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Efficacy of a FIB and Planar Preparation Technique for Producing TEM Lamella of CLS

by Samuel G Hirsch

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DEVCOM Army Research Laboratory

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<p>An unconventional method for creating transmission electron microscopy (TEM) lamella via focused-ion beam (FIB) was evaluated. While the conventional method involves using the FIB to mill and extract a lamella normal to a material's surface, the method tested here involves extracting a lamella in plane with the surface. The benefits of using the unconventional, or planar, method and when it might be used are contrasted with the conventional method. As an example, a TEM-quality lamella is made from a cut and polished bulk sample of calcium lanthanum sulfide using the planar method. Details of the method and parameters used in the procedure, as well as TEM analysis, are discussed.</p>					
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1. Introduction

The conventional method of preparing lamella for transmission electron microscopy (TEM) via focused-ion beam (FIB) is the cross-sectional (X-section), lift-out procedure. In this method a region of interest (ROI) is selected, and the FIB is used to mill perpendicularly into the surface, usually several microns deep. A lamella is then extracted resembling a slice of cake (Fig. 1a). This method is necessary for looking at layers and fine for homogeneous materials with no distinguishing features or grains much smaller than the lamella itself. However, this is less so for inhomogeneous materials that may display important features on the surface, which cannot be assured to extend below. In this technical note an unconventional FIB procedure was used to extract a section of the material in plane with the surface (Fig. 1b).^{1,2} This so-called planar-, or plan-view, method attempts to produce a lamella parallel to the surface, only 100–200 nm deep from the surface after final thinning to electron transparency. Ideally, this method will provide data that is more representative of the ROI. This plan-view method was implemented and tested for efficacy in the microanalysis of a feature on the surface of a cut and polished sample of hot-pressed calcium lanthanum sulfide (CLS).

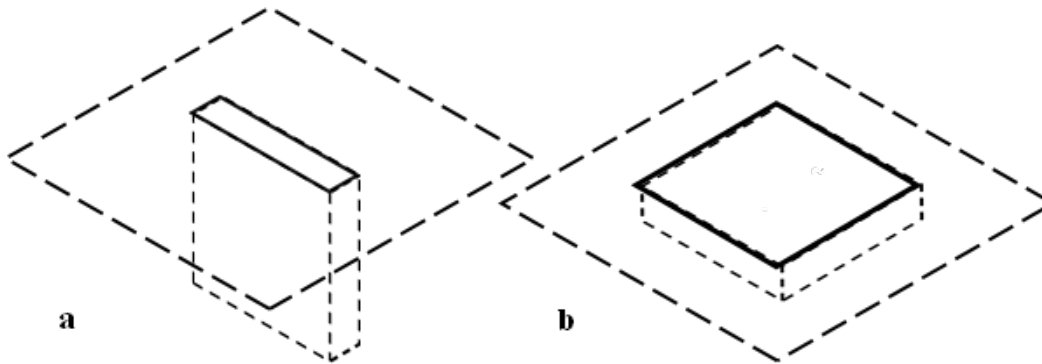


Fig. 1 Lamella with respect to the surface: (a) normal to, (b) in the plane of

2. Background

Figure 2 illustrates one issue with using the X-section method. Figure 2a shows an ROI where three phases of material are identified on the surface. In Fig. 2b we see that upon milling into the sample, the phases shift to the left, deeper into the material, with the darker phase exhibiting the majority of the exposed material. The lightest phase in the upper left occupies only a small fraction of the exposed face and, being on the very edge may become even smaller after milling for lift-out and final thinning of the lamella.

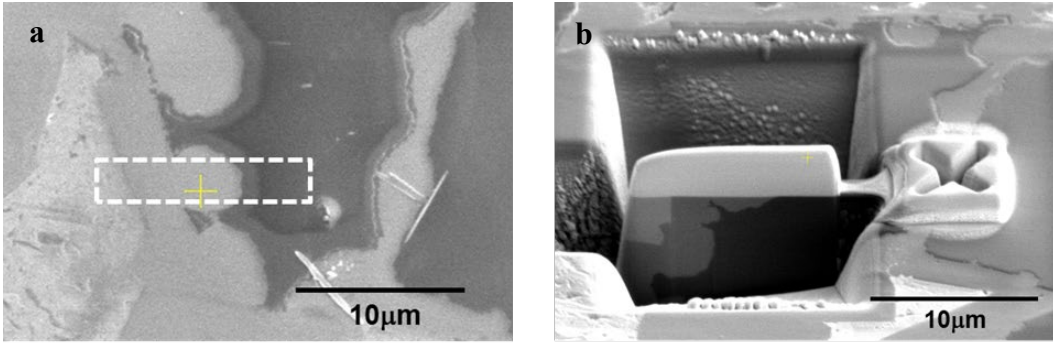


Fig. 2 (a) Dashed line indicating region from which a lamella was milled and where three separate phases are evident. (b) After FIB milling showing how the phases extend into the material below the surface.

Figures 3 and 4 illustrate some advantages of using the planar method. Figure 3a displays three large grains that come to a triple point. First, if the grains are on the order of or larger than the size of lamella that can be made it would be very difficult to come across a triple point using the conventional method. Second, by being able to identify the triple point on the surface it can be centered in the middle of the lamella. If investigating a grain boundary, it too can be centered. In Fig. 3b there are many small grains, each of the same phase and each with their own crystallographic orientation. Using electron backscatter detection these orientations can be identified beforehand and then selected as to which may be of interest before milling.

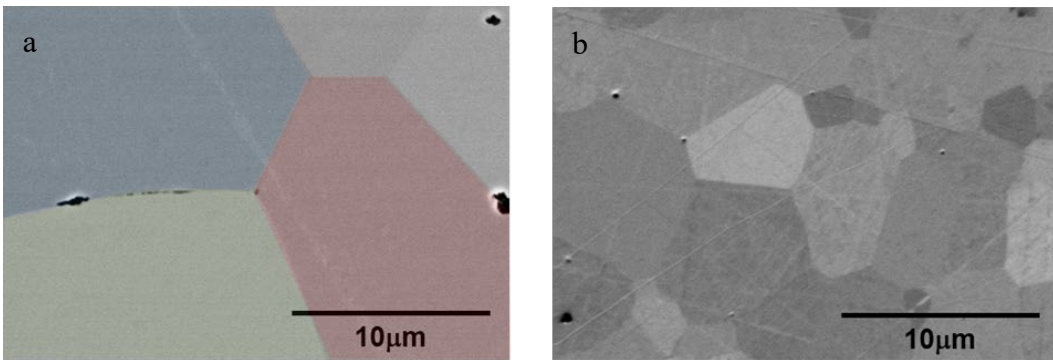


Fig. 3 (a) Scanning electron microscopy (SEM) micrograph with pseudo color added to highlight triple point at intersection of three large grains. (b) SEM micrograph showing grains of the same phase with different crystallographic orientations.

Figure 4 shows two types of inclusions. In Fig. 4a a group of crystallites is clearly shown on the surface but with no indication of how deeply they may penetrate the bulk. If they happen to be shallow there may be very little material to work with. By using the planar method, however, the chances of including those crystallites in the final lamella improve even if they are shallow. Figure 4b shows a circular, or more likely spherical, inclusion where different phases are distributed

asymmetrically. Milling a single slice across the width of the ROI (dashed line) using the X-section method would destroy most of the inclusion and not guarantee that all the phases would be present in the final lamella. But with a planar lift-out there is good chance the entire area (rectangular line) can be lifted out and a full analysis carried out.

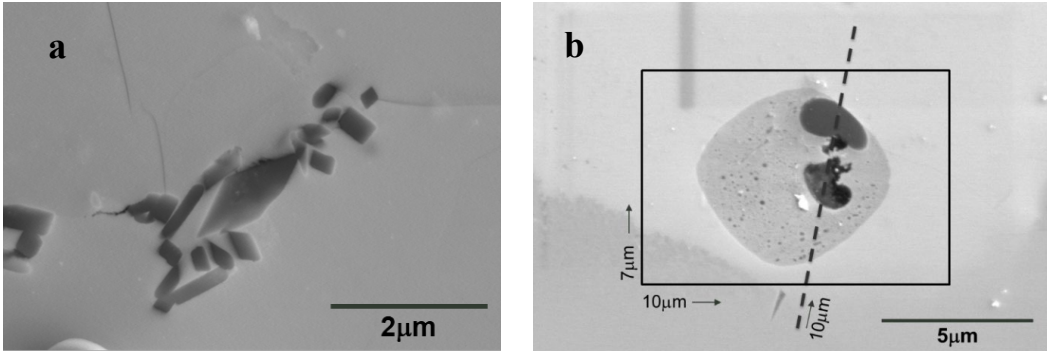


Fig. 4 (a) Inclusion made of several small crystallites on the surface. (b) Inclusion with micron- and nano-sized crystallites dispersed asymmetrically. Dashed line representing the location of a possible X-section lift-out. Solid line representing an area from which a planar lift-out could be made.

3. Experimental Procedure

The equipment used in the procedure to create the planar lamella was a Thermo Fisher Scientific Helios G4 UX Dual Beam state-of-the-art microscope obtained in 2019. It features a field-emission SEM along with a gallium-ion beam, each with maximum acceleration voltage of 30 kV. The manufacturer's default tilt angle between the two beams is 52°. The increased amount of current available, up to 63 nA on the G4 UX over older FIB models, offers greater depth of milling in a shorter amount of time. Without this important improvement this method would be impractical time wise—especially with materials that mill slowly. Also, the FIB was equipped with a Thermo Fisher Scientific EasyLift Nanomanipulator for lift-out, whose speed and accuracy have vastly improved over older models, making extraction of the lamella easier and faster.

For this technical note a cut and polished bulk sample of hot-pressed CLS, with a composition of CaLa_2S_4 , was used. Figure 5 shows the ROI, identified by the dashed line, that was chosen to include a triple point with unidentified phases at the center and along the grain boundaries. It was anticipated that the oblong dark area in the center was likely a void and unlikely to significantly widen just below the surface.

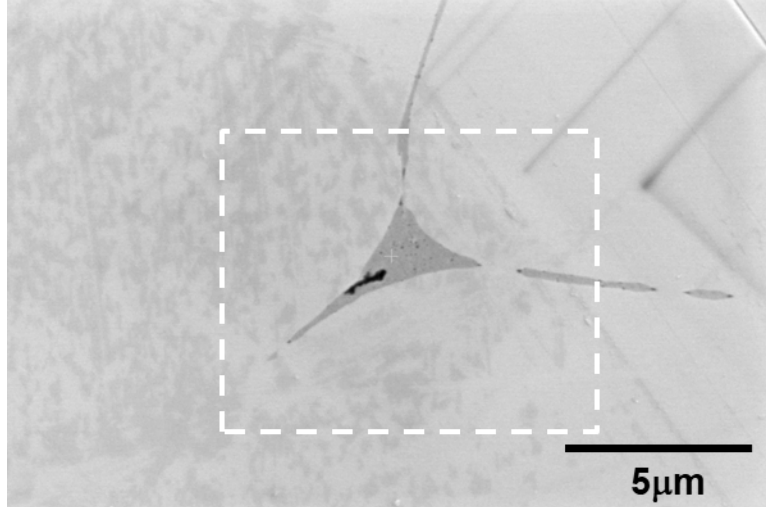


Fig. 5 ROI chosen for lift-out

Following is a description of the lift-out procedure and many of the associated parameters used. A full list of all the parameters used are listed in Table 1.

Table 1 Process parameters

Action	Beam	V (kV)	I (nA)	x (mm)	y (mm)	z (mm)	Thick	Cut
Pt deposition	e ⁻	5	1.6	10	8	1.0	1.5 mm	...
Pt deposition	i ⁺	30	0.44	10	8	1.5	1.0 mm	...
Trenching	i ⁺	30	9.1	5 * y	x * 5	5.0	...	RC
Undercuts	i ⁺	30	2.4	16	2	9.0	...	CC
Pt deposition	e ⁻	5	1.6	10.5	1	1.0	1.0 mm	...
(over front edge)	i ⁺	30	0.090	10.5	1	1.0	1.0 mm	...
Wedge removal (target rotated 180°)	i ⁺	30	2.4	10	3	7.0	...	CC
Thinning								
Pt face	i ⁺	30	1.2	10	0.5–1.0	8.0	...	CC
(tilt < +1.0°)			0.75	10	0.5–1.0	7.0	...	CC
			0.44	10	0.5–1.0	7.0	...	CC
			0.26	10	0.5–1.0	7.0	...	CC
Reverse side	i ⁺	30	1.2	10	0.5–1.0	8.0	...	CC
(target rotated 180°)			0.75	10	0.5–1.0	7.0	...	CC
(tilt ≤ +1.0°)			0.44	10	0.5–1.0	7.0	...	CC
			0.26	10	0.5–1.0	7.0	...	CC

Notes: e⁻ = electron beam; i⁺ = ion beam; RC = regular cross section; CC = cleaning cross section

As with a conventional lift-out, a protective layer of platinum was applied over the ROI. Usually, a thin coat of approximately $0.5\ \mu\text{m}$ is first put down by the electron beam followed by a thicker coating of approximately $2\ \mu\text{m}$ with the ion beam. For this technical note, however, the initial coating was $1.0\ \mu\text{m}$ followed by a $1.5\text{-}\mu\text{m}$ coating. This change was made to account for drift that occurred while using the ion beam. This drift was probably due to excess charge buildup on the surface as CLS is an electrically insulating ceramic. The area covered was $8 \times 10\ \mu\text{m}$, which is four times more than would normally be used for the conventional method with a corresponding increase in time. This equated to about 20 min versus the standard 5 min.

For milling trenches around the ROI, rectangular boxes were set up along each edge of the ROI with widths extending out from its edges set at $5\ \mu\text{m}$, leaving a bridge on the right-hand side for holding the sample in place when attaching to the EasyLift (Fig. 6). Also, an area opposite to the bridge was milled to allow the EasyLift to abut the target below the surface. This is sometimes necessary to improve attachment of the target to the EasyLift. The milling mode used for trenching was the “regular cross-section pattern,” as it is termed in the Helios software. The Helios default rescanning parameter of four times was used, which improved the efficiency of milling and reduced the milling time. As CLS is a rather soft ceramic material, the Z dimension, which affects the milling depth, was set at $5\ \mu\text{m}$. Trenching needed to be wider than for the conventional method as two deep cuts underneath the target were needed to separate it from the bulk. The depth of these cuts is important as redeposition can be an issue with large samples. Figure 7 shows the results from trenching.

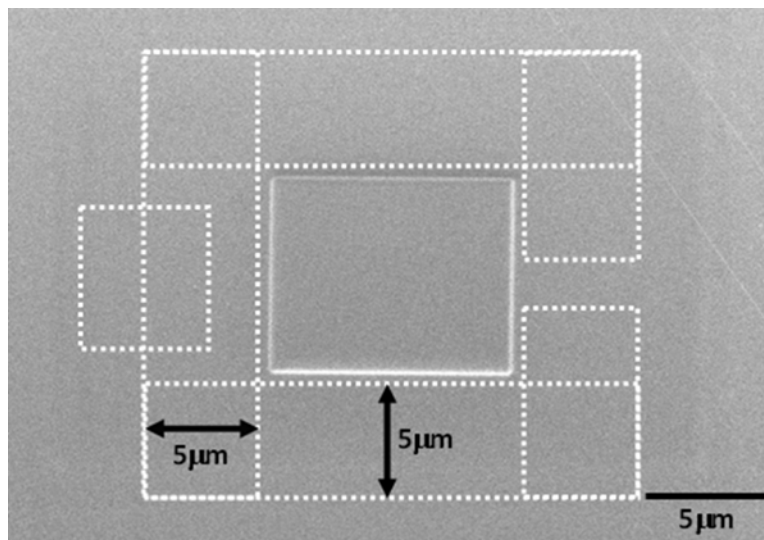


Fig. 6 Patterns used for milling out trenches

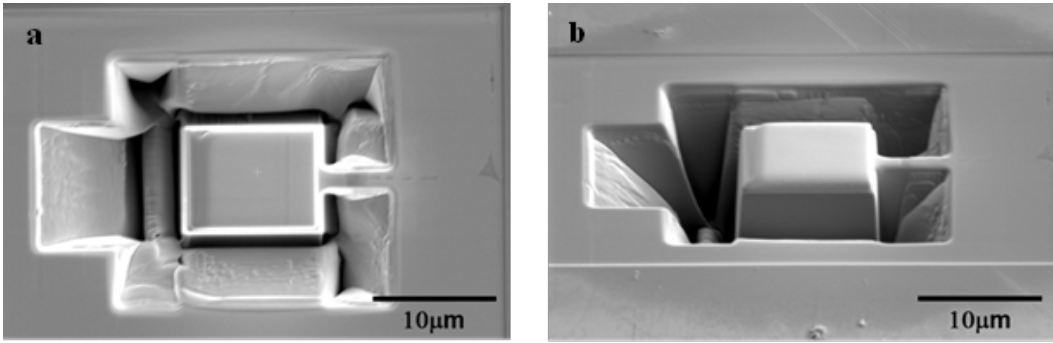


Fig. 7 (a) Top view and (b) side view after milling

During the trenching process a void began to show on the reverse side, as noted by the dashed line in Fig. 8. This is not something that could have been foreseen a priori and using a conventional lift-out might have jeopardized the entire process with the sample falling apart while thinning.

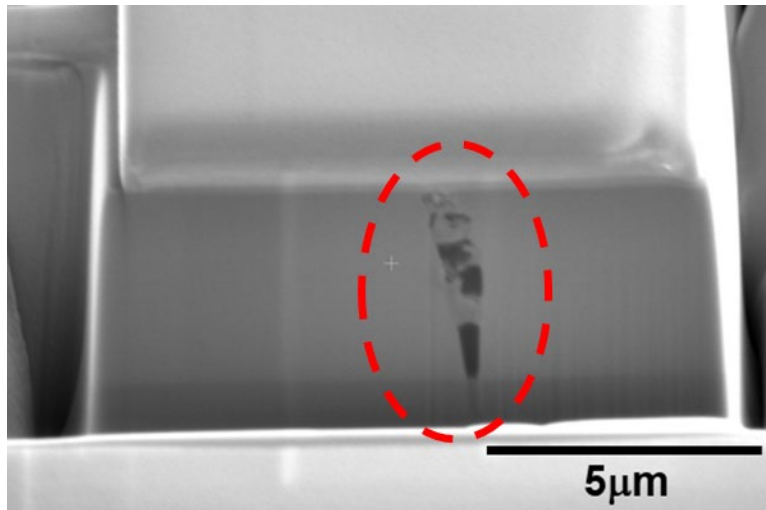


Fig. 8 Void revealed on reverse side

With the trenches now cut deep and wide, two narrower but deeper cuts are made underneath the target, parallel to the longer sides. The angle of the cuts must balance the amount of material needing to be removed, the amount of possible redeposition, and the amount of material remaining on the target that will be removed during the thinning process. Most FIBs are limited by the manufacturer's range on the tilt axis. The Helios G4 UX is limited to 60°. Also, with today's higher current FIBs, removing the extra material from the wedge before thinning will take relatively little time. Somewhere around 45° is usually the optimum angle and was used here. It is important to make two clean cuts all the way through the material so as not to have to recut and risk creating more redeposition, which then must be cut again and again (Fig. 9).

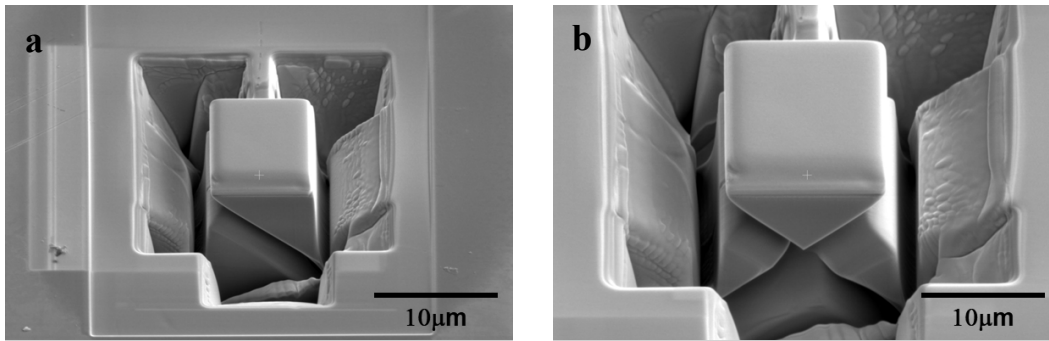


Fig. 9 Side view after making (a) one undercut and (b) after both undercuts

Once the two cuts are made, extracting the target is the same as with the conventional cross-sectional method. The target surface is aligned normal to the electron beam, the EasyLift is attached to the target with platinum, and the bridge is cut (Fig. 10).

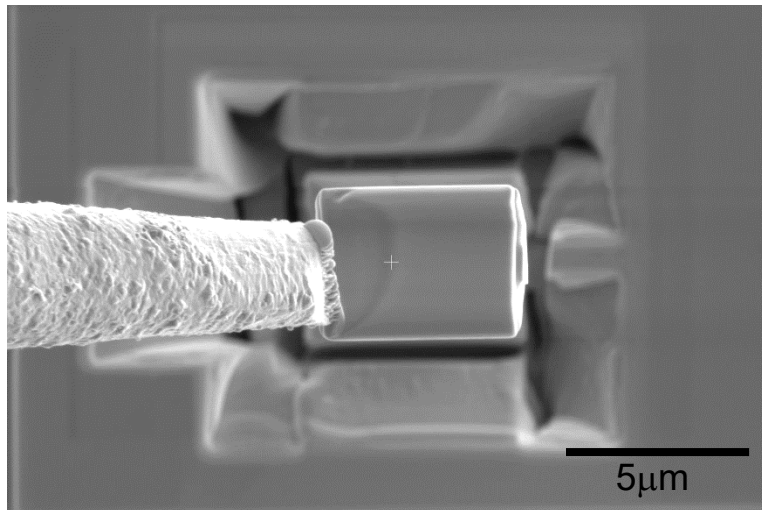


Fig. 10 Target attached to EasyLift and extracted from the surface

With the target now tilted at 90° from the conventional lift-out, the TEM grid must also be tilted to 90° to align the two in parallel for attachment. This was achieved by attaching the grid holder to a stage mount already angled at 45° , then tilting the whole stage another 45° on the tilt axis of the FIB. Attaching the target with platinum was then straightforward. After attachment another protective layer of platinum was added on top of the target as would be needed in a conventional cross section. The width in the Y direction need only be $1\text{--}2\ \mu\text{m}$ and extend from the edge of the surface platinum back toward the wedge (Fig. 11). Again, a thin layer was deposited using the electron beam followed by a thicker layer with the ion beam.

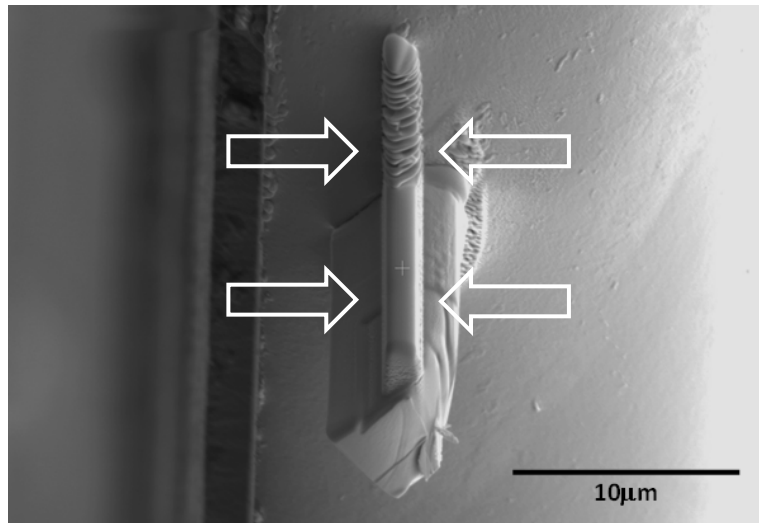


Fig. 11 Arrows pointing to platinum deposited on edge of target, just above the original surface

When using the planar method, the thinning process requires more care be taken to remove as little of the target's original surface as is practical. The first milling step was to remove most of the wedge with a relatively large current at 30 kV. This made the front and rear faces parallel and still protected the front from redeposition. Next, the platinum on the front surface was milled away using progressively smaller currents until the original surface was fully exposed with a smooth finish. Thinning from the reverse side then proceeded as usual until achieving an approximately 150-nm-thick lamella. Figure 12 shows the completed lamella as compared with the original ROI.

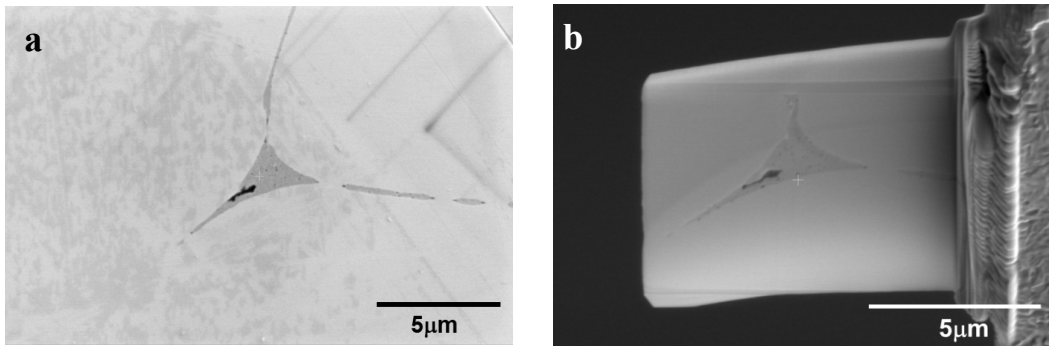


Fig. 12 SEM micrographs of (a) original ROI and (b) completed lamella

4. Results

The finished lamella was analyzed using a JEOL TEM, model 2100F; GATAN high angle annular dark field detector, model 806; and EDAX X-ray energy dispersive spectroscopy (XEDS) detector, model Elite T with silicon drift detector.

The TEM accelerating voltage used was 200 kV. Figure 13a shows a bright field scanning transmission electron microscopy (STEM) image along with an annular dark field STEM image (Fig. 13b). There is a noticeable void in the middle of the triple-point area, as noted previously, and possibly two smaller voids in the upper-grain boundary. Had the larger void extended deeper into the material, normal to the surface, a conventional cross-sectional lamella may have had difficulty holding together.

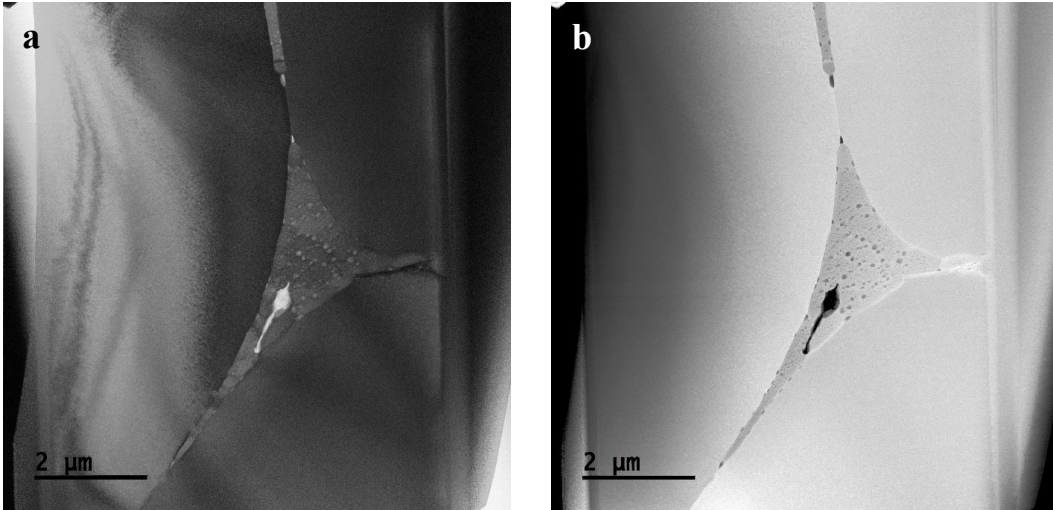


Fig. 13 STEM micrographs of completed lamella: (a) bright field and (b) annular dark field

Figure 14 shows detail of the annular dark field STEM image (Fig. 13b) containing the central triple-point area along with corresponding XEDS maps acquired for calcium, lanthanum, sulfur, aluminum, oxygen, and silicon. The intensity of light to dark in each map corresponds to the relative concentrations of each element. The presence and absence of these elements indicate that there are several possible phases represented other than the bulk CLS. The resolution of the maps is very good at better than 50 nm; however, even greater resolution could be obtained to conduct quantitative phase analysis.

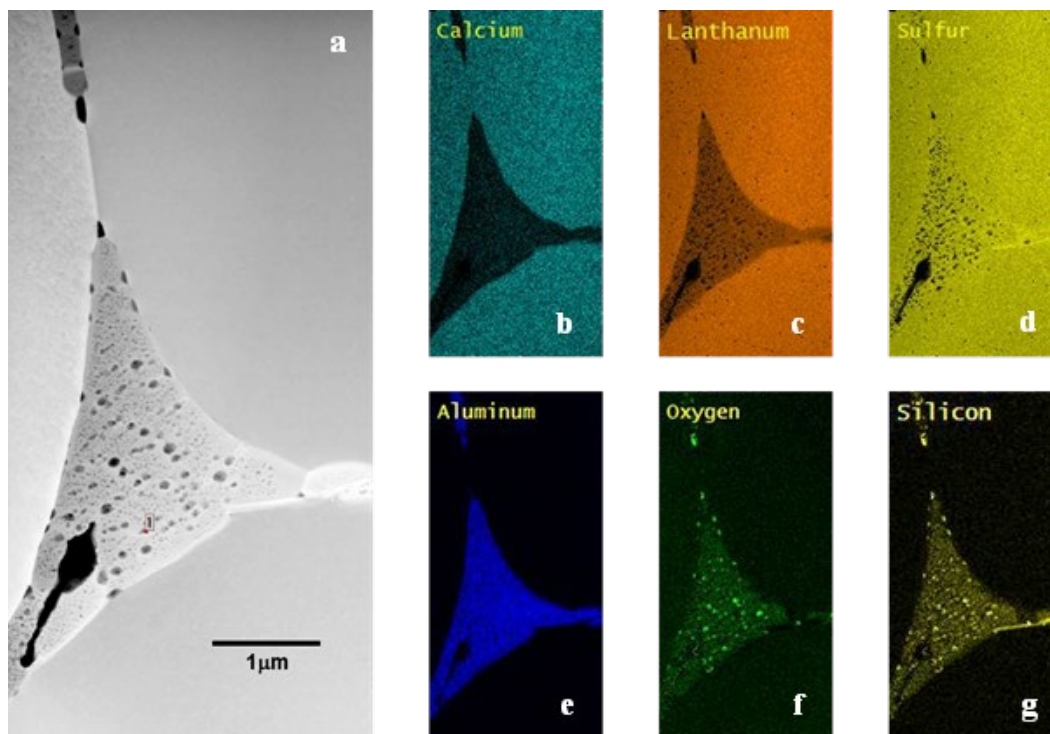


Fig. 14 (a) Detail of Fig. 13b; (b–f) XEDS maps of calcium, lanthanum, sulfur, aluminum, oxygen, and silicon, respectively

Comparing some aspects of the planar method with conventional cross section it is noted that the amount of platinum required for similar size lamella is five times more than for the conventional method and four times more for the areal coverage plus one more for the platinum added to the top of the wedge after mounting to the grid. For the planar method used here the added time to deposit the initial platinum protective layer was on the order of 20 min as compared to only 5 min for the conventional method, which is not unreasonable. The extra milling time required, however, was approximately 50% greater, which could be more inconvenient. Also, this method would be difficult to set up for automation whereas the cross-sectional method is usually much simpler to execute.

5. Conclusions

The best aspect of the planar method is ultimately “what you see is what you get,” otherwise known as WYSIWYG. It may not replace the necessity of looking below the surface of a material in cross section, but if you have already cut into and polished your bulk material, you may not have to waste time probing into the sample when what is important is visible right at the surface. For homogenous materials this may be superfluous, but if you are looking for a very specific feature, then this is a good option as it can save time, effort, and resources in the long run.

6. References

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List of Symbols, Abbreviations, and Acronyms

CC	cleaning cross section
CLS	calcium lanthanum sulfide
FIB	focused-ion beam
i^+	ion beam
RC	regular cross section
ROI	region of interest
SEM	scanning electron microscopy
STEM	scanning transmission electron microscopy
TEM	transmission electron microscopy (or microscope)
XEDS	X-ray energy dispersive spectroscopy
WYSIWYG	what you see is what you get
X-section	cross-sectional

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