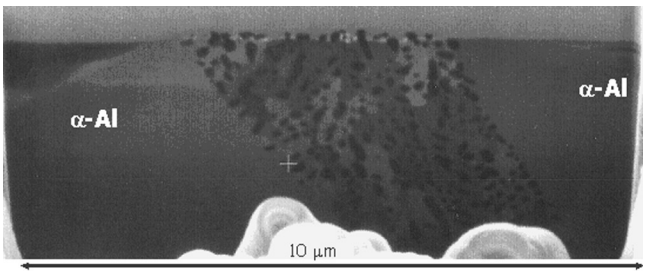


Fig. 5—Microstructure of the Al-7 pct Si-0.02 pct Sr sample showing the location (white rectangular box) where the sample for TEM work was produced by FIB milling. The schematics illustrate the microstructure in the white box.



(*a*)



(*b*)

Fig. 6—Micrographs of the modified Al-7 pct Si-0.02 pct Sr. (*a*) A primary aluminum dendrite on the right-hand side and a eutectic region. (*b*) Primary aluminum dendrites on both sides of a eutectic region. In both cases, notice the small aluminum grains within the eutectic region and the modified nature of the eutectic silicon.

Al-7 pct Si-0.02 pct Sr alloy* of Figure 5. Figure 6(a) shows a primary aluminum dendrite bordering a eutectic region, and Figure 6(b) shows two primary aluminum dendrites separated by a region of Al-Si eutectic. Notice the modification of the eutectic silicon into very fine fibrous crystals; more importantly, note the presence of a very small aluminum eutectic grains within the Al-Si eutectic phase. Modification of the eutectic silicon morphology can be easily seen by optical microscopy; however, the high resolution of the TEM is necessary to resolve the small aluminum grains within the eutectic phase. No damage in the form of excess dislocations resulting from FIB sample preparation is observed. However, it is not yet known if this technique

*Certain chemical additives to Al-Si alloys, *e.g.*, Sr and Na, modify the morphology of the eutectic silicon from a needlelike morphology to a much finer fibrous morphology.[4] The mechanism by which this chemical modification occurs is a matter of controversy,[2] but Lu and Hellawell[5,6] are credited with the most widely accepted theory.

induces excess twinning of Si, or if surface layer atoms have been disturbed. Disruption of surface layer atoms may limit the ability to perform high-resolution TEM work. At this time, we can conclude that FIB milling produces samples of sufficient quality for conventional TEM analysis.

The ability to perform TEM work on cast Al-Si eutectic alloys in specific microstructural regions is commensurate with the ability to quickly prepare electron transparent samples without inhibiting analysis by the introduction of defect structures. The FIB milling technique allows the preparation of such samples, and is currently being used to characterize mechanisms underlying the evolution and chemical modification of the eutectic phase in cast aluminum-silicon alloys.

REFERENCES

1. A. Pacz: U.S. Patent No. 1,387,900, 1921.
2. M.M. Makhlouf and H.V. Guthy: *J. Light Met.*, 2001, vol. 1, pp. 199-218.
3. L.A. Giannuzzi and F.A. Stevie: *Micron*, 1999, vol. 30, pp. 197-204.
4. J.E. Gruzleski and B. Closset: *Treatment of Liquid Aluminum Silicon Alloys*, American Foundrymen Society, Des Plaines, IL, June 1990.
5. S. Lu and A. Hellawell: *J. Cryst. Growth*, 1985, vol. 73, pp. 316-21.
6. S. Lu and A. Hellawell: *Metall. Trans. A*, 1987, vol. 18A, pp. 1721-33.

Mechanical Milling–Induced Deformation Twinning in Fcc Materials with High Stacking Fault Energy

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Various mechanical milling/alloying techniques have been widely used to produce nanocrystalline and other nonequilibrium structures, sometimes in large quantities. Approximately 4000 research papers investigating the processing, properties, microstructures, and applications of the milled materials have been published to date.[1] In these studies, it is frequently stated that various crystal defects such as dislocations, vacancies, stacking faults, and increased grain boundaries are generated due to heavy deformation caused by mechanical milling. However, only limited individual observations of crystal defects in the milled materials have been reported; two notable exceptions are the evolution of dislocation structure in milled Ru and Al-Ru, which was examined in detail by high-resolution transmission electron microscopy (HRTEM)[2] and the HRTEM observation of deformation twins in the milled Cu.[3] Deformation twins[4] or annealing twins[5] were believed to contribute to the strengthening of materials. On the basis of the apparent grain-refinement effect of twinning with twin boundaries acting as obstacles to dislocations,[4,5,6] several modified Hall–Petch equations have been established to exhibit the influence of twins on the strength and work-hardening rate of materials.[4,5] Detailed studies[7,8,9] on the microstructure and mechanical performance of nanocrystalline Cu and Pd, synthesized by inert gas condensation followed by hot pressing at 1.4 GPa pressures, have been performed. The HRTEM and X-ray diffraction (XRD) studies, as well as conventional transmission electron microscopy (TEM) observations, indicated the presence of a large number of highly twinned grains, whose origin is possibly related to the inert gas condensation process[7] or formed by shear stresses during sintering of the nanocrystalline powder.[8] The twinned structure was thought to be responsible for the higher creep resistance in the nanocrystalline Cu and Pd, because twin boundaries are poor paths for vacancy diffusion and resist grain boundary sliding.[9] In this article, the presence of deformations twins in several milled fcc metals and alloys with high stacking fault energy (100 to 200 ergs/cm^2) was observed using conventional TEM and HRTEM, and the density of twins was analyzed using XRD.

Commercially available metal and alloy powders were mechanically cryomilled in a modified Union Process 01-ST (Union Process, Inc., Akron, OH) attritor with a grinding tank capacity of 0.0057 m^3 at a rate of 180 rpm in a stainless tank containing stainless steel balls. The ball-to-powder mass ratio used was 20:1 (500 g powder was loaded for each milling). Liquid nitrogen was continuously introduced into the tank during the milling. The flux of liquid nitrogen was properly controlled using a valve so that the milling temperature, monitored by a thermocouple, was maintained at a relatively constant value of 100 K. For comparison purposes, the milling was also carried out in methanol as well as in hexane. Nominal chemical compositions of the metal and alloy powders used and milling conditions are listed in Tables I and II, respectively.

To address the influence of annealing treatment on microstructure, the as-cryomilled INCONEL 625 powders were sealed in stainless steel cans after back filling with nitrogen and isothermally treated for different times at designated temperatures.

The XRD measurements were performed by using Cu K_α (λ = 0.150418 nm) radiation in a SIEMENS* D5000 diffractometer. The TEM studies were conducted on a PHILIPS** CM 20 microscope operated at 200 keV and HRTEM examinations were carried out in a JEOL† 3000 microscope operated at 300 keV. The TEM and HRTEM samples of cryomilled powder were prepared using the following procedure. The powders and epoxy were mixed to create slurry, which was then mounted into a stainless steel nut, sliced from a stainless steel pipe with an outside diameter of 3 mm, to form a 3-mm-diameter disk, which was then

*SIEMENS is a trademark of Siemen's Electrical Equipment, Toronto.

**PHILIPS is a trademark of Philips Electronic Instruments Corp., Mahwah, NJ.

†JEOL is a trademark of Japan Electron Optics Ltd., Tokyo.

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