Specimen preparation for correlating transmission electron microscopy and atom probe tomography of mesoscale features

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Abstract

Atom-probe tomography (APT) provides atomic-scale spatial and compositional resolution that is ideally suited for the analysis of grain boundaries. The small sample volume analyzed in APT presents, however, a challenge for capturing mesoscale features, such as grain boundaries. A new site-specific method utilizing transmission electron microscopy (TEM) for the precise selection and isolation of mesoscale microstructural features in a focused-ion-beam (FIB) microscope lift-out sample, from below the original surface of the bulk sample, for targeted preparation of an APT microtip by FIB–SEM microscopy is presented. This methodology is demonstrated for the targeted extraction of a prior austenite grain boundary in a martensitic steel alloy; it can, however, be easily applied to other mesoscale features, such as heterophase interfaces, precipitates, and the tips of cracks. © 2014 Elsevier B.V. All rights reserved.

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1. Introduction

Atom-probe tomography (APT) and atom-probe field-ion microscopy (APFIM) have been used to analyze point, line and planar imperfections in materials since the beginning of their development in the late 1960s [1–11]. The use of APT to study grain boundaries (GBs) in metallic alloys is of particular interest, as their structure and composition can determine many properties of the bulk material, such as ductility, toughness and corrosion resistance [12–18].

In the early years of the FIM and the APFIM techniques, specimens were prepared by electropolishing to form a microtip geometry, which would permit both field ionization and field evaporation of specimens [6,19]. While successful, electropolishing alone does not readily permit site specific targeting of imperfections or mesoscale features. To address this problem, methods have been developed using transmission electron microscopy (TEM) to identify features of interest and then electropolishing repeatedly to position the feature of interest in the microtip, which is subsequently analyzed [19–25]. In addition to the pulse electropolishing approach, broad ion beams have been used to position features of interest within the microtip in materials that do not pulse electropolish reliably, such as cemented carbides [19,26]. These methodologies may involve many steps depending on the feature being investigated, and relies heavily on the desired feature occurring within a reasonable distance from the apex of the electropolished microtip. With the development of focused ion-beam (FIB) microscopes, it became possible to produce APT specimens with their microtips containing GBs and other features of interest, with both predictability and accuracy [27–31]. Various FIB–SEM-microscope based methodologies have been developed to lift-out sections of GBs and place them on single microtips or on silicon microrods residing on microtip arrays for analyses [27,28]. Alternatively, techniques involving FIB–SEM microscopy ion-beam milling and analyzing specimens in situ have also been successful for the study of different features, such as GBs and heterophase interfaces [29,32], which are central to the control of materials properties. In steels, for example, it has been demonstrated that segregation of elements such as copper, tin, antimony, phosphorus and sulfur to austenite GBs during hot working leads to a loss of ductility [12–14].

Despite their importance in determining material properties, microstructural features with a spacing in the 5–40 μm range (corresponding to ASTM grain size 6–12 in metals [33]) have not been analyzed readily with APT, due to difficulties in preparation of microtips using either electropolishing or existing FIB–SEM microscopy lift-out methods. An important example of these mesoscale features is prior austenite GBs, which serve as nucleation sites for...
carbides and intermetallic precipitates, further reducing ductility, toughness and corrosion resistance [13,15–18]; therefore, characterization of compositional changes at these features is critical for understanding the evolution of microstructures in steels. During an electropolishing procedure, a large number of specimens and repeated electropolishing steps are required to locate an interface. For example, in the methods presented by Henjered and Norden [19] and Krakauer and Seidman [20,22–25], an approximately 1–1.5 μm long section of the specimen is rendered electron transparent after each electropolishing step [19,22]. As such, if 500 nm of the viewed volume is removed with each polishing step, 20 electropolishing steps will be required to view 10 μm of the specimen. In addition to this large number of steps, materials with a mixture of microscale and mesoscale features, such as the lath boundaries and prior austenite GBs in martensitic steels, require more intensive analyses employing selected area diffraction (SAD), using a TEM, of each boundary encountered to determine whether or not it is of interest. While this approach is certainly possible it requires a significant amount of TEM analysis.

More recently, specimen preparation has been performed using FIB–SEM microscope lift-out procedures, which are more site-specific than conventional electropolishing techniques; standard lift-out techniques require, however, a straight and several micron long section of a GB to extract a specimen [27]. Cairney et al., Feller et al. and Tytko et al. have demonstrated methods that have successfully analyzed grains in the 5–40 μm size range, which involves lifting out a rod of material with a GB located near its end, then shaping the end of the rod into an APT specimen [34–38]. These techniques provide an excellent methodology for the fast, reliable and relatively damage-free preparation of samples containing GBs, but they have two major limitations when considering preparation methods for materials containing mesoscale features. First, it does not permit TEM observation of the broader microstructure in the vicinity of the tip to be prepared prior to shaping the APT microtip. This step can be critical for targeting the correct features in some materials, such as a martensitic steel with a small lath size, which is discussed in this article and maraging-TRIP steels [39]. Second, as with most FIB–SEM microscope techniques, the GB must be in a region that is fairly close to the surface of the bulk sample. While this typically is not an issue, samples where the GBs have been etched or otherwise corroded require that the GB must be in a region that is fairly close to the surface of the bulk sample. While this typically is not an issue, samples where the GBs have been etched or otherwise corroded require that the APT microtip be extracted from below the surface of the bulk sample to avoid the region of the GB that has been attacked by the etchant.

In this article, we present a methodology for analyzing mesoscale features, such as GBs or heterophase interfaces, at depths of several microns below a sample surface. The methodology involves targeting the feature utilizing the ion channeling contrast generated by the interaction of the ion beam with different crystal orientations, followed by an iterative FIB–SEM microscope ion-beam milling and TEM imaging process [34–36]. The FIB lift-out specimen produced is significantly larger than used in other approaches so that the atom probe specimen can be formed from beneath the layer initially damaged by the ion channeling contrast imaging, as the Ga⁺ damage is dependent only on the ion acceleration voltage and sample crystallography [34–36]. Our new approach permits either GBs or heterophase interfaces, in this case prior austenite GBs spaced approximately 10 μm apart, amidst the lath boundaries, to be located precisely in the lift-out specimen prior to the formation of an APT specimen. In this manner, the probability is increased of capturing the desired mesoscale feature in the microtip of an APT specimen and readily identifying the correct boundary employing a single-tilt TEM specimen holder. Samples prepared utilizing this methodology display a high success rate in locating the desired GB in the analyzed region for analysis by TEM and subsequently studying it during an APT analysis.

This methodology as demonstrated in this paper, can be used to select desired mesoscale features from among large numbers of microscale features at depths of several microns below the surface of the sample, which has applications beyond grain boundaries. Irradiated materials [40,41], such as nuclear reactor steels, can be analyzed at a specific depth with the specimen’s z-axis parallel to the surface of the sample to ensure that all features within the dataset received the same dose or to analyze a section of a GB at a specific depth from the irradiated surface; current methods only allow for microtips to be prepared parallel to the bulk sample surface if they are extracted from a region very close to the bulk sample’s surface. Thin-films and semiconductor device structures [42,43], such as electrode-gate-substrate triple points, can also be prepared with the device-substrate interface parallel to the analysis direction in the APT to minimize the probability of the specimen rupturing during analysis.

2. Samples and experimental apparatus

A martensitic ultrahigh strength steel, currently marketed as PremoMet™ by Carpenter Technology Corporation, was used to validate the methodology presented in this article. This steel was austenitized at 1191 K for 1.5 h, quenched, cryogenically treated at 200 K for 8 h and tempered at 533 K for 2 h. The microstructure contains a prior austenite grain diameter of approximately 10 μm and a sub-micron martensitic lath diameter. As such, the features of interest, the prior austenite GBs, must be precisely selected and distinguished carefully from among the more numerous lath boundaries.

FIB–SEM microscopy ion-beam milling was performed using a FEI Strata DB-235 dual-beam FIB microscope equipped with an Omniprobe Model 100.7 micromanipulator. The ion-beam column in this instrument permits Ga⁺ ion beam milling at either 5 or 30 kV. Ar⁺ ion milling was also performed in a Fischione model 1010 ion mill to further thin the lift-out specimen. TEM imaging was performed in a JEOL JEM-2100 with a LaB₆ electron source, a standard JEOL double-tilt specimen holder, and a Fischione Model 2050 on-axis rotation tomography holder designed to hold wire specimens and permit 360° rotation of a specimen. The APT analyses were conducted utilizing a LEAP 4000X-Si ( Cameca, Madison, WI) at a base temperature of 80 K, a pulse energy of 20–50 pJ, a pulse repetition rate of 500 kHz, and an ion detection rate of 0.20 to 0.5 ion pulse⁻¹. This instrument utilizes a local-electrode and laser pulsing with a picosecond 355 nm wavelength ultraviolet laser, which enables the analysis of insulating materials and minimizes specimen fracture [44].

3. Specimen preparation

The specimen preparation technique can be separated into three distinct phases: (A) feature-specific lift-out and observation by TEM; (B) transfer to an electropolished wire; and (C) a final specimen sharpening procedure based on the TEM images and examination of the microtip in TEM. Specifically, the methodology involves: (1) locating a boundary of interest using ion-channeling contrast; (2) extracting the specimen with the micromanipulator and attaching it to an Omniprobe grid; (3) thinning the sample until it is electron transparent using a FIB–SEM microscope and Ar⁺ ion milling; (4) imaging utilizing TEM to locate the mesoscale feature of interest, here a prior austenite grain boundary; (5) transferring the specimen onto an electropolished wire suitable for loading into the APT; (6) cutting the specimen, using ion-beam milling, based on the TEM images to target the feature of interest; (7) annular milling to form an APT microtip of the desired location; and (8) observation of the final APT microtip with TEM by capturing a TEM tilt series for
direct correlation with the APT analyses, which involves characterizing the GB and its location in the APT microtip. The sample is imaged in step 4 using a standard TEM sample holder, and in step 7 using the Fischione on-axis holder. After this preparation protocol, the microtip is transferred to the APT for analysis.

3.1. Feature-specific lift-out

The first step of preparation is to locate the region of interest (ROI) using secondary electron imaging. After the area has been located, the GB may be targeted by operating the ion beam at a low brightness/high contrast setting to enhance the ion-channeling contrast. This method works well with martensitic steels, where it permits prior austenite GBs to be identified by following the morphology of the prior austenite grains, Fig. 1a. Ion imaging may not be required in materials that form a passivation layer because the grain structure may still be resolved by electron channeling contrast in spite of, for example, the existence of a thin oxide layer. Gallium ion implantation for this method is less of concern than in other APT preparation techniques as the microtip is formed from material that is several microns below the exposed surface of the sample. Transport of ions in matter (TRIM) [45] calculations for 30 kV Ga\(^+\) ions in pure Fe and the alloy discussed in this article yield a stopping range of approximately 11 nm, which is significantly less than the 27 nm reported for Si, although ion channeling and radiation-damage enhanced diffusion can increase the implantation depth by up to 20 times [46–48]. Therefore, if ion channeling contrast is used gallium implantation into the micropip can be avoided by forming the micropip from a feature 300 nm or more below the surface that was imaged.

Once the ROI is located, a standard TEM sample is prepared perpendicular to the GB of interest. A nominally 3 × 20 × 1 \(\mu\)m\(^3\) thick Pt capping layer is deposited to protect the sample from Ga\(^+\) ion implantation, Fig. 1b. Stepped trenches, used to reduce milling time, 22 × 12 × 6 \(\mu\)m\(^3\) are then ion milled on either side of the Pt cap with the bulk sample tilted normal to the Ga\(^+\) ion beam. The edges of the lift-out specimen are trimmed by milling a 24 × 3 × 4 \(\mu\)m\(^3\) rectangular pattern on both sides of the specimen, tilting the sample ±1.5\(^\circ\) as needed to expose the area to be trimmed, to produce parallel sides on the specimen.

The specimen undercuts are made at a 7\(^\circ\) stage tilt angle, after which the micromanipulator is inserted at 0\(^\circ\) tilt and attached to the lift-out specimen before it is Ga\(^+\) ion milled free from the bulk sample. This specimen is then transferred to an Omniprobe grid and Pt-welded on each side. Thinning is accomplished with a pattern which mills each raster line of the pattern to full depth before moving to the next raster line, located closer to the center of the specimen, using a 300 pA Ga\(^+\) ion-beam current. The specimen is first tilted to −1.5\(^\circ\) to mill the side not visible to the SEM beam, then the final milling is completed for +1.5\(^\circ\), so that progress can be monitored using the SEM beam. The final thickness of the specimen is approximately 250–300 nm; below this thickness the martensitic steel samples begin to bend due to residual stress and ion implantation [49,50], making later thinning and specimen manipulation more difficult.

Following thinning the specimen is removed from the FIB–SEM microscope and transferred to the TEM. Even though the specimen is relatively thick for TEM, the martensitic lath structure is still resolvable using a large condenser aperture and a slightly condensing the electron beam, Fig. 1c. The micrographs captured at this stage are saved for later use in the specimen sharpening step to target the prior austenite GB. The GB of interest is identified in the TEM micrograph by tracing a dividing line between lath orientations, and is located in the FIB–SEM microscope using the edge contours of the TEM sample as a reference, Fig. 1d.

Additionally, some of the specimens were subjected to Ar\(^+\) ion milling at this stage to thin the specimen further. The specimen shown in Fig. 4 underwent argon milling for a total of 10 min at

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**Fig. 1.** (a) FIB–SEM microscope ion-beam image showing the microstructure of the martensitic steel with selected prior austenite grain boundaries indicated by arrows. (b) Pt capping layers for TEM samples placed perpendicular to grain boundaries. (c) TEM image of a thick lift-out, with a visible lath structure. (d) The same image, contrast enhanced, with a section of the prior austenite grain boundary marked with a dotted line and the desired location for the APT sample marked with a white arrow. White lines connect the region of interest with the edge topography.
5 kV, 10 min at 3 kV, and 8 min at 2 kV; all steps utilized 5 mA. We have found, however, that argon ion milling of the specimen should be avoided when possible while the specimen is in the TEM lift-out specimen geometry due to potential damage to the specimen.

3.2. Transfer to electropolished wire

After TEM imaging, the specimen is transferred back to the FIB–SEM microscope, and the Omniprobe is attached to the post of the Omniprobe grid, using electron-beam deposited Pt, on which the specimen is located. The section of the grid post attached to both the specimen and Omniprobe is then cut free using the a 30 kV 20 nA Ga\(^+\) ion-beam current, the largest aperture available on the DB-235 FIB–SEM instrument, to minimize the time needed to ion mill through the thickness of the grid post, Fig. 2a.

During the steps described, the specimen is not imaged, as the high Ga\(^+\) ion currents used to cut the grid can render the sample unusable due to Ga ion implantation into the sample along its thinnest dimension. To prevent Ga\(^+\) ion implantation, the opposite side of the grid post from the specimen is initially used to focus the Ga\(^+\) ion beam. The Ga\(^+\) ion beam is then shifted until the Pt weld holding the specimen to the post is observable. The micromanipulator is then moved to the grid post near the specimen weld using only the electron beam and attached with e\(^-\)-beam deposited Pt. The 20 nA Ga\(^+\) ion beam current aperture is selected and the Ga\(^+\) ion beam is again focused onto the far side of the post. The micromanipulator is used as a fiducial marker to prevent accidental exposure to Ga\(^+\) while shifting the Ga\(^+\) ion beam back to the region to be milled. The first two cuts are made in this position, after which the Ga\(^+\) ion beam is shifted below the plane of the specimen, then shifted to the edge of the post to finish detaching the section containing the sample.

After the section of the grid containing the specimen of interest is lifted out, the chamber is vented, and the Omniprobe lift-out stage is removed and replaced with a custom fabricated stage that securely holds a 1.02 mm diam. wire in either vertical or horizontal positions. An approximately 20 mm long tungsten wire segment, sharpened by electropolishing in potassium hydroxide [51–53], is placed in the stage in the horizontal position, and the chamber is re-evacuated. The tip of the wire, now aligned perpendicular to the Ga\(^+\) ion beam, is milled flat in a region of approximately the same diameter as the section of the grid post that has been removed. The grid section containing the specimen is then placed flush with the wire tip and attached with Pt on both sides; first on one side, Fig. 2b, then after releasing the micromanipulator, the stage is rotated 180° and the Pt is deposited on the opposite side of the wire/grid-section connection. The chamber is then vented, the wire moved from the horizontal to the vertical position, and the chamber re-evacuated for the final specimen sharpening stage, described below.

3.3. Final specimen sharpening

The electron beam is focused on the specimen (the highest point on the sample stage) for the final sharpening procedure,
after which the stage is rotated and tilted to accommodate any small mis-alignments, such that the specimen sits in a precisely vertical position, with its broad side parallel to the electron-beam and facing the Ga⁺ ion-beam. The 50 pA Ga⁺ ion-beam is then focused on the grid section, turned off, translated over the specimen and a single high-resolution image recorded, Fig. 3a. The edges of the specimen are then used as fiducial markers with which the TEM images are aligned with the FIB images, and the specimen is Ga⁺ ion beam milled into a wedge shape with the ROI just below the apex of the wedge, Fig. 3b.

Pt is electron-beam deposited on the tip of the wedge in preparation for the final thinning step, after which the specimen is tilted so that the Ga⁺ ion beam is parallel with the specimen’s long axis. An annular milling pattern is utilized to form the wedge into an APT specimen using a 0.50 μm inner radius pattern at 300 pA beam current followed by a 0.15 μm inner radius pattern at 50 pA beam current. A final step is performed at 5 kV, the lowest Ga⁺ ion acceleration voltage available in the DB235, using 3 μm circular pattern and a slightly defocused ion beam. This step is performed to remove as much of the radiation region damaged by the 30 kV Ga⁺ ions as possible [27], to remove the remainder of the Pt cover to prevent specimen fracture [54], and if required to slightly erode the micropit to precisely position the GB. The defocused Ga⁺ ion-beam with a circular pattern is utilized because it has been found to maintain the shape of the micropit, similar to in broad beam ion milling of surfaces [55], while a focused beam has been found to introduce asymmetries while milling at 5 kV in the DB-235 FIB–SEM microscope. A bright-field TEM tomographic tilt series is recorded after the final ion-milling step, two TEM images are displayed in Fig. 4e and f, using the Fischione on-axis rotation tomography specimen holder to load the electropolished wires on which the APT specimens sit.

4. Results and discussion

4.1. Analysis of PremoMet steel

A total of five APT specimens of PremoMet steel have been prepared using this method, from one of which 49 million ions were collected by APT, without the specimen fracturing (Appendix A). This specimen is presented in Fig. 4, with reconstruction containing carbon shown in (a), argon in (b), gallium in (c) and (d), TEM images at two tilts shown in (e) and (f), gallium concentration profiles in (f) and (g) and concentration of carbon, argon and gallium across the prior austenite grain boundary in (i). The carbon and argon segregation displayed in Fig. 4a clearly highlights the prior austenite GB displayed in Fig. 4e and f. Additionally, Fig. 4b indicates that the Ar⁺ ions from the exploitative ion-milling penetrated deep within the approximately 300 nm thick sample at the 2–5 kV accelerating voltages, even though the TRIM calculations yield a stopping range in iron of 4.0 nm (skewness=0.63, stragggle=2.2 nm, kurtosis=3.22) for 5 kV Ar⁺. This is likely due to the increased acceptance angle for ion channeling at low energies followed by diffusion to the grain boundary [56–59] although other explanations are possible, such as species specific enhanced grain boundary diffusion similar to Ga in the aluminum alloy system [60,61]. The bright-field TEM micrographs in Fig. 4e and f are from the TEM tilt series, displaying two tilts with the grains on either side of the prior austenite GB, which highlights the curved nature of the prior austenite GB between it and the adjoining grain. Fig. 4g is an APT 1-D concentration profile obtained using an analysis cylinder placed perpendicular to this GB, which demonstrates that the carbon is enriched to 10.75 at%, while the argon concentration along the GB from the ion-milling step is 0.56 at%. Further information on the composition of the prior austenite boundary shown will be provided in a future article [62].

The TEM micrographs provide the requisite information for calibrating the APT reconstruction dimensionally, which can be performed in the absence of crystallographic poles in the field-desorption images. Reconstructions of the dataset with different reconstruction parameters were overlaid onto the TEM micrograph using Adobe Photoshop; the difference in magnifications between the TEM micrograph and the APT 3-D reconstruction was then calculated using ImageJ [63] (Appendix B). The dataset displayed, Fig. 4, contains both the GB and a vanadium carbide precipitate, which serve as fiducial markers for calibration purposes. To minimize errors due to a possible relative rotation of the APT 3-D reconstruction about its z-axis relative to the TEM micrograph, only the z-axis component of distances between points are used to evaluate the fit and serve as the best possible reference points between the TEM image and APT reconstruction, as the z-axis aligns closely with the axis of rotation of the TEM holder.

Based on this calculation, an evaporation field of 29.5 V nm⁻¹ at an estimated field factor of k = 3.3 and an image compression factor of 1.65 provides the best fit along the z-axis, while maintaining an appropriate angle of the GB with respect to the principal axes of the 3-D reconstruction. The evaporation field, 29.5 V nm⁻¹, is 165 smaller than the empirical value for iron of 35 V nm⁻¹ [9–11,64]; the recorded base temperature of 80 K is, however, lower than the actual specimen temperatures during UV laser-assisted field evaporation due to heating by the laser beam (Appendix C). This lower evaporation field based on elevated temperature is consistent with the results of Wada and Arslan et al. [65,66]. Finally, the calibration methodology herein is only an estimate, while other methods based on field-desorption images provide greater precision [66–70].

The Ga⁺ implantation into the analyzed volume was minimal in this procedure, Fig. 4c, d and h, i, with no Ga concentration along the GB as occurred with Ar, Fig. 4b. This difference was likely due to the fact that only three images were recorded at 50 pA with the Ga⁺ ion beam at a significant angle to the thinnest part of the sample, whereas the sample underwent 28 min of Ar⁺ ion milling, even though the projected stopping range for the kV Ar⁺ is only 4.0 nm as previously described. The majority of the Ga implantation is confined to a small region on one side of the apex of the micropit, Fig. 4c and d. The dotted lines in Fig. 4d correspond to the direction of the concentration profiles for Ga, Fig. 4g and h, which indicate a maximum Ga concentration of 3.68 at%; this is greater than the maximum of 2 at% found by Thompson et al. [27] using their site-specific technique; it decreases, however, to < 2 at% at 3 nm and <1 at% at 8 nm from the edge of the dataset. The total Ga concentration in the dataset is 0.036 at%, and the concentration in the vertical center of the dataset, Fig. 4d, is 0.027 at%. TRIM calculations yield a stopping range in iron of 10.9 nm (skewness=0.68, stragggle=5.3 nm, kurtosis=2.59) for 30 kV Ga⁺. The target is, however, amorphous in the TRIM calculations; ion channeling in a crystalline material results in much greater penetrations. Materials with a smaller atomic number, such as silicon, also have a larger stopping range, which increases the probability that Ga⁺ ions will penetrate to the region or feature of interest. Therefore, great care must be taken to minimize Ga⁺ ion exposure, even though the region calculated to be most heavily damaged is removed during the annular milling step, as the sample is about 300 nm thick when the image during the final sharpening procedure is recorded.

4.2. Proposed combinations with other analysis methods

As demonstrated in the preparation section, our procedure can be used successfully in conjunction with other characterization techniques, such as TEM. Since the specimen is kept relatively
thick to prevent bending, while on the Omniprobe grid, electron backscattering diffraction (EBSD) may also be employed to analyze the orientation relationships within the specimen, especially if the FIB–SEM microscope utilized is equipped with an EBSD system. Orientation imaging microscopy (OIM) in the TEM could also be used in conjunction with this method, but will be significantly more difficult due to the specimen remaining thicker than is optimal for this technique. These techniques can be used to characterize the orientation relationship at the grain boundary selected for APT analysis [71].

EBSD may be performed on the bulk sample surface before the lift-out process described herein, to select a grain boundary or heterophase interface, as an alternative to using ion channeling contrast [71]. This variation of the outlined methodology could be used when ion channeling is of great concern, such as during the lift-out of GBs in a corroded sample; we have shown, however, that Ga implantation during the initial step of this technique is generally not an issue due to the depth from which the sample can be taken, even though TRIM calculations are known to underestimate the penetration of gallium due to ion channeling and radiation assisted diffusion.

The specimen preparation on a sharpened wire suitable for a TEM on-axis tomography holder is important since it allows a microtip to be easily manipulated, while minimizing the possibility of mechanical damage during handling. This specimen geometry also facilitates examination of a specimen after preparation by recording a TEM tilt series to confirm the presence and location of the mesoscale feature of interest, and in principle permits the creation of an electron tomographic reconstruction if a suitable contrast mechanism is available [66].

### 4.3. Comparison to current techniques

Our results demonstrate that this specimen preparation technique provides a precise method for targeting mesoscale features in the 5–40 μm size range located several microns below the sample surface. The original pulsed electropolishing techniques provided a methodology to locate features of interest within an APT specimen volume; they function, however, as an iterative selection tool, where the sample is polished until a feature of interest is located and positioned in the volume of the microtip to be analyzed [19,20,22]. As discussed, these methods require multiple polishing steps, each of which is accompanied by extensive TEM diffraction analyses to locate the mesoscale feature of interest, especially so in the case of martensitic steels due to the large number of lath boundaries contained within each grain. While these methods have proven to be successful, they do not provide the most convenient route for preparing site-specific samples by current standards.

FIB–SEM microscope based techniques, Kelly et al. [72] and Thompson et al. [27], provide methods to lift-out areas containing specific GBs, and the newer method described by Felfer et al. [36] integrates TEM into the FIB–SEM microscope preparation procedure. These methods, however, do not provide information on where the feature is located below the sample's surface. By first lifting out a relatively large TEM sample, the method demonstrated in this article permits a mesoscale feature to be precisely selected from its surrounding mesoscale features at a depth of several microns below the surface. This new capability is useful for several problems. First, it permits the use of ion-channeling contrast, as the sample can be extracted at depths greater than the maximum range...
of an implanted Ga atom. Second, specimens can be prepared from features such as crack tips without the crack or pre-crack oxide running through the rest of the sample as in other methods [73,74]. Moreover, these techniques suggest preparation from a plan view of the crack, which requires a location that is only partially corroded or cracked to ensure that an APT specimen can be fabricated from near the crack’s tip. By preparing the TEM specimen in a cross-sectional geometry across the crack, the method demonstrated herein provides a route for targeting the region immediately below the crack tip instead of a section of a GB or interface that, while next to a section that was corroded, was not itself attacked.

5. Conclusions
- A new method for the preparation of atom-probe tomograph microtips containing mesoscale features for correlative investigations employing atom-probe tomography (APT), transmission electron microscopy (TEM) and additional auxiliary methods is demonstrated.
- This preparation technique is applied to the study of prior-austenite grain boundaries (GB) in PremoMet, a commercial martensitic ultrahigh strength steel alloy, and the specimens created with this technique are shown to field-evaporate smoothly and produce useful APT and TEM datasets.
- Lift-out specimens are prepared perpendicular to the grain boundary, analyzed in TEM, and a microtip containing the GB is prepared based on the TEM analysis.
- The APT microtips produced by this method are well suited for analysis using an on-axis tomography holder in a TEM. Calibration of the APT dataset using the TEM images is demonstrated.
- The need to avoid extended Ar⁺ ion milling, often used for cleaning TEM foils, is also discussed, as the 3-D reconstructed APT dataset clearly shows argon concentrated along the grain boundary.
- This technique permits the preparation of specimens containing mesoscale features, such as GBs, from materials in which their spacing and the proximity of other features renders both electropolishing and traditional lift-out techniques difficult.
- Potential future applications include the study of specific grain boundaries that have experienced corrosive attack, hydrogen embrittlement or irradiation, as well as semiconductor device structures and interfaces.

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Appendix A. Supplementary information
Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.ultramic.2014.05.005.

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