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Site-specific Sample Preparation by Concentrated Ar Ion Milling for Post-mortem Atomic Resolution Imaging of Rapidly Solidified Al-Cu Thin Films After Pulsed Laser Melting

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Rapid solidification (RS) is the rapid extraction of thermal energy during the transition from liquid at high temperature to solid at ambient temperature by electron and laser beams [1]. RS of alloys results in the formation of metastable phases, refinement of primary and secondary products from solidification, and solute concentrations exceeding the equilibrium solid solubility limits [2]. Dynamic transmission electron microscopy (DTEM) in movie mode allowed for in situ TEM measurements of solid-liquid interface velocity during RS and direct observation of motion during solidification of the interface in Al-Cu alloys after laser irradiation as in [3,4]. DTEM experiments are typically performed on thin films deposited on Si_3N_4 windowed TEM grids with a total thickness of 130 to 200 nm. This large specimen thickness prevents atomic resolution imaging by scanning TEM (STEM) and TEM due to multiple overlapping grains. Typical specimen polishing techniques, e.g., conventional dimple grinding and focused ion beam (FIB) lift-out, are not viable due to destruction of RS microstructure during mechanical preparation and Ga ion beam modification during FIB preparation. In this work, we present site-specific preparation by concentrated Ar ion beam (CIB) [NanoMill® TEM specimen preparation system, Fischione Instruments] milling of metallic thin films supported by Si_3N_4 windowed TEM grids after pulsed laser induced melting and RS. We highlight the importance of site-specific specimen preparation to obtain artifact-free specimens with large areas of multiple grains presenting in the morphologically complex Al-alloy RS microstructure to enable atomic resolution study of metastable microstructural features existing on the RS microstructures.

Al-Cu thin films (140 nm thickness) were deposited by electron beam evaporation on Si_3N_4 windowed TEM grids (Fig. 1a). RS by pulsed laser melting was performed on the Al-Cu alloy film using a single 248 nm (KrF) excimer laser pulse of 15 ns duration. The region of interest on the specimen was identified and marked using electron beam-induced deposition of Pt using a dual-beam FIB; subsequently, low-energy CIB milling was employed at 900 eV (Fig. 1b). Thereafter, high resolution TEM (HRTEM) and aberration-corrected STEM imaging were performed.

Energy-filtered TEM (EFTEM) thickness maps show the specimen before (Fig. 2a) and after (Fig. 2b) CIB milling of the banded region of the RS microstructure (Fig.1b). The banded region was found to have one phase (1P) and two phase (2P) regions, with θ'-Al_2Cu particles found in the 2P region. Relative thickness reduction of about 80% from t/λ ~1.2-1.4 to t/λ ~ 0.2, with uniform electron transparent areas after CIB milling, is observed (Fig.2b). The thickness of the specimen after CIB Ar milling was calculated as 28 to 32 nm, which is suitable for aberration-corrected STEM imaging. Fig. 2c shows atomic resolution high-angle annular dark field STEM (HAADF-STEM) image of the 2P region with Gunnier-Preston (GP) zones, 1.4 to 2.8 nm in diameter, in bright contrast. GP zones formed by natural aging at room temperature of more than 90 days between laser induced RS and STEM image acquisitions. The evolution of the θ'-Al_2Cu particles and results from specimens after in situ RS DTEM experiments polished by CIB milling will be discussed. Analysis of the HRTEM and STEM images before and after the aging process will be presented.

References:
[1] EJ Lavernia & TS Srivatsan, J. Mater. Sci., **45(2)**, (2010), pp. 287–325.
[2] W Boettinger et al. , Acta Mater., **48(1)**, (2000), pp. 43–70.
[3] JT McKeown et al. , Acta Mater. **65**, (2014), pp. 56–68.
[4] V Bathula et al. , Acta Mater. **195** (2020), pp. 341-357.

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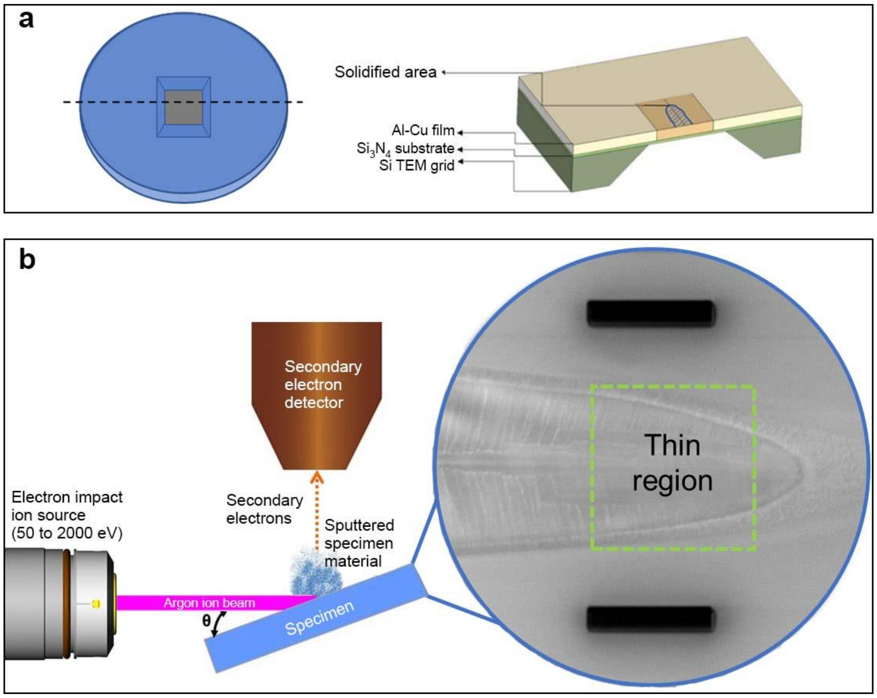

Figure 1. Representation of the backside and cross-sectional views of Si_3N_4 windowed TEM grids in (**a**). The concentrated Ar ion beam, specimen configuration, and secondary electron detector in the concentrated Ar ion beam milling system **(b)**. Inset is a SEM image showing the area of interest (green box), which was located using the electron beam-induced deposition of Pt.

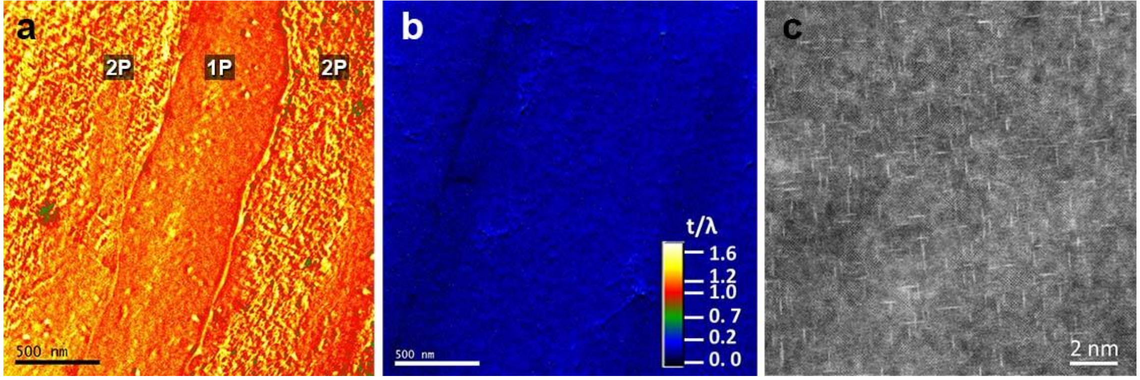

Figure 2. EFTEM maps of the RS Al-Cu thin film before (**a**) and after (**b**) concentrated ion beam Ar milling with relative thicknesses of t/λ=1.2 to 1.4 and t/λ=0.2, respectively. After Ar milling, the specimen was suitable for atomic resolution HAADF-STEM (**c**). A [110] orientation of Al with particles identified as Cu-based Gunnier-Preston (GP) zones, in high intensities, are observed.