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Towards better 3-D reconstructions by combining electron tomography and atom-probe tomography

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ABSTRACT

Scanning transmission electron microscope tomography and atom-probe tomography are both three-dimensional techniques on the nanoscale. We demonstrate here the combination of the techniques by analyzing the very same volume of an Al–Ag alloy specimen. This comparison allows us to directly visualize the theoretically known artifacts of each technique experimentally, providing insight into the optimal parameters to use for reconstructions and assessing the quality of each reconstruction. The combination of the techniques for accurate morphology and compositional information in three dimensions at the nanoscale provides a route for a new level of materials characterization and understanding.

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1. Introduction

Advances in nanotechnology are often coupled to advances in characterization. As nanomaterials are three-dimensional (3-D) in nature, it is critical to improve our current 3-D techniques. The ultimate goal would be to determine the 3-D positions and chemical identity of each atom accurately in any materials system. Achieving the resolution necessary to attain this goal is crucial for the analysis and understanding of nanoscale materials where the key mechanisms occur at the atomic scale. Two different techniques to characterize 3-D structures of nanomaterials have been used extensively: scanning transmission electron microscope (STEM) tomography and atom-probe tomography (APT).

Electron tomography (ET) has been used successfully in the biological sciences for decades [1–3] and its application to inorganic materials using STEM has developed rapidly over recent years [4–6]. A fairly large range of volumes of material can be analyzed in 3-D, for example as small as $10\times10\times10\,nm^3$ to as large as $300\times300\times300\,nm^3$, with the 3-D resolution varying from just under 1 to ~5 nm depending on the volume size, the number of images acquired, the tilt range achieved, and the alignment and reconstruction methods. However, the standard acquisition and reconstruction parameters used today yield a resolution of ~1 nm in all the three spatial dimensions [7]. On the other hand, APT provides 3-D structural and chemical information on the atomic scale, but at the cost of a smaller volume of material (can be up to $150\times150\times100$–$500\,nm^3$). For a review of the recent progress made in the field of APT and the evolution of the technique since its origin as field ion microscopy, see [8].

ET and APT have different limitations and provide complementary information. In this paper we present the correlation between STEM tomography and APT of the very same sample. We demonstrate the feasibility of such an approach by applying it to an Al–Ag alloy containing nanoscale precipitates. By analyzing the respective artifacts, we show that an overall more accurate reconstruction can be obtained by combining the two techniques, leading to more reliable spatial and chemical information. We first review the characteristics and limitations of the two techniques.

1.1. Electron (STEM) tomography

STEM is an established technique [9,10] in which a focused electron beam is scanned across the specimen, and the scattered intensity is typically collected on a high angle annular dark field

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(HAADF) detector (though other detectors, such as bright field detectors, can also be used to collect the signal). Due to the detection geometry and physics of the scattering process, the images are sensitive to the atomic number of the species imaged, and, more beneficially for tomography, the images are mostly incoherent [11,12]. The coherency of images in conventional TEM has blocked advancements in TEM tomography for inorganic materials because they yield unreliable reconstructions. This is due to the projection requirement of tomography which states that the signal from the material should be a monotonic function of that physical property. As a result of the changing intensities in TEM images from diffraction contrast as the sample is tilted, the projection requirement is not satisfied. STEM on the other hand only suffers from diffraction contrast in a few images (when the specimen reaches zone axes), and these small number of images can be removed if desired during the reconstruction process, although they do not appear to have a large effect on the reconstructions in general.

In ET, the resolution is determined primarily by the number of images acquired, the tilt range that is achieved, and the volume analyzed [13,14]. Too few images lead to a muddled reconstruction of the original object, and too small of a tilt range amplifies the missing wedge artifact [4]. These restrictions define anisotropic resolution in the 3-D tomograms: the x-axis (along the holder) has the highest resolution, the y-axis (in plane but perpendicular to x) has a resolution similar to x, and the z-axis (parallel to the beam and perpendicular to the tilt axis) has the worst resolution.

While using new holder technology to tilt to $\pm 90^\circ$ can eliminate the missing wedge [15], the number of images acquired must also increase correspondingly, which leads to other factors that can limit resolution such as beam damage and contamination. Therefore, it is necessary to determine what is best for the particular specimen involved, and in many cases a high resolution ($\sim$1 nm) tilt series can be obtained even with $\pm 70^\circ$ or 75° if the number of images is sufficient.

1.2. Atom-probe tomography (APT)

APT relies on the controlled field evaporation of atoms from a sharp needle-shaped specimen. The identity of the evaporated ions is determined by time-of-flight mass spectrometry, while their original positions on the specimen surface are deduced by a simple projection law from the position of the hit on a 2-D position-sensitive detector [16]. The depth information is contained in the sequence of evaporation of each atom. Depending on the design of the atom-probe instrument used for analysis and on the specimen itself, the sampled 2-D area of the needle can vary from about 30 nm $\times$ 30 nm up to 150 nm $\times$ 150 nm, and the depth depends on the smooth evaporation of the sample, usually yielding 50 to hundreds of nm of data.

Sub-nanoscale spatial and chemical resolutions are usually achieved. However, a number of artifacts can limit the overall resolution. The origin of these artifacts is two-fold: the material microstructure leading to complex evaporation processes and the assumptions made in the reconstruction schemes. For instance, the presence of multiple phases with different evaporation fields leads to non-uniform evaporation [17] and the resulting complex specimen shape that evolves as phases evaporate is not taken into account in any existing reconstruction scheme. Among the manifestations of these non-uniformities are interface smearing in the reconstructed datasets and inaccurate composition analysis near interfaces. The schemes commonly used for APT data reconstruction assume that the shape of the specimen is spherical although it is known that because of crystallographic variation of the evaporation field the steady-state tip shape exhibits facets along low-index planes [18]. Despite the simplicity of the reconstruction process, some materials can be represented with a high degree of accuracy. Atomic planes, for instance, can be reconstructed when the normal to a densely packed crystal plane is close to the tip axis. The main limitation in obtaining accurate 3-D reconstruction is the detailed understanding of the evaporation processes taking place in complex materials, and in order to justify the implementation of more complex algorithms one needs to verify their validity. Knowing *a priori* the 3-D structure of the volume of material to be analyzed by APT is therefore crucial.

2. Data acquisition and processing

A needle-shaped specimen from a binary Al alloy containing 3 at% Ag was produced by a standard electro-polishing method using a mixture of 1:3 nitric acid to methanol [16]. The specimen has a radius of curvature at the tip of less than 100 nm to make it suitable for APT The alloy was annealed at 580 °C, water quenched, and aged at 230 °C for 45 min followed by 130 °C for 30 min. As described in Ref. [19], Ag-rich Guinier–Preston (GP) zones of $\sim$10 nm diameter as well as a high density of smaller Ag-rich clusters are formed during these annealing conditions. Since APT is a destructive technique, the STEM tomography was performed first, and the specimen was immediately transferred to the atom-probe for further analysis. Due to the nature of this paper in bringing together two techniques and fields that might not be familiar with the other, we explain the analysis of the data in detail that would normally be assumed within that field and excluded from publications.

2.1. Electron (STEM) tomography

The tilt series for STEM tomography was acquired on a 200 kV JEOL 2010F field-emission microscope using the Fischione on-axis tomography holder. The tilt range achieved was +80° to −80°, with a 2° increment, resulting in 81 images ($\pm 90^\circ$ could not be achieved due to eucentric height/goniometer limitations on the electropolished specimen). The images were aligned manually and reconstructed using an iterative reconstruction algorithm [20]. The reconstructed volume was then segmented using the Amira software, and the intensity on the surface of the specimen in the reconstruction was removed manually such that the particles would be clearly visible for comparison with the APT data. Therefore, the particles on the very edge of the dataset will not have accurate shapes or sizes, and those particles are not used in any analysis. We note that some thresholding must be chosen during the segmentation process, and here it was chosen such that only the large GP zones are visible. This therefore excludes the very small (1–2 nm) clusters and other intensity arising from the presence of Ag incorporation into the Al matrix. No 3-D scaling of the reconstructed volume or other forms of manipulation of the data was performed. The full resulting dataset is shown in Fig. 1, with one of the 81 Z-contrast images shown in Fig. 1(a), and the same projection of the 3-D reconstruction in Fig. 1(b). The movie of the full reconstructed volume can be seen in the Supporting Information section as Movie 1.

2.2. Atom-probe tomography

The APT was performed using an Imago LEAP-3000^{TM} microscope and the data was processed in the IVAS^{TM} software. During the analysis, the tip was held at 60 K in ultra-high vacuum conditions ($<5\times10^{-11}$ Torr), and analysis was performed using