



METHOD ARTICLE

REVISED Inhibition of vacuum sublimation artefacts for (Scanning) Transmission Electron Microscopy ((S)TEM) of sulphur samples *via* encapsulation

[version 2; peer review: 2 approved]

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Abstract

Lithium-sulfur battery is one of promising candidates for next-generation energy storage device due to the sulfur cathode material with low cost and nontoxicity, and super high theoretical energy density (nearly 2600Wh kg⁻¹) and specific energy (2567Wh kg⁻¹). Sulphur, however, poses a few interesting challenges before it can gain widespread utilisation. The biggest issue is known as the polysulphide shuttling effect which contributes to rapid capacity loss after cycling. Accurate characterisation of sulphur cathodic materials becomes critical to our understanding polysulphide shuttling effect in the quest of finding mitigating solutions. Electron microscopy is playing a crucial role in battery research in determining structure–property–function relations. However, sulphur undergoes sublimation at a point above the typical pressures found in the column of a transmission electron microscope (TEM) at room temperature. This makes the imaging and characterisation of any sort of nanostructured sulphur samples challenging, as the material will be modified or even disappear rapidly as soon as it is inserted into the TEM vacuum. As a result, materials characterised by such methods are prone to deviation from normal conditions to a great extent. To prevent this, a novel method of encapsulating sulphur particles between silicon nitride (SiN_x) membranes is demonstrated in this work.

Keywords

Transmission Electron Microscopy (TEM), sulfur, in-situ, sublimation, gas cell.

Open Peer Review

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1. **Andrew A. Stewart** , University of Limerick, Limerick, Ireland

2. **Toma Susi** , University of Vienna, Vienna, Austria

Any reports and responses or comments on the article can be found at the end of the article.



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Author roles: **Ronan O:** Formal Analysis, Investigation, Methodology, Validation, Writing – Original Draft Preparation; **Downing C:** Conceptualization, Writing – Review & Editing; **Nicolosi V:** Funding Acquisition, Project Administration, Resources, Supervision, Writing – Review & Editing

Competing interests: No competing interests were disclosed.

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REVISED Amendments from Version 1

The authors wish to thank the reviewers for their helpful comments, and have made changes accordingly. Text has been updated as per reviewer's suggestions.

Linked data have been updated to include original data files (viewable in Digital Micrograph (Gatan, Inc) and/or TEM Image Acquisition (FEI)) as well as .jpeg and .tif versions.

Reviewer comments on typing errors addressed.

Comments on specific reviewer questions added: Discussion added on EDX/EELS limitations with the method outlined. Discussion added on resolution/contrast limitations with the method outlined. Discussion added on imaging of air-sensitive samples using this method. Discussion added on sample preparation limitations with the method outlined. Discussion added on benefits of avoiding temperature changes in cryo-EM method.

Any further responses from the reviewers can be found at the end of the article

Introduction

With increased interest in sulphur as a candidate cathode material for high-performance lithium-ion batteries, the characterisation of sulphur to determine structure–property–function relations has become increasingly important in recent years^{1–3}. The characterisation of sulphur by electron microscopy poses a number of interesting challenges. The foremost issue is the high-vacuum environment required for electron microscopy; in the low pressure regime sulphur may undergo sublimation under high vacuum at room temperature, rendering it challenging to image in the transmission electron microscope (TEM)^{4–6}.

In order to prevent sublimation and give the microscopist enough time to complete imaging before the sample disappears entirely, a sample must be held in the solid region of its phase diagram^{5–7}. This can be achieved in a number of ways for samples that can undergo sublimation; by cryogenically cooling the sample to a temperature past the sublimation line, by increasing the pressure surrounding the sample to a level greater than its equilibrium vapor pressure (for sulphur; $\sim 10^{-7}$ Torr @ 20°C), or a combination of both^{4,5,7}.

At the typical TEM vacuum pressures (approx. 7×10^{-8} Torr), sulphur exists in the gas phase^{4,8}. To overcome this issue, cryogenic cooling to below 250 K or increasing pressure around the sample is required to keep sulphur in the solid orthorhombic phase^{4,5,7}.

Both methods have been previously demonstrated by Levin *et al.*⁵, showing stable imaging of sulphur using a Cryo-holder for the TEM and using an ambient pressure SEM. Building on this work, a novel method for imaging sulphur in the TEM using a SiN_x window gas cell holder is here proposed. With the introduction of commercially available gas cell holders, a sample can be held at a set pressure with any compatible gas connected to the sample holder. In this work, the sample was left open to air at atmospheric pressure for experimental simplicity. Given that the only necessary modification to the system would be the pressure, a set pressure of any non-reactive gas (such as N₂ or Ar) would also work if the

sample of interest is also sensitive to air. The concept of protectively encapsulating a sample to inhibit degradation in the microscope environment has been used in previous work by Kwon *et al.* to limit oxidation of copper nanowires, and a conceptually similar solution is demonstrated in this work⁹. However, the increased thickness added to the sample from the SiN_x windows used in this method will reduce the contrast of the image formed, and the field of view and viewable sample area is limited by the size of the viewing window relative to standard TEM grids⁹. Such compromises are considered acceptable for the sake of sample stability.

Methods

The sulphur particles were observed *via* field-emission (scanning) transmission electron microscopy ((S)TEM) (Titan, Thermo Fisher Scientific Inc.). The macrostructure and elemental composition of the sulphur particles were analysed using (S)TEM coupled with simultaneous energy dispersive X-ray spectroscopy (EDX; Ametek 30mm² EDX detector). A vacuum of 7×10^{-8} Torr and an acceleration voltage of 300 keV were employed for both TEM and STEM imaging during the measurements.

A sample of elemental sulphur was prepared by dispersing the as purchased sulphur powder (325 mesh, 99.5% purity, Alpha Aesar) in isopropanol (IPA) *via* ultrasonic bath sonication (Fisherbrand 11207 operated at 37kHz)¹⁰. The dispersion was deposited on a lacey carbon 400 mesh Cu grid (01896-F, TED PELLA Inc.) by dropcast for observation in vacuum, and allowed to dry in air. Sulphur particles were deposited onto Climate E-chips (P.T.GH.SS.2, DENS Solutions B.V. Delft, Netherlands, shown in Figure 4) in the same facile dropcast manner. Each chip, which consisted of an ~ 30 -nm-thick SiN_x membrane, was mounted in a Climate TEM holder (DENS Solutions B.V. Delft, Netherlands.). Here the SiN_x membrane acts as sample support while simultaneously isolating the sample from the vacuum environment.

The sample was loaded into the microscope, and a sulphur particle was chosen as the region of interest. The column valves were closed between each consecutive image to reduce any beam-induced artefacts for both standard Cu grid experiment and SiN_x window encapsulated experiment.

The encapsulated sample was left open to atmosphere at 1 bar during the experiment.

Results

In order to demonstrate sublimation of sulphur in normal TEM vacuum conditions a sulphur sample was prepared as described above and introduced into the microscope. As can be seen in the bright-field (BF) TEM images in Figure 1, the sulphur particle undergoes sublimation in the column vacuum over a period of time as little as 10 minutes. After this time, the residual material becomes vacuum-stable and does not sublime further. It is believed to be super-sublimated polymeric sulphur, according to literature sources^{11,12}. This is known to have a much lower vapour pressure than elemental sulphur¹¹ and is comparable to the experiments conducted by Levin *et al.*⁵. Examination of the residue by EDX STEM mapping at 300 keV confirms the residue as sulphur as evidenced by the S-K_α peak at 2.3keV (Figure 2).

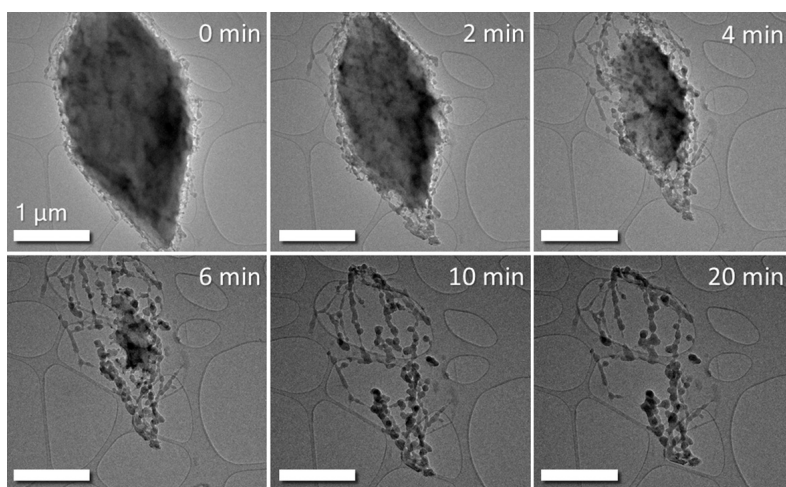


Figure 1. BF TEM time series images of sulphur particle subliming in TEM column vacuum of 7×10^{-8} Torr. The large particle in the region of interest is seen to dramatically shrink over time under vacuum exposure. Column valves were closed between images to prevent beam-induced effects altering the sublimation process. Scale bar is $1 \mu\text{m}$ in all above images.

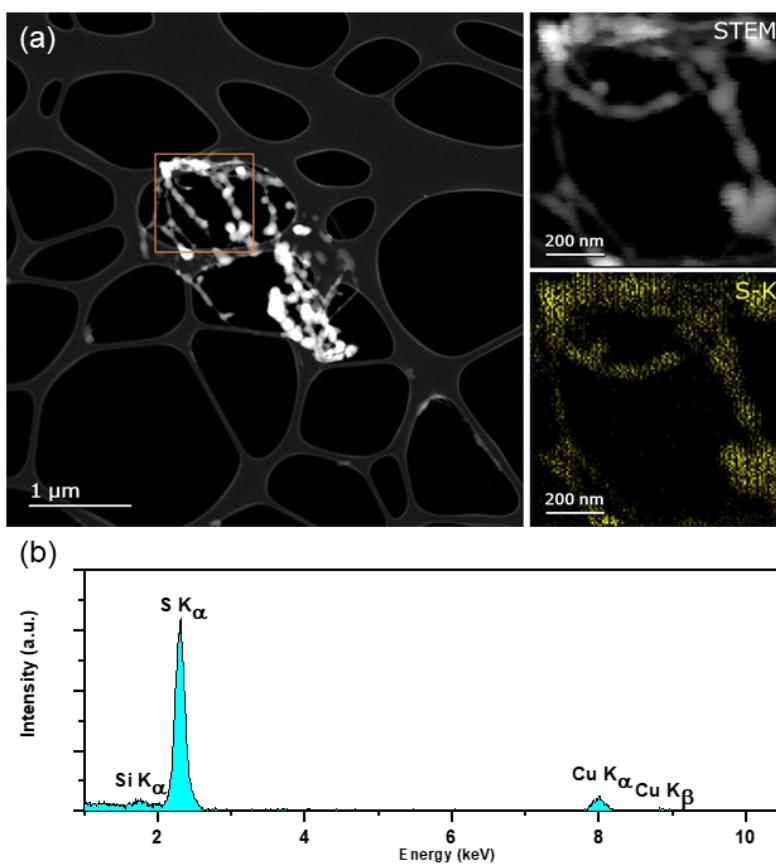


Figure 2. **a)** HAADF STEM of residual S post-sublimation acquired on FEI Titan @300keV. Inset shows STEM image of highlighted ROI and EDX maps of S corresponding to the EDX spectrum. **b)** Integrated spectrum of region of interest, confirming elements listed in a) are present. Maps confirm that the residue is indeed composed of sulphur. (Si peak due to an EDX spectral artefact; an internal fluorescence peak from the silicon window on the silicon drift detector likely. Cu peak is a background signal from the TEM grid.)

In order to investigate the potential of the commercial gas cell sample holder MEMS chips^{13,14} as a method to image the sulphur without sublimation the sample was deposited on SiN_x window DENS Solutions Climate MEMS chips (DENS Solutions B.V. Delft, Netherlands.), inserted into the column, and imaged over a period of 2 hours using the same experimental conditions as the previous experiment (300 keV TEM and STEM with a dwell time of 10ms/pixel and beam current of 0.5nA for EDX analysis). Unlike the previous experiment, the initial state of the particle is maintained in the column vacuum, and no change in particle morphology was observed over the observation period of 2 hours (as seen in Figure 3) in the high-angle annular dark field (HAADF) STEM image. The particle was kept at ambient pressure (1 bar) inside the Climate holder.

The EDX spectrum acquired after the 2 hours showing S-K_α and Si-K_α peaks (located at 2.3keV and 1.74keV respectively) in Figure 3c confirms that the particle is indeed sulphur supported on a SiN_x window. The suppression of the sublimation clearly demonstrates the viability of the use of windowed in-situ sample holders for imaging and characterisation of high vapour pressure materials in the TEM.

Being an electrically insulating material, sulphur is still very susceptible to radiolytic ionisation damage from the electron

beam, and careful consideration must be made to the beam current passing through the sample for both quantitative and qualitative spectroscopy¹⁵⁻¹⁸. For example, in order to acquire data for an EDX map of the sulphur particle, higher beam doses (using longer pixel dwell times or a higher beam current) must be used in order to generate enough counts for the detector. This has the effect of damaging the sample heavily due to sulphur's susceptibility to knock-on damage, and alternate methods must be employed to characterise the elemental composition of the sample with this method¹⁵⁻¹⁸. It is still possible to collect spectra to confirm elemental composition of the sample *via* EDX using a lower dose^{19,20}, as can be seen in Figure 3. In this regard, the cryo-EM method utilised by Levin *et al.*⁵ has its benefits over the SiN_x window approach that can be found in many in-situ holders. Cryogenic electron microscopy has the secondary benefit of slowing or even inhibiting beam induced artefacts and sublimation artefacts simultaneously, allowing for easier collection of mapping data²¹. However, cryo-EM may induce a phase change as a result of the change in temperature, depending on the sample being observed, and this method avoids this particular issue⁷.

Conclusions

The ability to image potentially vacuum sensitive materials for energy storage applications such as sulphur composites without

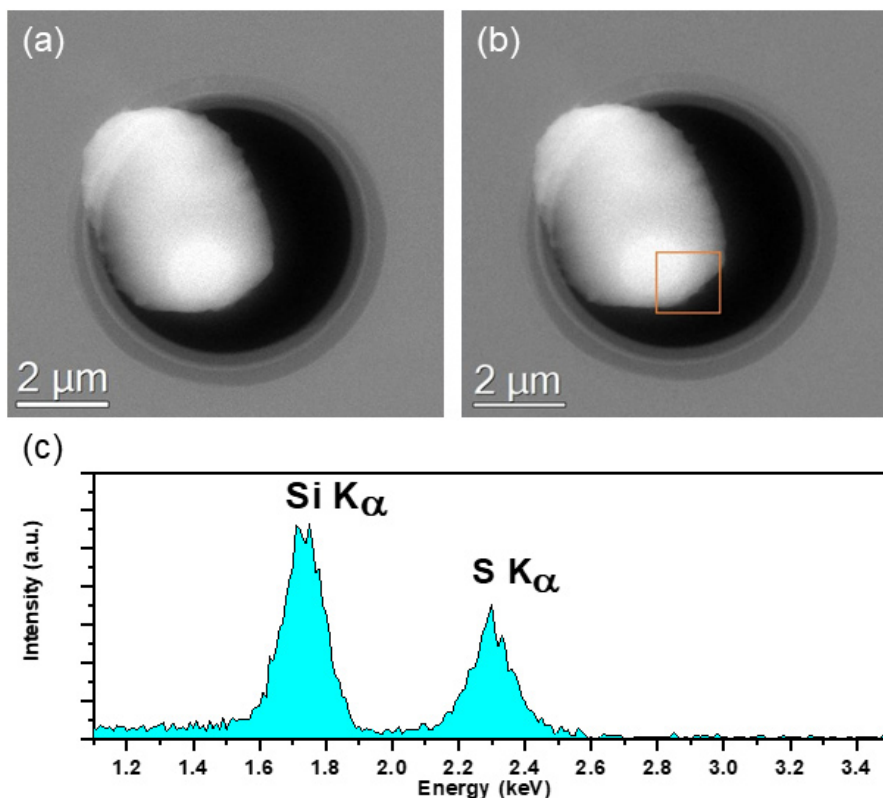


Figure 3. HAADF STEM images of a sulphur particle before (a) and after (b) a 2-hour exposure inside the TEM column vacuum. The particle is isolated from the column vacuum by the SiN_x window of the gas cell chips, thus preventing sublimation of the sample. (c) Integrated EDX Spectra of highlighted ROI of sulphur particle on SiN_x window of the gas cell chips after 2-hour exposure shown in (b) acquired @300 keV in FEI Titan equipped with an Ametek 30mm² EDX detector. S and Si peaks from the S particle and SiN_x window respectively clearly detected.

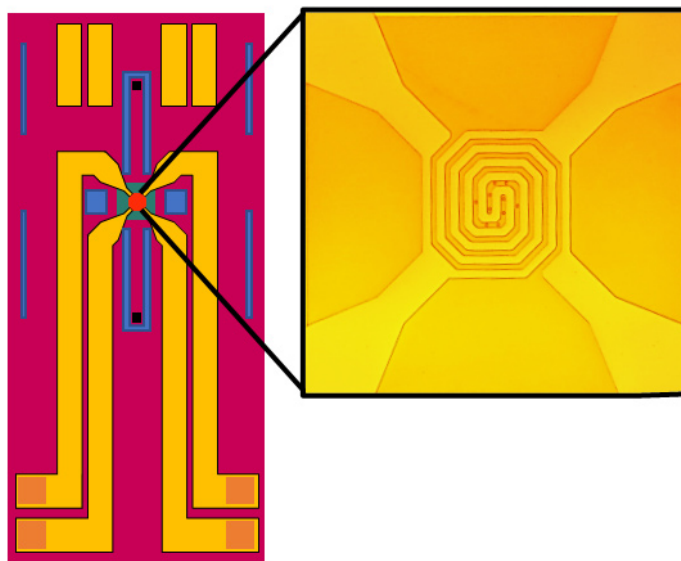


Figure 4. Schematic view of DENS Solutions Climate SiN_x window chips. SiN_x windows isolate the gas flow inside the holder from the vacuum of the TEM column. Heating coil visible over SiN_x windows.

sublimation artefacts creates new possibilities for their characterisation. To control sublimation in TEM analysis, an encapsulation method utilizing a commercial gas cell TEM sample holder is demonstrated. Where before, samples would sublime before imaging could take place, this novel encapsulation method allows for sample stability over the time frames necessary for both imaging, and spectroscopic analysis by EDX, or even EELS with ultra-thin SiN_x windows (providing the sample of interest does not contain peaks/edges that would be obfuscated by Si or N peaks/edges). The proposed method is suitable for the TEM analysis of other specimens that may undergo sublimation at TEM vacuum pressures, such as zinc, magnesium, or some polymers to name a few. Such techniques would have been impossible for such materials in the past and presents exciting new opportunities for battery materials researchers.

Data availability

Open Science Framework: Inhibition of vacuum sublimation artefacts for (Scanning) Transmission Electron Microscopy ((S)TEM) of sulphur samples via encapsulation, <https://doi.org/10.17605/OSF.IO/Z4MDV>²².

This project contains the following underlying data:

- Fig 1_1.dm3
- Fig 1_1.jpg
- Fig 1_1.tif
- Fig 1_2.dm3
- Fig 1_2.jpg
- Fig 1_2.tif
- Fig 1_3.dm3
- Fig 1_3.jpg
- Fig 1_3.tif
- Fig 1_4.dm3
- Fig 1_4.jpg
- Fig 1_4.tif
- Fig 1_5.dm3
- Fig 1_5.jpg
- Fig 1_5.tif
- Fig 1_6.dm3
- Fig 1_6.jpg
- Fig 1_6.tif
- Fig 2_1.tif
- Fig 2_2.tif
- Fig 2_3.tif
- Fig 2_Integrated EDX spectrum.csv
- Fig 3.tif
- Fig 3_1.tif
- Fig 3_Integrated EDX spectrum.csv
- Fig3_1.emi
- Fig3_1_1.ser
- Fig3_2.emi

- Fig3_2_1.ser
- S_window-EDX.txt
- S-EDX.txt
- Sublimated Sulphur Drift corrected EDX spectrum image.emi
- Sublimated Sulphur Drift corrected EDX spectrum image_1.ser
- Sulphur Window Drift corrected EDX spectrum image.emi
- Sulphur Window Drift corrected EDX spectrum

image_1.ser

Data are available under the terms of the [Creative Commons Zero “No rights reserved” data waiver](#) (CC-BY 4.0 Public domain dedication).

Acknowledgments

The authors acknowledge the Advanced Microscopy Laboratory for the provision of their facilities. Microscopy characterization and analysis has been performed at the CRANN Advanced Microscopy Laboratory (AML) (www.tcd.ie/crann/aml/).

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<http://www.doi.org/10.17605/OSF.IO/Z4MDV>

Open Peer Review

Current Peer Review Status:  

Version 2

Reviewer Report 17 February 2022

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Toma Susi 

Faculty of Physics, University of Vienna, Vienna, Austria

Thanks to the authors for the revisions – the new discussion could have been a little more extensive, but I understand this is not the main point here, and it's fine that the issues are only briefly mentioned.

Competing Interests: No competing interests were disclosed.

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Reviewer Report 16 February 2022

<https://doi.org/10.21956/openreseurope.15606.r28480>

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Andrew A. Stewart 

Department of Physics and the Bernal Institute, University of Limerick, Limerick, Ireland

The article can in my view be accepted, the points have all been addressed and all the data now being available fits into the open ethos of the journal.

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Transmission Electron Microscopy

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Version 1

Reviewer Report 21 January 2022

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Toma Susi 

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Although I am not an expert on battery materials, the authors convincingly argue for the importance of studying sulphur-based composites. For this purpose, (scanning) transmission electron microscopy and spectroscopy are indeed ideally suited, and the prevention of sulphur sublimation is an experimental challenge that the presented method appears to nicely address.

The article can in my view be already accepted, although I have some suggestions for improvement that the authors should consider.

Most importantly, the work would greatly benefit from a balanced discussion of the potential downsides that encapsulation within SiN_x membranes might bring. For example, how much more laborious is the sample preparation, and is the area that can be studied limited in an important way? How does the presence of the membrane materials affect imaging resolution or contrast? Does the presence of Si and N in the membranes interfere with EDX or EELS characterisation of some elements that might be important to identify in battery composites?

As regards to open data, I would urge the authors to not use a lossy compressed file format such as .jpg for any data items. Instead, uncompressed .tif or .png formats should be used, or even better yet, the original data files should also be included without any overlaid graphics.

Finally, I have minor suggestions for copyediting corrections:

- Use of the acronym in the title seems unnecessary.
- The phrase "super high" in the abstract seems a little unscientific, but perhaps this is common usage in the field.
- Often a space is missing between numerical quantities and units.
- There is some apparent repetition in the text, e.g. the Titan instrument provider is repeated,

and the closing of column valves is mentioned twice on p. 3.

- A few typos are present, e.g. in "300keV TEM and STEM **will** a dwell time" on p.3, "confirming elements **listen** in a)" in the caption of Figure 2 on p.4, and "Being an electrical insulating material" (as opposed to "electrical insulator" or "electrically insulating material") on p.6.

Is the rationale for developing the new method (or application) clearly explained?

Yes

Is the description of the method technically sound?

Yes

Are sufficient details provided to allow replication of the method development and its use by others?

Yes

If any results are presented, are all the source data underlying the results available to ensure full reproducibility?

Partly

Are the conclusions about the method and its performance adequately supported by the findings presented in the article?

Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Transmission electron microscopy

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Reviewer Report 18 January 2022

<https://doi.org/10.21956/openreseurope.15517.r28271>

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This article addresses the important problem of materials with high vapour or sublimation pressures within the low pressure environment of the Transmission Electron Microscope (TEM),

suggesting the use of gas cells to encapsulate the material and observe it at atmospheric pressure to circumvent the high vacuum problems caused by the instrumentation. Sulphur is used for the purposes of demonstrating the effectiveness of the gas cell.

The comparison of the sulphur particles is made with a particles on a carbon substrate and compares the results with the closed cell made of silicon nitride.

The figures 1&2 clearly show the disintegration of the particle in the presence of the high vacuum and the electron beam, with figure 3 showing the stability of the particle in the presence of the gas phase at atmospheric pressure. What is omitted is the difficulties of producing such samples where the placement of the sample on the ultra thin windows of the SiN_x chips is either luck or careful placement with a FIB, compared to a carbon grid where almost any region of the grid is reasonable to collect data from.

A point not addressed in the text is using the high pressure room temperature approach can also avoid any phase changes as a result of temperature changes. Just using atmosphere can be problematic for some samples which are more sensitive to water vapour and its radicals caused by the electron beam.

While the data is provided the nature of the data output from some of the vendors software will be missing significant metadata required to fully interperate the images without further information, this is a general problem of TEM experiments currently. The authors could indicate which software can be used to read the files to obtain the metadata such as Velox or Hyperspy, would be a way of partially addressing this issue.

Is the rationale for developing the new method (or application) clearly explained?

Yes

Is the description of the method technically sound?

Yes

Are sufficient details provided to allow replication of the method development and its use by others?

Yes

If any results are presented, are all the source data underlying the results available to ensure full reproducibility?

Yes

Are the conclusions about the method and its performance adequately supported by the findings presented in the article?

Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Transmission Electron Microscopy

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.