Korea Advanced Institute of Science and Technology

**291 Daehak-ro, Yuseong-gu Daejeon 34141 Korea | Tel: +82-42-350-3322 | Email: jongmin.yuk@kaist.ac.kr**

September 05, 2018

To: Editor Prof. Xiaoyan Cao

**Editor of *Journal of Visualized Experiments***

Re: **Manuscript ID: JOVE58676**

Dear Prof. Xiaoyan Cao,

We received the reviewer’s comments on our manuscript entitled **"Preparation of graphene liquid cells for lithium ion battery material observation"** submitted to ***Journal of Visualized Experiments.***

We would like to first thank the editor and reviewer for his/her prompt yet careful and thorough report. We greatly appreciate his/her comments and recommendations and we have made all efforts to address them properly in blue in provided response letter. The changes in the revised manuscript according to the reviewers’ directions are clearly highlighted in red. We believe we have made the required revisions recommended by the reviewers and look forward to acceptance of the revised manuscript.

Sincerely Yours,

Jong Min Yuk, Assistant professor

Tel) 82-42-350-3322

Fax) 82-42-350-3310

Email) jongmin.yuk@kaist.ac.kr

W1-1 #3304

Dept. of Materials Science and Engineering

Korea Advance Institute of Science and Technology (KAIST)

291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea

**<Response letter to the editor’s comments>**

**Editor’s remark: Your manuscript, JoVE58676R1 "Preparation of graphene liquid cells for lithium ion battery material observation," has been editorially reviewed and the following comments need to be addressed. Please track the changes to identify all of the manuscript edits. After revising the submission, please also upload a separate document that addresses each of the editorial comments individually with the revised manuscript:**

**(Response)** We appreciate the editor’s kind comments in the manuscript, which strengthen our manuscript. Especially, the combination of some of the steps improves the overall quality of the paper, for which we respect the most. For all the editorial comments individually, we have made the changes in the revised manuscript. For the comments that did not have any changes, they were in agreement what the reviewer had suggested (such as some of the comments regarding the combination of two sub-steps into one step).

**1.** **After you have made all the recommended changes to your protocol (see specific comments below), please highlight 2.75 pages or less of the Protocol (including headings and spacing) that identifies the essential steps of the protocol for the video, i.e., the steps that should be visualized to tell the most cohesive story of the Protocol.**

**(Response)** We appreciate the editor’s kind comments, which will strengthen the manuscript.

**2. Please include all relevant details that are required to perform the step in the highlighting. For example: If step 2.5 is highlighted for filming and the details of how to perform the step are given in steps 2.5.1 and 2.5.2, then the sub-steps where the details are provided must be highlighted.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, we also highlighted the sub-steps where the details are provided. For instance, we have additionally highlighted some parts of the section 2.2.8, where it gives more details into how the cell is assembled. We have highlighted both the steps and the sub-steps that add more detail to the experimental steps.

**3. 1.2.9. Please specify which software.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, we revised our manuscript and added specific program name in Jove\_Materials excel file.

(Revised manuscript, protocol 1.2.9)

1.2.9. Roll the roller (100 rpm) and initiate the electrospinning program software.

**4. 1.2.10 How? Using a voltage bias?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, we need to indicate how we modulated the applied voltage. Based on this point, we have revised the manuscript.

(Revised manuscript, protocol 1.2.10)

1.2.10. Lastly, modulate the applied voltage by using a voltage bias (**Figure 1a**) between 10-25 kV to allow the electrojetting of the Taylor cones to operate the electrospinning process.

**5. 1.3.2 I have combined shorter steps. Please check for accuracy here.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. The currently combined steps are accurate.

**6. 1.3.3 Please check the accuracy here as I rephrased the sentences. Please also note that I highlighted this step for filming because it shows the final product of this step and including it tells a complete story of fabrication of SnO2 nanotubes.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, the rephrased sentence precisely describes what we had wanted to explain. Moreover, this tells a complete theory of fabrication of SnO2 nanotubes.

**7. 2.1 What slurry? Electrode slurry?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, it is important to indicate what kind of slurry it is. It is the electrode slurry and we added this in our manuscript.

(Revised manuscript, protocol 2.1)

The electrode slurry is composed of 10 wt% binder, 10 wt% Super P carbon, and 80 wt% of active materials (in this case, SnO2 nanotubes).

**8. 2.1 Carbon in what form, carbon black?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, it is important to indicate what carbon it is. The form of carbon is Super P carbon and we added this information in our manuscript.

(Revised manuscript, protocol 2.1)

The electrode slurry is composed of 10 wt% binder, 10 wt% Super P carbon, and 80 wt% of active materials (in this case, SnO2 nanotubes).

**9. 2.1.1 What glass substrate?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, it is important to indicate what glass substrate it is. It is the rectangular glass substrate with dimension of 25 cm x 15 cm x 0.5 cm. We have revised the manuscript:

(Revised manuscript, protocol 2.1.1)

2.1.1. Cut the copper (Cu) foil into 10 cm width × 30 cm length and fix it on a rectangular glass substrate (25cm x 15 cm x 0.5 cm) by using ethanol.

**10. 2.1.5 Diameter?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. 60 μm is not diameter, but thickness of the slurry. We specified this and revised our manuscript.

(Revised manuscript, protocol 2.1.5)

2.1.5. When the slurry is well prepared, place the slurry on the top side of the Cu foil on the glass substrate, and cast it evenly using a casting roller.

Note: Usually thickness of slurry is 60 μm but can be higher or lower).

**11. 2.2.1 I have combined shorter steps. Please check for accuracy.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, the combination of shorter steps in 2.2.1 needs to be checked for accuracy. The currently combined shorter steps are accurate.

**12. 2.2.2 I rephrased this sentence. Please check whether the original meaning has been retained.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, rephrased sentence in 2.2.2 needs to be checked for retaining the original meaning. The rephrased sentence has retained the original meaning.

**13. 2.2.9 At what conditions? Temperature?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, it is important to indicate what condition it is. We have indicated the condition, as shown below:

(Revised manuscript, protocol 2.2.9)

2.2.9. Measure the open circuit voltage (OCV) by a digital multimeter and age the battery cell at room temperature for 1-2 days.

**14. 2.3.3 Please specify the specific current values applied.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor commented, it is important to specify the specific current values. We have indicated the current values, as shown below:

(Revised manuscript, protocol 2.3.3)

2.3.3. Insert the electrochemical battery cells in the battery cell tester. Apply the current (corresponding to 0.05 A g-1 for formation cycle and various current densities in the range of 0.1 A g-1 to 10.0 A g-1 for cycle tests and rate capabilities) for each battery cell using the battery cell tester program.

**15. 3.1.1 This has been rephrased. Please check for accuracy.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. The meaning of rephrased sentence is correct and we left this as it is.

**16. 3.1.2 Do you mean Cu foil pieces, as 3.1.1 cuts the foil into pieces?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. The Cu foil is 10 cm x 3 cm that is cut in 3.1.1. We rephrased this sentence in order not to make any confusion.

(Revised manuscript, protocol 3.1.2)

3.1.2. Rinse Cu foil from 3.1.1 with isopropyl alcohol (IPA) to remove dust or contaminants and treat it with 100 mL of 20 wt% phosphoric acid (H3PO4) for 20 min to remove native oxide on the surface in a glass Petri dish. Then place Cu foil in DI water for another 10 min to fully rinse remaining H3PO4.

**17. 3.1.2 Please specify the concentration and volume of H3PO4 used.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. The concentration of H3PO4 is 20 weight% and volume is 100 mL. We added this information in our manuscript.

(Revised manuscript, protocol 3.1.3)

3.1.2. Rinse Cu foil from 3.1.1 with isopropyl alcohol (IPA) to remove dust or contaminants and treat it with 100 mL of 20 wt% phosphoric acid (H3PO4) for 20 min to remove native oxide on the surface in a glass Petri dish. Then place Cu foil in DI water for another 10 min to fully rinse remaining H3PO4.

**18. 3.1.5 Do you mean hold at 1000 °C for 40 min or elevate the temperature to 1000 °C in 40 min?**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. The temperature is elevated from room temperature (25 °C) to 1000 °C in 40 minutes. We specified this in our manuscript.

(Revised manuscript, protocol 3.1.5)

3.1.5. Elevate the temperature from room temperature (25 °C) to 1000 °C for 40 min with 10 sccm of H2 gas flow. Maintain the temperature of chamber for another 40 min to anneal the Cu foil.

**19. 3.2.1 Is etching performed in a specific instrument? Please specify.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. The plasma etching is conducted with a specific plasma cleaner instrument (Femtoscience VITA). We added this information in our manuscript and Jove Materials excel file.

(Revised manuscript, protocol 3.2.1)

3.2.1. To remove graphene on the backside of Cu foil, conduct plasma etching using plasma cleaner by following settings: flow of Ar (100 sccm), time (60 s), power (30 W), and base pressure (5.0 x 10-2 Torr).

**20. 3.2.2 Please specify in which step graphene is synthesized.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As-synthesized graphene is the one synthesized in protocol 3.1. We specified this in our manuscript.

(Revised manuscript, protocol 3.2.2)

3.2.2. Cut Cu foil with graphene which was synthesized in 3.1, by 3 mm x 3 mm with scissors. Place Cu foil pieces between two slide glasses and press to make them flat.

Note: 4 Cu foil pieces are placed together between two slide glasses.

**21. 3.2.2 How many pieces are placed between two slide glasses.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. Usually, 4 Cu foil pieces are placed between two slide glasses. This will be also included in movie, so readers will be able to understand this process clearly. We added this information in our manuscript.

(Revised manuscript, protocol 3.2.2)

3.2.2. Cut Cu foil with graphene which was synthesized in 3.1, by 3 mm x 3 mm with scissors. Place Cu foil pieces between two slide glasses and press to make them flat.

Note: 4 Cu foil pieces are placed together between two slide glasses.

**22. 3.2.4 What is suctioned, IPA?**

We appreciate the editor’s comments, which will strengthen the manuscript. IPA is suctioned at this process because the only liquid exist here is IPA. We revised this step so that readers are not confused.

(Revised manuscript, protocol 3.2.4)

3.2.4. Suction IPA with a micropipet tip that is connected to a rotary pump. After suctioning, dry Au grid/Cu foil at 50 °C for 5 min.

**23. 3.2.6 This is unclear. Do you mean contaminants from etchant?**

We appreciate the editor’s comments, which will strengthen the manuscript. This phrase originally means contaminants from etchant, as the editor pointed out. We revised our manuscript.

(Revised manuscript, protocol 3.2.6)

3.2.6. Scoop Au grids Pt loop and move it to a glass Petri dish filled with DI water at 50 °C, in order to fully remove remaining contaminants from etchant16.

**24. 3.2.7 Please specify conditions. In an oven? At what temperature?**

We appreciate the editor’s comments, which will strengthen the manuscript. The Au grids with graphene are not dried at special condition. The drying condition is at room temperature and atmospheric pressure.

(Revised manuscript, protocol 3.2.7)

3.2.7. Scoop Au grids from DI water and dry them for 6 h at room temperature and atmospheric pressure.

**25. 3.3.1 Please check for accuracy.**

**(Response)** We appreciate the editor’s kind comments, which will strengthen the manuscript. The revised sentence of the editor is accurate.

**26. 3.3.4 Please check for accuracy.**

**(Response)** We appreciate the editor’s kind comments, which will strengthen the manuscript. As shown in Figure 2d, the other grid is held with a tweezer and placed on the top of the bottom one. We slightly changed this step with editor’s revised one.

(Revised manuscript, protocol 3.2.7)

3.3.4. Hold another graphene transferred grid with a tweezer and place it on the top of the bottom grid.

Note: This procedure must be done quickly before electrolyte is dried (**Figure 2d**).

**27. 3.3.4 Figure 3c does not show grid. Please reference the correct figure. Figure 2d?**

**(Response)** We appreciate the editor’s kind comments, which will strengthen the manuscript. As the editor pointed out, Figure 2d is correct. We revised our manuscript based on the editor’s comment.

(Revised manuscript, protocol 3.2.7)

3.3.4. Hold another graphene transferred grid with a tweezer and place it on the top of the bottom grid.

Note: This procedure must be done quickly before electrolyte is dried (**Figure 2d**).

**28. 4.2.1, 4.2.2 Please provide more specific details about how these steps are done.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor pointed out, specific details are needed in these steps.

(Revised manuscript, protocol 4.2.1)

4.2.1. Find the region where SnO2 nanotube is encapsulated with liquid electrolyte.

Note: To find out whether liquid exist around SnO2 nanotube, irradiate electron beam for few seconds. If some movement of liquid or decomposition of electrolyte is observed, it is highly likely that the area is encapsulated with liquid.

(Revised manuscript, protocol 4.2.2)

4.2.2. Do alignment for TEM and set electron beam dosage to initiate the reaction by adjusting brightness knob.

Note: Suitable alignment for TEM includes user alignment such as Z height alignment, gun tilt/shift, beam tilt/shift, aperture alignment and stigmation alignment. These procedures are better done in other area (right next to the region found in 4.2.1) in order not to give any damage to SnO2 nanotube and liquid electrolyte. The electron beam dose rate for initiating the lithiation is usually ~ 103 , but it may differ with every TEM instrument.

**29. 4.2.2 Every TEM instrument or TEM measurement? Please specify.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor pointed out, the protocol should be specified. The original meaning of this phrase is every TEM instrument.

(Revised manuscript, protocol 4.2.2)

4.2.2. Do alignment for TEM and set electron beam dosage to initiate the reaction by adjusting brightness knob.

Note: Suitable alignment for TEM includes user alignment such as Z height alignment, gun tilt/shift, beam tilt/shift, aperture alignment and stigmation alignment. These procedures are better done in other area (right next to the region found in 4.2.1) in order not to give any damage to SnO2 nanotube and liquid electrolyte. The electron beam dose rate for initiating the lithiation is usually ~ 103 , but it may differ with every TEM instrument.

**30. Please define SAED.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor pointed out, SAED must be defined. We revised our manuscript.

(Revised manuscript, page 9, line 6)

TEM image and corresponding selected area electron diffraction (SAED) pattern of transferred graphene are shown in **Figures 5c** and **5d**.

**31. Please describe Movie S1 in the figure legend.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor pointed out, SAED must be defined. We revised the figure legend part in our manuscript.

(Revised manuscript, page 9, line 40-41)

**Movie S1**. Movie showing litihation of GLC. The surface of SnO2 nanotube is visualized inside liquid electrolyte.

**31. Figure 5. Please add one sentence to describe this figure in general.**

**(Response)** We appreciate the editor’s comments, which will strengthen the manuscript. As the editor pointed out, one sentence to describe Figure 5 is needed. We added this information in our manuscript.

(Revised manuscript, page 9, line 34)

**Figure 5.** Characterization of synthesized graphene. (a) Raman spectrum, (b) SEM image, (c) TEM image, and (d) SAED pattern of the monolayer graphene.

**32. Figure 6. Please spell out SEI.**

**(Response)** We appreciate the editor’s kind comments, which will strengthen the manuscript. As the editor pointed out, SEI must be defined. We defined solid electrolyte interphase (SEI) in revised our manuscript.

(Revised manuscript, page 8, line 38)

The irreversible formation of Li2O during conversion reaction of SnO2 along with unstable formation of solid electrolyte interphase (SEI) layer resulted in poor reversible reaction with Li in the formation cycle.