Hardware and Techniques for Cross-Correlative TEM and Atom Probe Analysis

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Introduction

Atom probe tomography has primarily been used for atomic scale characterization of high electrical conductivity materials [1]. A high electrical field applied to needle-shaped specimens evaporates surface atoms, and a time of flight measurement determines each atom's identity. A 2-dimensional detector determines each atom's original position on the specimen. When repeated successively over many surface monolayers, the original specimen can be reconstructed into a 3-dimensional representation. In order to have an accurate 3-D reconstruction of the original, the field required for atomic evaporation must be known *a-priori*. For many metallic materials, this evaporation field is well characterized, and 3-D reconstructions can be achieved with reasonable accuracy.

Compared with conventional atom probes, the use of a local electrode has been shown to increase the sustainable evaporation rate and field of view [2]. The localized electric field produced by the local electrode enables arrays of specimens to be analyzed, as opposed to a single, electropolished wire needle. Specimen arrays increase throughput by minimizing exchange to UHV and cryogenic temperatures, as well as increasing material statistics through analysis of many specimens. In order to take advantage of these specimen arrays, preparation techniques utilizing *in-situ* FIB liftout techniques were developed [3]. These techniques allow routine preparation of nominally 100nm diameter specimens. The FIB also enables much improved control of the specimen diameter so the atom probe experiments can be tuned accordingly.

The maturation of local electrode and laser pulsed atom probe hardware, as well as FIB specimen preparation techniques, have enabled atom probe analysis of non-traditional materials such as semiconductors, ceramics, and some organic materials to become more commonplace [4]. For most of these materials, the evaporation field is not well characterized. For example, oxides and III-V materials tend to evaporate in clusters of atoms, rather than individual atoms [5]. The physics of cluster evaporation in atom probe experiments are not well understood, and the evaporation field required is also not well characterized. In order to increase the accuracy of the 3-D reconstructions in non-traditional materials, the evaporation field and its progression during an atom probe experiment should be calculated using the specimen geometric features, such as tip radius and shank angle.

While a combination FIB and SEM can give some information about atom probe specimen structure, higher resolution characterization of specimens using TEM and STEM can further increase reconstruction accuracy. TEM can image not only the specimen radius and shank angle with higher precision, but also can give the internal structure of interfaces and precipitates. Diffraction and high resolution imaging can give information about the orientation of crystallographic axes with respect to the specimen, and thus allow accurate scaling of the reconstruction in the *z*-direction. Analytical

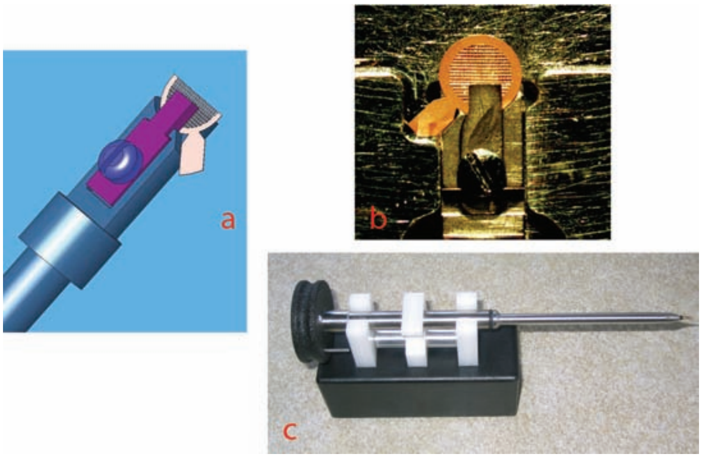

Figure 1. (a) CAD representation of the FIB / TEM / Atom Probe specimen effector and grid design. (b) Grid and effector mounted in loading jig. (c) Specimen carrier mounted on a TEM stage designed for the FEI Co. TF20ST.

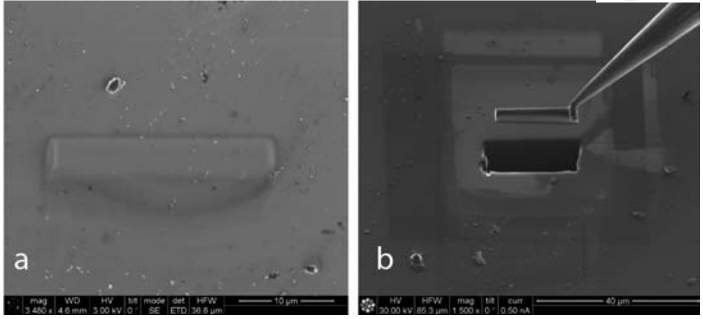

Figure 2. SEM and FIB images illustrating the in-situ specimen extraction and mounting of atom probe specimens. (a) Protective Pt deposition over area of interest using FIB-based gas injector deposition. (b) In-situ liftout of the wedge-shaped area of interest.

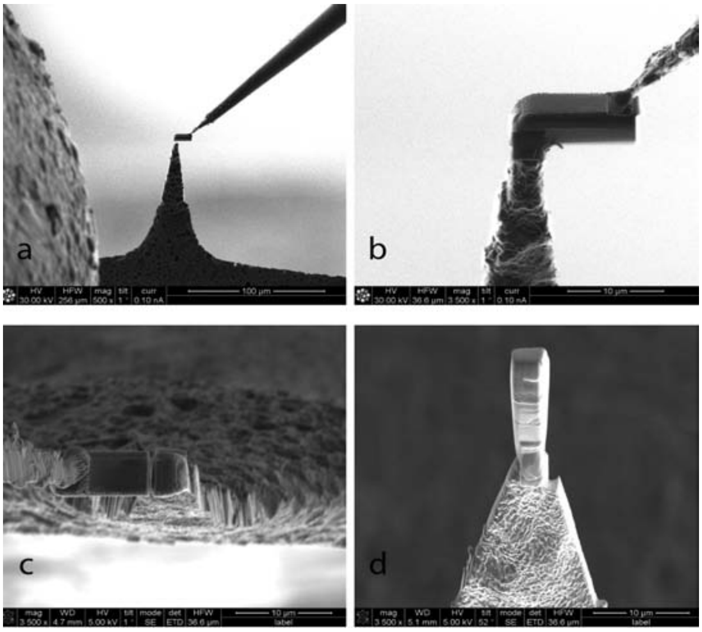

Figure 3. SEM and FIB images illustrating the specimen wedge mounting to the TEM grid posts. (a) Specimen approach to the TEM grid post, also showing the Pt GIS. (b) Specimen mounted to the TEM post with Pt. (c) Top-down image of the specimen wedge sectioned from the mounted specimen using the FIB. (d) Specimen mounted on the post before final preparation.


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techniques such as EDS and EELS as well as STEM-HAADF imaging can also give preliminary information about the composition of precipitates and interfaces, which again can aid in the scaling of the evaporation field during atom probe analysis and reconstruction.

In addition to pre-atom probe TEM analysis, post-atom probe analysis gives insight into how specimens with varying compositions evaporate under the influence of a high electrical field or laser pulse. This is particularly useful in the simultaneous analysis of high and low conductivity materials (such as high-*k* dielectric / gate interfaces or oxide precipitates in metals), where the lower conductivity materials alter the local electric field and thus the sequence of evaporation. It has been previously observed that oxide inclusions evaporate much later in sequence than a metallic glass matrix [6], purportedly due to the lack of sufficient field to break the oxide bonds and cause evaporation.

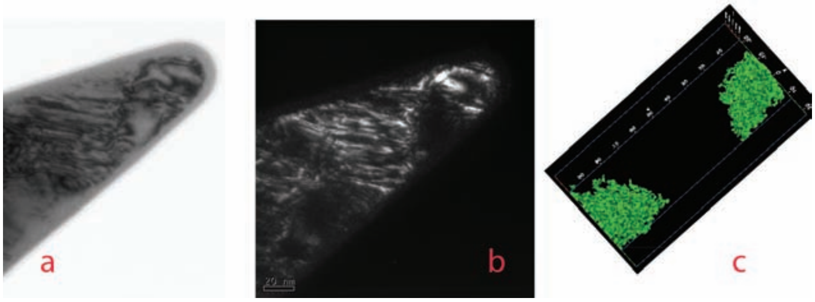

Figure 6. (a) Bright field and (b) dark field TEM images of a TiNiPt specimen. (c) Atom probe reconstruction of the same specimen in (a) and (b), illustrating the location of Ni-rich regions of the specimen.

Post-atom probe analysis can also give insight into why certain atom probe specimens fail during analysis, such as delamination at interfaces or dielectric breakdown. Finally, post-analysis also has the advantage of quantifying specimen irregularities induced by the laser or electric field, such as amorphization or phase separation.

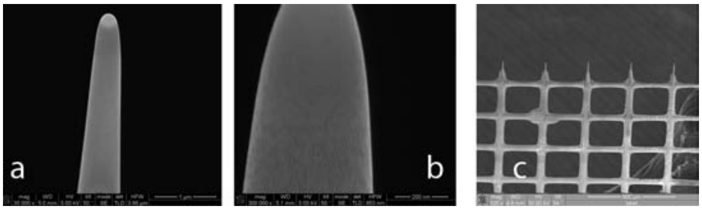

Figure 4. SEM images of the final specimen preparation steps. (a) Final shaped specimen following 5kV cleanup step also illustrating the region of interest. (b) Higher magnification image illustrating structure in the ROI. (c) Image of the grid posts after final preparation of the specimens.

Top-down specimen preparation

The usefulness of performing TEM and atom probe on the same specimen makes it beneficial to have an easy method for transferring specimen arrays from the FIB to the TEM and finally to the atom probe. To that end, as well as to minimize the handling of the fragile specimens, UNT and Hummingbird Scientific Instruments developed hardware to accept FIB liftout specimens onto 1-D arrays (Figure 1). Since the geometry of the local electrode necessitates specimens be separated by approximately 200 μm, we utilized standard 3 mm diameter, 100 mesh grids sectioned such that a vertical portion of each grid bar extended approximately 150 μm beyond each horizontal grid bar.

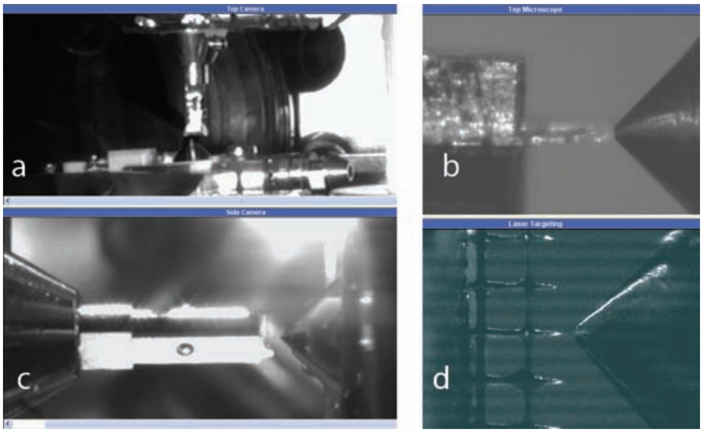

Figure 5. Optical microscope images of the TEM grid mounted specimens inside of the local electrode atom probe. (a) Top and side low-magnification images of the grid mount in proximity to the local electrode. (b) Higher magnification images of the grid mounted specimens in the analysis position next to the local electrode.

A jig was specially designed to guide the grids into proper position, after which the grids are secured into the end effectors by a clamp tightened with a screw (figure 1b). The jig also is designed to align and position the grid for proper sectioning. Once securely held in the grid effector, the grid does not need to be directly handled again for any of the processing or analysis. Figure 1c shows the grid effector loaded onto the TEM stage, specifically designed for an FEI Co. Tecnai F20ST. Since the grid effector has a narrow horizontal cross section of approximately 3mm, very high tilts (±80° using the center post) are possible. Using a modified grid effector and specialized grids, similar tilts have been demonstrated in small pole piece instruments. Once loaded into the TEM, the atom probe specimens are inherently positioned directly down the alpha tilt axis of the stage, allowing excellent tomography data to be acquired [7].

Once loaded and sectioned, the grids are further processed to minimize the bar cross section and therefore minimize FIB milling time. The grid effector can then be loaded into the FIB (in this case an FEI Nova 200 equipped with an Omniprobe Autoprobe 250) with the grid bars oriented vertically. FIB specimen preparation is similar to that developed for 2-D arrays. First, a region of interest is protected with e-beam and ion beam deposited Pt (Figure 2). FIB cutting is next used to cut a wedge-shaped cantilever from the region of interest. This piece is attached to an *in-situ* manipulator and transported to the posts on the grid. There, it is sectioned into an individual specimen by attaching the base of the wedge to a post and then FIB cutting to remove this area from the rest of the extracted wedge (Figure 3). This process is repeated to produce between 6 and 8 specimens for each liftout.

Sectioned specimens now attached to the grid posts are tilted parallel to the ion beam and sharpened using a series of decreasing diameter annular mills to produce the geometry required for atom probe analysis. Figure 4 shows a series of SEM images illustrating these final preparation steps. Milling can be terminated before the region of interest is reached by continual imaging with the SEM column. Reducing the FIB accelerating voltage to 5kV or lower removes a significant portion of the amorphization damage induced by the FIB [3].

Due to their diameter typically being in the electron transparency region, sharpened atom probe specimens can be directly transferred to the TEM stage (figure 1) and analyzed for relevant

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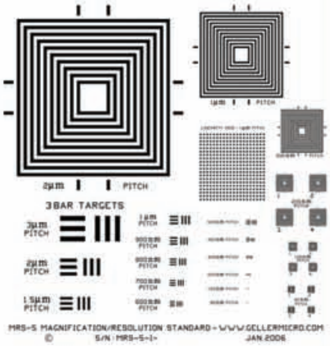

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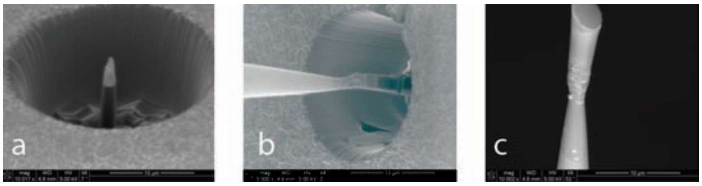

Figure 7. SEM images of (a) a FIB milled site-specific cylindrical specimen and (b) a manipulator needle attached to the top surface of the specimen using Pt deposition. (c) SEM image of the specimen following a FIB cut to remove the site-specific specimen.

structural information including their geometry, crystallography, and internal interfaces. As noted earlier, the specimens can also be easily examined using TEM tomography to high tilts. While obvious, it is nonetheless worthwhile noting that cylindrical specimens do not have the difficulties of planar specimens, in that the effective thickness remains constant with tilt angle.

Following TEM examination, the 1-D specimen effector then fits directly into a specimen holder ("puck") for atom probe analysis. The grid effector was designed such that the specimens sit at the center of the TEM stage and also have the clearance to fit near the local electrode within the atom probe. Figure 5 illustrates the 1-D grid design when used in the atom probe. The 1-D array allows for simple alignment of the specimens with the local electrode as well as with the laser pulse. One particular advantage of the 1-D array over 2-D arrays is that when properly aligned in laser pulsed mode, any laser energy that is not absorbed by the specimen simply passes through. 2-D arrays can facilitate the absorption of excess laser energy by specimens sitting within the laser's flight path beyond the sample being analyzed.

An example of cross-correlative TEM and atom probe analysis is illustrated in figure 6. This particular specimen is a TiNiPt alloy where segregation at the nanoscale can dominate the high temperature mechanical properties. Bright field and dark field images of a specimen prior to atom probe analysis are shown in figures 6a and 6b. These images were used to set the specimen diameter and shank angle for the atom probe reconstruction, as well as to note initially where phase segregation may be occurring. Figure 5c shows a nickel isosurface reconstruction of the atom probe analysis, completed using an Imago Scientific Instruments LEAP 3000x in laser pulsed mode. From this reconstruction, the atomic concentration and geometry of the Ni-rich particles and the matrix was directly determined in 3-D and compared with the TEM images of the particles.

Site-specific backside specimen preparation

In many applications, atom probe and TEM analysis is desired at highly site specific positions. Examples include phase segregated region in alloys, grain boundaries in ceramics, individual devices in semiconductor components, or even mechanical failures such as crack tips and indentations. In some cases, the FIB wedge-liftout specimen preparation technique can be utilized, but for others it may be difficult to properly align the feature of interest axially on the post and retain the feature of interest at the specimen tip after FIB milling. This is especially true for materials that have different sputtering rates, such as oxides in metallic matrices and the widely varying compositions present in semiconductor devices.

Atom probe analysis can be significantly inhibited by the presence of insulating materials within the specimen. Dielectric layers can lead to field concentrations, and ultimately specimen fracture during analysis. Our group has noted this effect significantly in semiconductor devices processed on silicon-on-insulator (SOI) wafers. In order to eliminate this issue, we have utilized backside (substrate side towards the atom probe detector) specimen preparation where the SOI layer is FIB milled away before analysis. Typically, this requires *ex-situ* specimen and nanomanipulator rotations that are time consuming and unreliable.

In order to expedite site-specific, backside atom probe specimen preparation, a simple adaptation to Omniprobe's Short Cut ™ technique [8] and grid geometry enabled this technique to be utilized. Preparation begins similar to the wedge liftout technique, where our area of interest is first protected using a combination of e-beam and ion beam deposited Pt to define and protect our area of interest. The major difference between the two techniques is in the amount of area defined. For the wedge technique, an area approximately 2 × 20 µm is required, where the site specific technique only requires a circular area approximately 500 nm in diameter.

After the area of interest has been defined by the metal deposition, a cylindrical post is cut from the sample (Figure 7a). The specimen is then rotated such that the nanomanipulator is parallel to the long axis of the cylinder. Again using FIB deposition, the nanomanipulator is attached to the cylinder (Figure 7b), and the cylinder cut loose (Figure 7c).

Once removed from the sample, the specimen is left attached to the nanomanipulator and loaded into the Short Cut jig *ex-situ* (Figure 8). An anvil subsequently cold forms the nanomanipulator needle into the grid material while concurrently sectioning the needle. The final specimen form (Figure 8b) gives line of sight access from the specimen tip to the atom probe optics. A specific advantage of forming the manipulator tip to the grid material is that TEM examination of the atom probe specimen is still achievable. If TEM is not going to be used, the nanomanipulator needle itself can be loaded into the atom probe. It should also be noted that since this

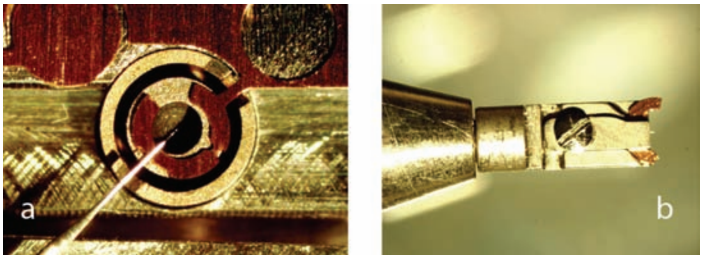

Figure 8. (a) Optical image of the nanomanipulator placed in the Short Cut™ anvil alignment jig. (b) Optical image of the nanomanipulator and site-specific specimen mounted to a TEM grid and placed in the TEM holder. The Cu grid is specially designed to allow atom probe analysis of the prepared specimen.

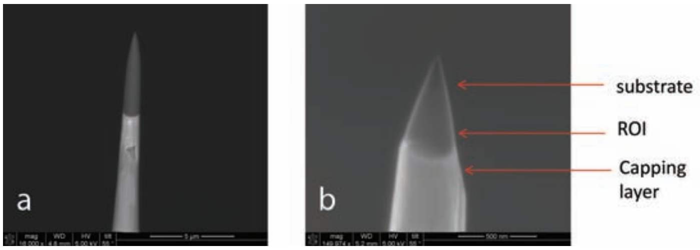

Figure 9. SEM images of the FIB shaped site-specific specimen illustrated in figures 7 and 8. (a) Image of the manipulator needle, Pt capping layer and weld, and specimen. (b) Image of the specimen after 5kV cleanup FIB mill. Note the specimen orientation difference from the first technique, with the substrate to be analyzed by atom probe before the ROI.

technique produces only a single specimen, a local electrode atom probe is not required.

Site-specific specimens prepared using this technique can subsequently be loaded into the grid effector used with the 1-D array (Figure 1). The effector and grid are then placed back into the FIB for final preparation of the specimen. Figure 9 shows the final geometry of these specimens once prepared. Again, in contrast with the wedge liftout technique, the sample substrate is facing the detector with the region of interest protected during FIB milling by the substrate.

In addition to being site-specific and eliminating layers that would normally cause difficulty during atom probe analysis, this technique is also much more rapid compared with the wedge liftout technique. This technique also requires less FIB time during initial metal deposition and specimen removal, as well as during final preparation FIB milling. Disadvantages are that only a single specimen is prepared (not usually a concern for site-specific preparation), and that some of the preparation must currently be completed *ex-situ*.

Utilizing EDS and EBSD in a combination FIB and SEM makes the best utilization of this site-specific technique. EDS spectral maps can illustrate areas of varying composition that are most interesting for atom probe analysis. EBSD maps illustrate variations in crystallographic phases and grain boundary orientations, again which may be of most interest for atom probe analysis.

The geometry of the specimens when using these site-specific techniques is particularly advantageous for TEM tomography. Cylindrical specimens do not change thickness with tilt angle, and the geometry of the grid and specimen are such that the grid will not enter the specimen field of view during tilting, making 360° rotations possible.

Summary

Techniques and hardware are introduced that enable the simple, reliable cross-correlative examination of FIB prepared atom probe specimens in the TEM. Utilizing previously developed FIB techniques in 1-D arrays, cross-correlative TEM and local electrode atom probe characterization is demonstrated. Adapting the Short Cut™ technique to atom probe specimens allows for rapid preparation of site-specific specimens from the substrate side of the sample rapidly and with high precision. ■

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References

1. Miller, M.K. (2000). *Atom Probe Tomography: Analysis at the Atomic Level*. New York: Kluwer Academic.
2. Kelly, T. F., et. al. (2004) First Data from a Commercial Local Electrode Atom Probe. *Microsc. Microanal.* **10**, 373-383.
3. Thompson, K., et. al. (2007) *In-situ* Site-specific Specimen Preparation for Atom Probe Tomography. *Ultramicroscopy* **107**, 131-139.
4. Kelly, T. F., et. al. (2007) Atom Probe Tomography of Electronic Materials. *Ann. Rev. Mat. Res.* **37**, 681-727.
5. Gorman, B. P. , Norman, A. G., Yan, Y. (2007). Atom Probe Analysis of III-V and Si-based Photovoltaic Structures. *Microsc. Microanal.* **13**, 493-502.
6. Gorman, B. P., *et. al.* (2008) Cross-Correlative TEM and Atom Probe Analysis of Partial Crystallization in NiNbSn Metallic Glasses. *Mat. Sci. and Tech.* in print.
7. Gorman, B. P., Diercks, D. R, Jaeger, D. (2008) Cross-correlative TEM and Atom Probe Tomography. *Microsc. Microanal. Suppl. 2*, in print.
8. Moore, T. M. (2006) Apparatus for Preparing a TEM Sample Holder. *U. S. Patent 7126132*.
