Enhancement of microwave absorption bandwidth of MXene nanocomposites through macroscopic design

Pritom J. Bora, T. R. Suresh Kumar and Daniel Q. Tan

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Revised submission: 24 June 2020
Final acceptance: 10 July 2020

Note: Reports are unedited and appear as submitted by the referee. The review history appears in chronological order.

Review History
RSOS-200456.R0 (Original submission)

Review form: Reviewer 1

Is the manuscript scientifically sound in its present form?
Yes

Are the interpretations and conclusions justified by the results?
No

Is the language acceptable?
Yes

Do you have any ethical concerns with this paper?
No

Have you any concerns about statistical analyses in this paper?
No

Recommendation?
Accept with minor revision (please list in comments)
Comments to the Author(s)
This manuscript proposed a macroscopic engineering method to improve microwave absorption based on MXene material. This manuscript is inspired by previous graphene devices design, yet interesting for MXene EMA research. The following are some comments about this manuscript.

1. In figure 1, more material characterizations are required to support the claimed composite materials of TiO2-Ti3C2Tx, and Fe3O4@TiO2-Ti3C2Tx. For example, XPS mapping is necessary to be used to present the element distribution. The SEM image can not show the presence of Fe.

2. The references of MXene based functional devices need to be updated, see a recent review "X. Jiang, A.V. Kuklin, A. Baev, et al. Physics Reports 848 (2020) 1-58”

3. The experimental pictures of the MXene coated pyramid matrix are missing and should be presented in the manuscript. How is the quality of the coating? The performance of the devices highly depends on the fabrication process and outcome.

4. The authors claimed ultra-light MXene devices, which might be inaccurate and lack of experimental evidence. The Ti3C2Tx multi-layer devices are not necessarily lighter than graphene-based devices.

Review form: Reviewer 2

Is the manuscript scientifically sound in its present form?
Yes

Are the interpretations and conclusions justified by the results?
Yes

Is the language acceptable?
Yes

Do you have any ethical concerns with this paper?
No

Have you any concerns about statistical analyses in this paper?
No

Recommendation?
Accept with minor revision (please list in comments)

Comments to the Author(s)
As a novel type of 2D materials, Mexene possesses important application prospect in field of microwave absorption materials. To enhance the microwave absorption width of Mexene-hybrid matrix materials, two types of Mexene hybrid materials were fabricated for the first time by macroscopic design approach through simulation method. The results are beneficial for the successful production of Mexene hybrid-matrix materials with wide microwave absorption bandwidth. I thus recommend its publication in this journal after minor revision as followings:

1) In the as-obtained Fe3O4@TiO2-Ti3C2Tx, please provide the particular size of Fe3O4 nanoparticles, distribution, and proportion?
2) What is the interaction type between the Fe3O4 nanoparticles and Mexene matrix?
We hope you are keeping well at this difficult and unusual time. We continue to value your support of the journal in these challenging circumstances. If Royal Society Open Science can assist you at all, please don't hesitate to let us know at the email address below.

Dear Dr Tan:

Title: Enhancement of microwave absorption bandwidth of MXene nanocomposites through macroscopic design  
Manuscript ID: RSOS-200456

Thank you for submitting the above manuscript to Royal Society Open Science. On behalf of the Editors and the Royal Society of Chemistry, I am pleased to inform you that your manuscript will be accepted for publication in Royal Society Open Science subject to minor revision in accordance with the referee suggestions. Please find the reviewers' comments at the end of this email.

The reviewers and handling editors have recommended publication, but also suggest some minor revisions to your manuscript. Therefore, I invite you to respond to the comments and revise your manuscript.

Because the schedule for publication is very tight, it is a condition of publication that you submit the revised version of your manuscript before 21-Jun-2020. Please note that the revision deadline will expire at 00.00am on this date. If you do not think you will be able to meet this date please let me know immediately.

To revise your manuscript, log into https://mc.manuscriptcentral.com/rsos and enter your Author Centre, where you will find your manuscript title listed under "Manuscripts with Decisions". Under "Actions," click on "Create a Revision." You will be unable to make your revisions on the originally submitted version of the manuscript. Instead, revise your manuscript and upload a new version through your Author Centre.

When submitting your revised manuscript, you will be able to respond to the comments made by the referees and upload a file "Response to Referees" in "Section 6 - File Upload". You can use this to document any changes you make to the original manuscript. In order to expedite the processing of the revised manuscript, please be as specific as possible in your response to the referees.

When uploading your revised files please make sure that you have:

1) A text file of the manuscript (tex, txt, rtf, docx or doc), references, tables (including captions) and figure captions. Do not upload a PDF as your "Main Document".
2) A separate electronic file of each figure (EPS or print-quality PDF preferred (either format should be produced directly from original creation package), or original software format)
3) Included a 100 word media summary of your paper when requested at submission. Please ensure you have entered correct contact details (email, institution and telephone) in your user account
4) Included the raw data to support the claims made in your paper. You can either include your data as electronic supplementary material or upload to a repository and include the relevant doi within your manuscript
5) All supplementary materials accompanying an accepted article will be treated as in their final form. Note that the Royal Society will neither edit nor typeset supplementary material and it will be hosted as provided. Please ensure that the supplementary material includes the paper details where possible (authors, article title, journal name).
Supplementary files will be published alongside the paper on the journal website and posted on the online figshare repository (https://figshare.com). The heading and legend provided for each supplementary file during the submission process will be used to create the figshare page, so please ensure these are accurate and informative so that your files can be found in searches. Files on figshare will be made available approximately one week before the accompanying article so that the supplementary material can be attributed a unique DOI.

Once again, thank you for submitting your manuscript to Royal Society Open Science. The chemistry content of Royal Society Open Science is published in collaboration with the Royal Society of Chemistry. I look forward to receiving your revision. If you have any questions at all, please do not hesitate to get in touch.

Kind regards,
Dr Laura Smith
Publishing Editor, Journals

Royal Society of Chemistry
Thomas Graham House
Science Park, Milton Road
Cambridge, CB4 0WF
Royal Society Open Science - Chemistry Editorial Office

On behalf of the Subject Editor Professor Anthony Stace and the Associate Editor Dr Chaohua Cui.

********************************************************************************

RSC Associate Editor:
Comments to the Author:
(There are no comments.)

RSC Subject Editor:
Comments to the Author:
(There are no comments.)

********************************************************************************

Reviewer comments to Author:
Reviewer: 1

Comments to the Author(s)
This manuscript proposed a macroscopic engineering method to improve microwave absorption based on MXene material. This manuscript is inspired by previous graphene devices design, yet interesting for MXene EMA research. The following are some comments about this manuscript.

1. In figure 1, more material characterizations are required to support the claimed composite materials of TiO2-Ti3C2Tx, and Fe3O4@TiO2-Ti3C2Tx. For example, XPS mapping is necessary to be used to present the element distribution. The SEM image can not show the presence of Fe.

2. The references of MXene based functional devices need to be updated, see a recent review "X. Jiang, A.V. Kuklin, A. Baev, et al. Physics Reports 848 (2020) 1-58”

3. The experimental pictures of the MXene coated pyramid matrix are missing and should be presented in the manuscript. How is the quality of the coating? The performance of the devices highly depends on the fabrication process and outcome.
4. The authors claimed ultra-light MXene devices, which might be inaccurate and lack of experimental evidence. The Ti3C2Tx multi-layer devices are not necessarily lighter than graphene-based devices.

Reviewer: 2

Comments to the Author(s)
As a novel type of 2D materials, Mexene possesses important application prospect in field of microwave absorption materials. To enhance the microwave absorption width of Mexene-hybrid matrix materials, two types of Mexene hybrid materials were fabricated for the first time by macroscopic design approach through simulation method. The results are beneficial for the successful production of Mexene hybrid-matrix materials with wide microwave absorption bandwidth. I thus recommend its publication in this journal after minor revision as followings: 1) In the as-obtained Fe3O4@TiO2-Ti3C2Tx, please provide the particular size of Fe3O4 nanoparticles, distribution, and proportion? 2) What is the interaction type between the Fe3O4 nanoparticles and Mexene matrix?

Author's Response to Decision Letter for (RSOS-200456.R0)

See Appendix A.

RSOS-200456.R1 (Revision)

Review form: Reviewer 1

Is the manuscript scientifically sound in its present form?
Yes

Are the interpretations and conclusions justified by the results?
Yes

Is the language acceptable?
Yes

Do you have any ethical concerns with this paper?
No

Have you any concerns about statistical analyses in this paper?
No

Recommendation?
Accept as is

Comments to the Author(s)
Acceptable now.
Decision letter (RSOS-200456.R1)

We hope you are keeping well at this difficult and unusual time. We continue to value your support of the journal in these challenging circumstances. If Royal Society Open Science can assist you at all, please don’t hesitate to let us know at the email address below.

Dear Dr Tan:

Title: Enhancement of microwave absorption bandwidth of MXene nanocomposites through macroscopic design
Manuscript ID: RSOS-200456.R1

It is a pleasure to accept your manuscript in its current form for publication in Royal Society Open Science. The chemistry content of Royal Society Open Science is published in collaboration with the Royal Society of Chemistry.

The comments of the reviewer(s) who reviewed your manuscript are included at the end of this email.

Thank you for your fine contribution. On behalf of the Editors of Royal Society Open Science and the Royal Society of Chemistry, I look forward to your continued contributions to the Journal.

Yours sincerely,
Dr Laura Smith
Publishing Editor, Journals

Royal Society of Chemistry
Thomas Graham House
Science Park, Milton Road
Cambridge, CB4 0WF
Royal Society Open Science - Chemistry Editorial Office

On behalf of the Subject Editor Professor Anthony Stace and the Associate Editor Dr Chaohua Cui.

*******

RSC Associate Editor:
Comments to the Author:
(There are no comments.)

RSC Subject Editor:
Comments to the Author:
(There are no comments.)

*******

Reviewer(s)' Comments to Author:
Reviewer: 1

Comments to the Author(s)
Acceptable now.
Appendix A

Responses to the comments of the reviewers (RSOS-200456)

The authors thank the reviewers for their useful comments and suggestions. We have thoroughly addressed the comments of the reviewers and made suitable modifications in the revised manuscript. The comments of the reviewers are in italics and our responses are in normal font. The modifications to the revised manuscript are highlighted in yellow.

Reviewer: 1

Comments to the Author(s):

This manuscript proposed a macroscopic engineering method to improve microwave absorption based on MXene material. This manuscript is inspired by previous graphene devices design, yet interesting for MXene EMA research.

We thank the reviewer for appreciating this work and recommending its publication.

The following are some comments about this manuscript. In figure 1, more material characterizations are required to support the claimed composite materials of TiO2-Ti3C2Tx, and Fe3O4@TiO2-Ti3C2Tx. For example, XPS mapping is necessary to be used to present the element distribution. The SEM image can not show the presence of Fe.

Response: Both the nanocomposites viz., TiO2-Ti3C2Tx, and Fe3O4@TiO2-Ti3C2Tx was synthesized according to the standard procedure adopted in the references [17] and [18], where all the characterizations were provided. In this work, the same synthetic procedure, the same conditions, and parameters of synthesis were used (the chemicals were also procured from the same suppliers). The objective of this work is more towards application point of view. However, as suggested by the reviewer, we added more elemental distribution information by providing EDX data in the revised manuscript (Supporting information). In addition, in the case of Fe3O4@TiO2-Ti3C2Tx, the permeability data is predominant, which thus indicates the presence of Fe through magnetic Fe3O4 in TiO2-Ti3C2Tx. Following modifications have been made in the revised manuscript,

The energy dispersive X-ray (EDX) confirms the presence of Fe (Figure S1, supporting information).
Figure S1. EDX spectra of Fe$_3$O$_4$@TiO$_2$-Ti$_3$C$_2$Tx MXene.

#2. The references of MXene based functional devices need to be updated, see a recent review "X. Jiang, A.V. Kučkin, A. Baev, et al. Physics Reports 848 (2020) 1–58”

Response: We thank the reviewer for this valuable suggestion. We have cited this paper in the revised manuscript.

Following modifications have been made in the revised manuscript:

References:
[14] Jiang X, Kučkin A V., Baev A, Ge Y, Ågren H, Zhang H, et al. Two-dimensional MXenes: From morphological to optical, electric, and magnetic properties and applications. Phys. Rep. 2020; 848, 1-58.

#3. The experimental pictures of the MXene coated pyramid matrix are missing and should be presented in the manuscript. How is the quality of the coating? The performance of the devices highly depends on the fabrication process and outcome.

Reply: The fabrication of pyramidal and multi-layered pyramidal structure was carried out by a solution process and was schematically shown in the Fig.S1 and S2 (Supporting information). Since it was fabricated through an in-situ solution process, so the intrinsic electromagnetic parameters ought to be predominant in the material. However, there might be some minor structural errors in the fabrication. As suggested, we provided the experimental sample picture of multi-layered MXene coated pyramid matrix in the revised manuscript (Supporting information). We also agree with the reviewer and mentioned the point of structure fabrication error in the revised manuscript. Following modifications have been made in the revised manuscript:
The variation of experimental results as compared to the simulated results is believed to be due to the errors in raw data fitting and structure fabrication.

**Figure S7.** (a) Schematic of broadband microwave absorption measurement technique, (b) prepared multi-layer MXene hybrid-matrix pyramid design

PS: The experiment was carried out in the Microwave Laboratory, Department of Electrical and Communication Engineering (ECE), Indian Institute of Science (IISc), Bangalore, India. Please note that because of present Covid-19 situation, Microwave Lab is being closed till September, 2020. The picture shown in Figure S7. (b) was taken earlier and more photos are not available now due to the close of the laboratory. We believe that the reviewer has well understood the situation and will recommend this manuscript to publish with the proof of experiment picture.

4. The authors claimed ultra-light MXene devices, which might be inaccurate and lack of experimental evidence. The Ti3C2Tx multi-layer devices are not necessarily lighter than graphene-based devices.

Reply: We thank the reviewer for pointing out this typographical error in the manuscript. We have modified the sentence in the revised manuscript. Following modifications have been made in the revised manuscript,

MXene, the new family of 2D materials having numerous nanoscale layers, is being considered as a novel microwave absorption material.
Reviewer: 2

Comments to the Author(s):

As a novel type of 2D materials, Mexene possesses important application prospect in field of microwave absorption materials. To enhance the microwave absorption width of Mexene-hybrid matrix materials, two types of Mexene hybrid materials were fabricated for the first time by macroscopic design approach through simulation method. The results are beneficial for the successful production of Mexene hybrid-matrix materials with wide microwave absorption bandwidth. I thus recommend its publication in this journal after minor revision as followings:

We thank the reviewer for appreciating this work and recommending its publication.

1) In the as-obtained Fe₃O₄@TiO₂-Ti₃C₂Tx, please provide the particular size of Fe₃O₄ nanoparticles, distribution, and proportion?

Reply: We thank the reviewer for this valuable suggestion. The size of Fe₃O₄ nanoparticles was obtained to be ~ 8 nm. As suggested, histogram was provided in the supporting information. Following modification have been made in the revised manuscript.

The Fe₃O₄ nanoparticle size was recorded using ImageJ software from SEM image and histogram was shown in the Figure S2 (Supporting information). The average particle size of the Fe₃O₄ nanoparticles was found to be ~ 8 nm.

Figure S2. Particle size distribution of Fe₃O₄ nanoparticles in the TiO₂-Ti₃C₂Tx MXene.
2) What is the interaction type between the Fe$_3$O$_4$ nanoparticles and Mexene matrix?

**Reply:** The interaction between Fe$_3$O$_4$ nanoparticles and MXene matrix as synthesized composite was explained elaborately in the references [17] and [18]. Since the objective of this work is towards application point of view, not synthetic oriented, we did not explain it previously. As suggested, the following modification have been made in the revised manuscript:

The interaction of Fe$_3$O$_4$ nanoparticles with MXene was explained in Ref. [17]. The exposed hydroxyl groups offer the possibility of binding with MXenes along with blending metal-oxygen stretching modes such as Fe–O and Ti–O [17].