Dear editor and reviewers:

Thank you very much for the comments on the manuscript “Preparation of biomass-based mesoporous carbon with higher nitrogen/oxygen-chelating adsorption for Cu(II) through microwave pre-pyrolysis”. We find the reviewers’ comments most helpful and genuinely appreciate your time and effort. We have made serious revision and explanation as recommended by the reviewers. Modified places are marked in red. We also deleted or corrected some words. We hope the revised paper could be in accordance with the instructions of JoVE.

**Response beneath each comment**

**Editorial comments:**

1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues. The JoVE editor will not copy-edit your manuscript and any errors in the submitted revision may be present in the published version.

**Response: The manuscript has been proofed carefully.**

2. Unfortunately, there are a few sections of the manuscript that show overlap with previously published work. Though there may be a limited number of ways to describe a technique, please use original language throughout the manuscript. Please see lines: 37-41, 52-61, 68-71, 166-167, 189-191, 218-220.

**Response: The statement on Line 52-61 (in original manuscript) has been deleted and replaced with other content according to the suggestion of reviewer 3#. Lines 37-41, 68-71, 166-167, 189-191, 218-220 (in original manuscript) have been revised to original language.**

3. Please obtain explicit copyright permission to reuse any figures from a previous publication. Explicit permission can be expressed in the form of a letter from the editor or a link to the editorial policy that allows re-prints. Please upload this information as a .doc or .docx file to your Editorial Manager account. The Figure must be cited appropriately in the Figure Legend, i.e. “This figure has been modified from [citation].”

**Response: We have gotten and uploaded the explicit permission. The figures are cited appropriately in the Figure Legend.**

4. Please upload each Figure individually to your Editorial Manager account as a .png or a .tiff file. Please combine all panels of one figure into a single image file.

**Response: Each Figure has been uploaded individually as a .png or a .tiff file.**

5. All tables should be uploaded separately to your Editorial Manager account in the form of an .xls or .xlsx file. Each table must be accompanied by a title and a description after the Representative Results of the manuscript text.

**Response: Each Table has been uploaded individually as an .xls or .xlsx file.**

6. Figure 1: Please explain the inset in its figure legend.

**Response: Revised as the editor’s advice.**

7. Please provide an institutional email address for each author if possible.

**Response:** **We have changed the original email address to an institutional email address for each author in the revised manuscript.**

8. Please rephrase the Short Abstract to clearly describe the protocol and its applications in complete sentences between 10-50 words: “Here, we present a protocol to …”

**Response: Revised as the editor’s advice.**

9. Please revise the Long Abstract to focus on the method being presented rather than the results of a specific experiment. Include a statement about the purpose of the method. A more detailed overview of the method and a summary of its advantages, limitations, and applications is appropriate. Please focus on the general types of results acquired.

**Response: Revised as the editor’s advice.**

10. Please revise the Introduction to include all of the following:

a) A clear statement of the overall goal of this method

b) The rationale behind the development and/or use of this technique

c) The advantages over alternative techniques with applicable references to previous studies

d) A description of the context of the technique in the wider body of literature

e) Information to help readers to determine whether the method is appropriate for their application

**Response: Revised as the editor’s advice.**

11. Please add more details to your protocol steps. Please ensure you answer the “how” question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action.

**Response: More details have been added to protocol steps.**

12. 1.2.2: Is 90 min the holding time at 500 °C, or the total time that the precursor stays in the furnace? Please specify.

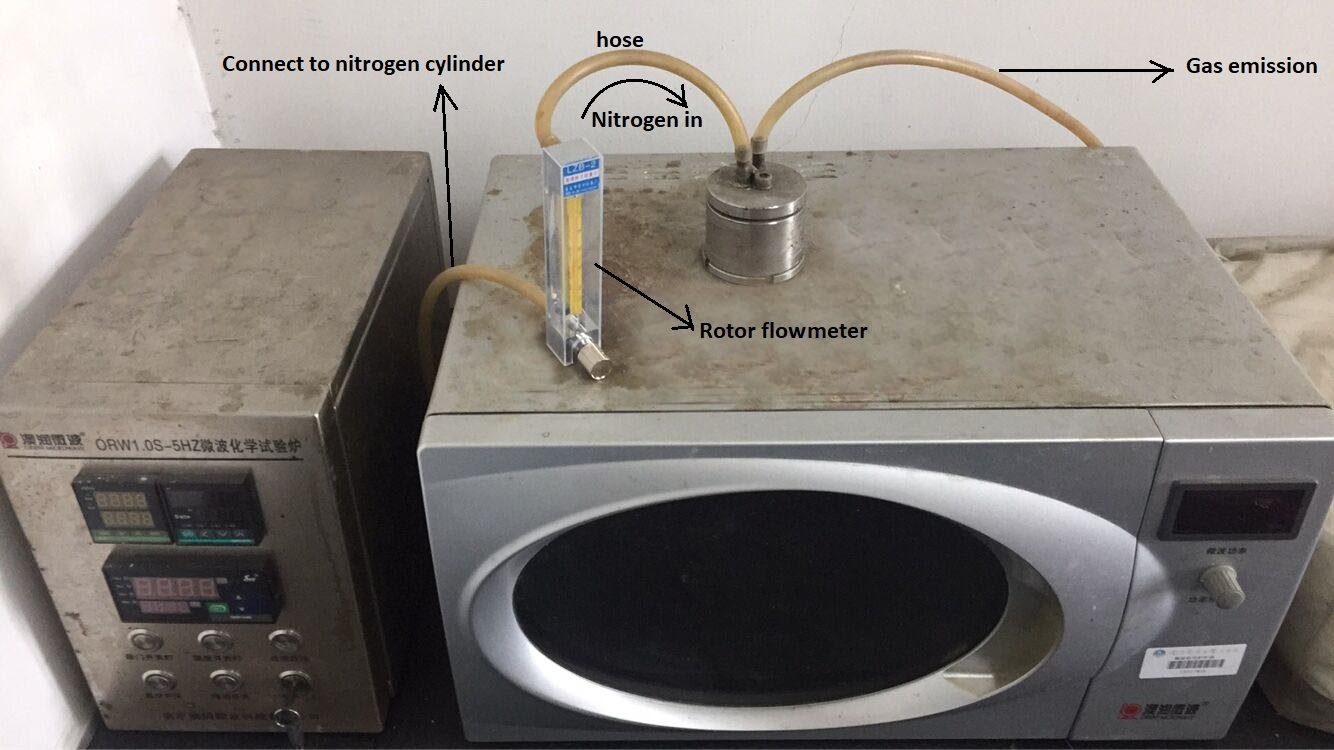
**Response: 90 min is the holding time. Relative protocol steps have been revised.**

13. 1.2.3: Please change “store” to “heat” as the temperature in the oven is relatively high (105 °C).

**Response: Revised as the editor’s advice.**

14. 1.3.2: Please describe how the nitrogen line is connected to the microwave and how the flow rate is controlled.

**Response: The N2 flow rate is controlled with a rotor flowmeter. The air inlet of the rotor flowmeter is connected to a nitrogen cylinder using a hose while the outlet is connected to the air inlet of microwave oven.**



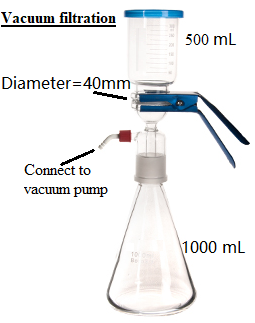
15. 1.3.3: Please describe how phosphates are removed, presumably by decanting the water? Is a filter or centrifuge used to separate the solid from the liquid?

**Response: Actually, phosphates are removed in step 1.3.4 “Filter the carbon by filter paper with vacuum filtration and rinse the sample with deionized water until…”. In order to disambiguate, the statement in step 1.3.3 “to remove phosphates (byproducts of H3PO4 with bagasse)” has been removed.**

16. Please include a space between all numbers and their corresponding units: 150 rpm; etc.

**Response: Revised as the editor’s advice.**

17. 1.3.4: Please describe how this is actually done. What is the filter size? Drying in the oven for how long?

**Response: Step 1.3.4 has been revised. Drying in the oven for 12 h. Filter the carbon by** **filter paper with vacuum filtration (as shown in the picture). In fact, the purpose of filtration is to separate the carbon from the liquid, so the size of the filter is not a factor.** 

18. 2.1.1: Is this done in a hood? If so, please specify.

**Response: Without a hood.**

19. 2.1.2/2.2.1/2.2.2: What is the stirring speed?

**Response: All steps related to stirring have been revised.**

20. 2.1.3: What is the filter size?

**Response: Filter the carbon by filter paper with vacuum filtration (See picture in comment 17) In fact, the purpose of filtration is to separate the carbon from the liquid, so the size of the filter is not a factor.**

21. 2.1.4: What is considered as a neutral pH, over 6 or 7?

**Response: The statement “a neutral pH” has been changed to “pH 6”.**

22. 2.2.3: Please specify the solution pH.

**Response: The statement “pH was constant” has been changed to “pH over 6”.**

23. 3.1.3: Please add more details here or include a reference.

**Response: We add a reference to step 3.1.3.**

24. 3.2.3: At what temperature and for how long are the carbon samples and potassium bromide dried?

**Response: The statement “fully” has been changed to “at 110 °C for 4 h”.**

25. 3.2.4: Please specify the parameters if applicable.

**Response: The parameters are not applicable. Besides, we add a reference to step 3.2.4.**

26. As we are a methods journal, please revise the Discussion to explicitly cover the following in detail in 3-6 paragraphs with citations:

a) Critical steps within the protocol

b) Any modifications and troubleshooting of the technique

c) Any limitations of the technique

d) The significance with respect to existing methods

e) Any future applications of the technique

**Response: Revised as the editor’s advice.**

27. Please do not abbreviate journal titles. Please include volume and issue numbers for all references.

**Response: Revised as the editor’s advice.**

**Reviewer 1**

Comments: The manuscript describes a GOOD work AND is well presented. Authors need following points to be included before reconsideration.

**Comment 1**: Abstract should contain some quantitative information also.

**Response: Thank you very much for your suggestion. According to the editor’s advices, the long abstract should focus on the method and its purpose *etc*., as JoVE is a methods journal. The abstract has been revised as editor’s advices, so it may not contain some quantitative information.**

**Comment 2**: English must be improved.

**Response: Thank you very much for your suggestion. The spelling and grammar issues have been proofed.**

**Comment 3**: Novelty of the work be established.

**Response: We have revised the ABSTRACT and INTRODUCTION seriously. It can be found that the novelty of the work is to present a protocol to synthesis nitrogen/oxygen dual-doped mesoporous carbon from biomass by chemical activation and two different pyrolysis methods followed up by modification. We demonstrate that the microwave pyrolysis benefits the subsequent modification process to simultaneously introduce more nitrogen/oxygen functional groups on the carbon.**

**Comment 4**: All the adsorbents reported be compared in a tabular form to establish the superiority of the work.

**Response: Revised as the reviewer’s advice. The following content is added in the revised manuscript as well as the comparison table (Table 4).**

**Table 4 compares the adsorption capacity of Cu(II) by the recently reported biomass-based activated carbon. It is found that MBAC-N has a higher adsorption capacity than other adsorbents reported in the literature, demonstrating it a promising adsorbent for removing Cu(II).**

|  |  |  |  |
| --- | --- | --- | --- |
| **Table 4.** |  |  |  |
| Adsorbents | pH | qe (mg g−1) | References |
| Wood-based granular activated carbon | 5.5 | 6.016 | Loganathan et al., 2018 |
| Baobab fruit shell-derived activated carbon | 6 | 3.0833 | Vunain et al., 2017 |
| Olive stone AC (COSAC) | 5 | 17.08 | Bohli et al., 2017 |
| Activated carbonfrom date stones | 5.5 | 18.68 | Bouhamed et al., 2016 |
| Walnut shell based activated carbon | 5 | 9.3 | Wu et al., 2018 |
| Plasma modified activated carbon | 21.4 |
| MBAC-N | 5 | 25.12 | This study |

Bohli T, Ouederni A, Villaescusa I. Simultaneous adsorption behavior of heavy metals onto microporous olive stones activated carbon: analysis of metal interactions[J]. Euro-Mediterranean Journal for Environmental Integration, 2017, 2(1):19.

Bouhamed F, Elouear Z, Bouzid J, et al. Multi-component adsorption of copper, nickel and zinc from aqueous solutions onto activated carbon prepared from date stones[J]. Environmental Science & Pollution Research, 2016, 23(16):1-6.

Loganathan P, Shim W G, Sounthararajah D P, et al. Modelling equilibrium adsorption of single, binary, and ternary combinations of Cu, Pb, and Zn onto granular activated carbon[J]. Environmental Science & Pollution Research, 2018(15):1-12.

Vunain E, Kenneth D, Biswick T. Synthesis and characterization of low-cost activated carbon prepared from Malawian baobab fruit shells by H3PO4 activation for removal of Cu(II) ions: equilibrium and kinetics studies[J]. Applied Water Science, 2017, 7(8):4301-4319.

Wu L, Wan W, Shang Z, et al. Surface modification of phosphoric acid activated carbon by using non-thermal plasma for enhancement of Cu(II) adsorption from aqueous solutions[J]. Separation & Purification Technology, 2018, 197.

**Comment 5**: Following refs. must be included in the Introduction part.

Materials Science and Engineering C ;Volume 32, Issue 1, 1 January 2012, Pages 12-17

Environmental Science and Pollution Research ;Volume 19, Issue 4, May 2012, Pages 1224-1228

Waste Management ;Volume 17, Issue 8, 1998, Pages 517-522

Journal of Colloid and Interface Science;Volume 342, Issue 2, 15 February 2010, Pages 518-527

Journal of Colloid and Interface Science ;Volume 337, Issue 2, 15 September 2009, Pages 345-354

Journal of Colloid and Interface Science ;Volume 340, Issue 1, 1 December 2009, Pages 16-26

Journal of Hazardous Materials ;Volume 185, Issue 1, 15 January 2011, Pages 17-23

RSC Advances ;Volume 2, Issue 16, 14 August 2012, Pages 6380-6388

Journal of Colloid and Interface Science ;Volume 344, Issue 2, 15 April 2010, Pages 497-507

Materials Science and Engineering C;Volume 31, Issue 5, 20 July 2011, Pages 1062-1067

Chemical Engineering Journal ; 180, 15 January 2012, Pages 81-90

Journal of Colloid and Interface Science ;Volume 371, Issue 1, 1 April 2012, Pages 101-106

Journal of Hazardous Materials ;Volume 183, Issue 1-3, November 2010, Pages 402-409

J. Mol. Liquids, 173 (2012) 153-163.

2003Separation Science and Technology 38 (2) , pp. 463-481

Environmental Engineering Research, 20(1) (2015)001-018.

Adv. Colloid Interface Sci, 211 (2014) 92-100.

Adv. Colloid Interface Sci., 193-194 (2013) 24-34.

Journal of Molecular Liquids 221 (2016) 1029-1033.

Journal of Molecular Liquids 221 (2016) 930-941.

Scientific Reports 6 (2016) 31641.

Journal of Saudi Chemical Society 19(5) (2015) 521-527.

Journal of Molecular Liquids 209 (2015) 374-380

RSC Advances 5 (2015)34645-34651.

Journal of Colloid and Interface Science 452 (2015) 126-133.

Journal of the Taiwan Institute of Chemical Engineers 45 (2014) 1910-1917.

Journal of Molecular Liquids 198 (2014) 409-412.

Journal of Molecular Liquids 193 (2014) 160-165

Materials Science and Engineering C 33 (2013) 2235-2244.

Journal of Molecular Liquids 181 (2013) 133-141.

Materials Science and Engineering C 33 (2013) 4725-4731.

Materials Science and Engineering: C 33 (2013) 91-98.

Journal of Molecular Liquids 177 (2013) 394-401.

Journal of Molecular Liquids 178 (2013) 88-93.

Environmental Science and Pollution Research 20 (5), 2828-2843(2013)

RSC advances 2 (22), 8381-8389(2012)

Journal of Colloid and interface Science 362 (2), 337-344(2011)

Encyclopedia of Surface and Colloid Science, 2004 Update Supplement 5, 1,2004

RSC advances 2 (22), 8381-8389,2012

Journal of Colloid and interface Science 362 (2), 337-344 ,2011

Materials Science and Engineering: C 33 (1), 91-98,2013

Industrial & Engineering Chemistry Research 50 (24), 13589-13613,2011

Journal of colloid and interface science 362 (2), 457-462 , 2011

Advances in colloid and interface science 211, 93-101,2014

Separation and purification technology 89, 245-251,2012

J. Mol. Liquids 173(2012)153-163.

International Journal of Environmental Analytical Chemistry 84, 947-964 (2004).

Environmental Science and Pollution Research 21, 3218-3229 (2014).

Journal of Molecular Liquids 219, 858-864 (2016).

Journal of Molecular Liquids 224, 171-176 (2016).

Journal of Molecular Liquids 211, 457-465 (2016).

Materials Science and Engineering: C, 33( 2013) 2235-2244.

J. Colloid Interface Sci., 452 (2015)126-133.

Chem Eng J. 284 (2016) 687-697.

RSC Adv., 5 (2015) 34645- 34651.

J. Mol. Liq., 177 (2013) 394-401.

Chemical Eng. J., 268(2015) 28-37.

J Colloids Surface Sci., 417 (2014) 420-430.

Materials Science & Engineering C 33 (2013)4725-4731.18438 - 18450.

Materials Science and Engineering CVolume 33, Issue 4, 1 May 2013, Pages 2235-2244

journal of Colloid and Interface ScienceVolume 452, August 05, 2015, Pages 126-133

Chemical Engineering JournalVolume 284, January 15, 2016, Pages 687-697

Journal of Molecular LiquidsVolume 177, January 2013, Pages 394-401

RSC Adv., 5 (2015) 18438 - 18450.

**Response: Thank you very much for your suggestion. We have learned a lot from the literatures you have provided. Taking into account the length of the paper and the content relevance of these references, twelve most relevant references have been cited in the revised manuscript, as follows:**

**Journal of colloid and interface science 362 (2), 457-462, 2011**

Adsorption process of methyl orange dye onto mesoporous carbon material–kinetic and thermodynamic studies

**Advances in colloid and interface science 211, 93-101,2014**

Processing methods, characteristics and adsorption behavior of tire derived carbons: A review

**RSC Adv., 5 (2015) 18438 - 18450.**

Removal of basic dye Auramine-O by ZnS:Cu nanoparticles loaded on activated carbon: optimization of parameters using response surface methodology with central composite design

**Environmental Science and Pollution Research 20 (5), 2828-2843(2013)**

Sorption of pollutants by porous carbon, carbon nanotubes and fullerene- An overview

**Environmental Science and Pollution Research; Volume 19, Issue 4, May 2012, Pages 1224-1228**

Column with CNT/magnesium oxide composite for lead(II) removal from water

**Industrial & Engineering Chemistry Research 50 (24), 13589-13613,2011**

Rice Husk and Its Ash as Low-Cost Adsorbents in Water and Wastewater Treatment

**Journal of Colloid and Interface Science ;Volume 340, Issue 1, 1 December 2009, Pages 16-26**

Adsorptive removal of hazardous anionic dye “Congo red” from wastewater using waste materials and recovery by desorption

**Adv. Colloid Interface Sci., 193-194 (2013) 24-34.**

Adsorptive removal of dyes from aqueous solution onto carbon nanotubes: A review

**Journal of Molecular Liquids 211, 457-465 (2016).**

Green Synthesis of Iron Nano-Impregnated Adsorbent for Fast Removal of Fluoride from Water

**Chemical Eng. J., 268(2015) 28-37.**

Modeling of competitive ultrasonic assisted removal of the dyes – Methylene blue and Safranin-O using Fe3O4 nanoparticles

**Chemical Engineering Journal, Volume 284, January 15, 2016, Pages 687-697**

Removal of hazardous dyes-BR 12 and methyl orange using graphene oxide as an adsorbent from aqueous phase

**Chemical Engineering Journal; 180, 15 January 2012, Pages 81-90**

Cadmium removal and recovery from aqueous solutions by novel adsorbents prepared from orange peel and Fe2O3 nanoparticles

**Reviewer 2**

Comments: This paper presents the use of modified activated carbon produced from microwave pyrolysis in the adsorption of copper ion. The protocol provided are descriptive. The results and discussions presented would be useful to the reader. Following are my comments for improvements:

**Comment 1**: Line 87-88, it's confusing on how the 15 wt% of phosphoric acid solution was prepared in 1:1 mass ratio. What is the SI unit for the "mass"?

**Response: The statement “Place 30 g of fine bagasse powder into 15 wt% of phosphoric acid solution in a 1:1 mass ratio for 24 h.” means “Place 30 g of fine bagasse powder into 30 g of phosphoric acid solution (15 wt%) for 24 h”. The ratio of their weight is 1:1. In order to disambiguate the sentences, all “mass” has been replaced by “weight”.**

**Comment 2**: Line 200-203, in order to claim the modified activated carbon produced from microwave pyrolysis contained more nitrogen/oxygen functional groups than the one prepared from conventional pyrolysis, the author should present the results of elemental composition (C, H, N, O, S) of both samples to support their claim.

**Response: Revised as the reviewer’s advice. The results of elemental composition of samples are listed in Table 1.**

**Comment 3**: The author should justify why the word "pre-pyrolysis" is used instead of "pyrolysis". Also, they should be consistent to the use of one term.

**Response: In this study, the pyrolysis process is followed up by modification. And we demonstrated that the microwave pyrolysis benefits the follow-up introduction of surface functional groups to improve the adsorption capacity than electric-heating pyrolysis. So the word “pre-pyrolysis” is used instead of “pyrolysis” in original manuscript. For ease of understanding, we have replaced “pre-pyrolysis” with “pyrolysis” in the text of the revised manuscript.**

**Comment 4**: In the introduction, the author should describe why chemical activation was preferred than physical activation. Following articles are suggested as both references and citations to the author:

-Microwave-assisted pyrolysis with chemical activation, an innovative method to convert orange peel into activated carbon with improved properties as dye adsorbent

-Production of activated carbon as catalyst support by microwave pyrolysis of palm kernel shell a comparative study of chemical versus physical activation.

**Response: Thank you very much for your suggestion. The INTRODUCTION has been revised as the reviewer’s advice. These references have been cited in the revised manuscript.**

**Comment 5**: The author should also describe why one step activation was preferred.

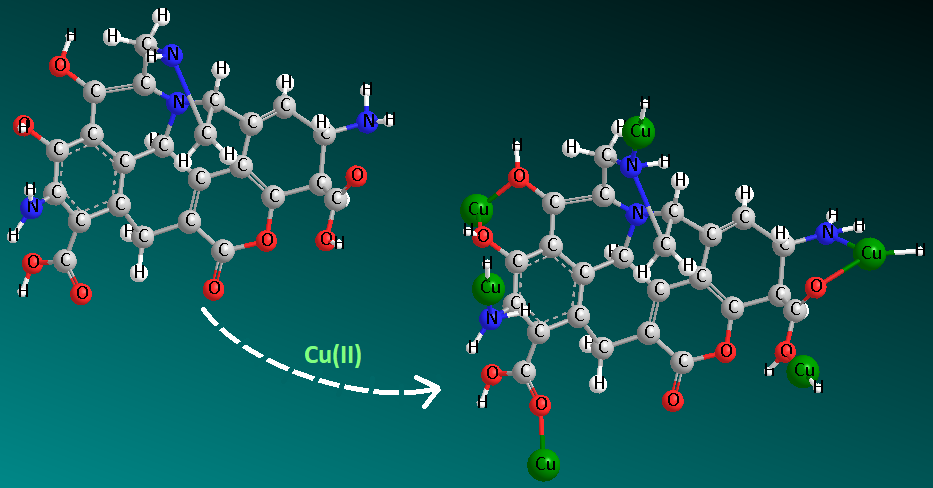
**Response: Thank you very much for your suggestion. Revised as the reviewer’s advice.**

**Reviewer 3**

Comments: This manuscript "Preparation of biomass-based mesoporous carbon with higher nitrogen/oxygen-chelating adsorption for Cu(II) through microwave pre-pyrolysis" is logical, the organization of sentence is good.

**Comment 1**: While they only describe the adsorption capacity increase after doping in this article, the reasons for the adsorption capacity increase did not explain, which needs to supplement. The manuscript should be improved according to the comments before considering for publications.

**Response: Thank you very much for your suggestion. Revised as the reviewer’s advice. The mechanism analysis is added in the revised manuscript as well as the reaction scheme (Figure 5).**

**Comment 2**: The analysis of mechanism needs to be strengthened instead of describe the increase in nitrogen/oxygen-chelating can improve the adsorption capacity of Cu(II) only. The specific adsorption mechanism of Cu(II) need to be supplement.

**Response: Revised as the reviewer’s advice. The mechanism analysis is added in the revised manuscript as well as the reaction scheme (Figure 5).**

**Comment 3**: The logical of INTRODUCTION should be organized, the conventional electric-heating doesn't need to contrast with Microwave irradiation too much, and the feasibility analysis of doping or advantage analysis of doping in previous should be used to elicit your own experimental research.

**Response: Thank you very much for your suggestion. The INTRODUCTION has been revised as the reviewer’s advice.**

**Comment 4**: The advantages of preparation of biomass-based mesoporous carbon need to be highlighted in comparison with other studies, and the adsorption mechanism of Cu(II) after doping should be appointed in.

**Response: Revised as the reviewer’s advice. The following content is added in the revised manuscript as well as the comparison table (Table 4).**

**Table 4 compares the adsorption capacity of Cu(II) by the recently reported biomass-based activated carbon. It is found that MBAC-N has a higher adsorption capacity than other adsorbents reported in the literature, demonstrating it a promising adsorbent for removing Cu(II).**

|  |  |  |  |
| --- | --- | --- | --- |
| **Table 4.** |  |  |  |
| Adsorbents | pH | qe (mg g−1) | References |
| Wood-based granular activated carbon | 5.5 | 6.016 | Loganathan et al., 2018 |
| Baobab fruit shell-derived activated carbon | 6 | 3.0833 | Vunain et al., 2017 |
| Olive stone AC (COSAC) | 5 | 17.08 | Bohli et al., 2017 |
| Activated carbonfrom date stones | 5.5 | 18.68 | Bouhamed et al., 2016 |
| Walnut shell based activated carbon | 5 | 9.3 | Wu et al., 2018 |
| Plasma modified activated carbon | 21.4 |
| MBAC-N | 5 | 25.12 | This study |

Bohli T, Ouederni A, Villaescusa I. Simultaneous adsorption behavior of heavy metals onto microporous olive stones activated carbon: analysis of metal interactions[J]. Euro-Mediterranean Journal for Environmental Integration, 2017, 2(1):19.

Bouhamed F, Elouear Z, Bouzid J, et al. Multi-component adsorption of copper, nickel and zinc from aqueous solutions onto activated carbon prepared from date stones[J]. Environmental Science & Pollution Research, 2016, 23(16):1-6.

Loganathan P, Shim W G, Sounthararajah D P, et al. Modelling equilibrium adsorption of single, binary, and ternary combinations of Cu, Pb, and Zn onto granular activated carbon[J]. Environmental Science & Pollution Research, 2018(15):1-12.

Vunain E, Kenneth D, Biswick T. Synthesis and characterization of low-cost activated carbon prepared from Malawian baobab fruit shells by H3PO4 activation for removal of Cu(II) ions: equilibrium and kinetics studies[J]. Applied Water Science, 2017, 7(8):4301-4319.

Wu L, Wan W, Shang Z, et al. Surface modification of phosphoric acid activated carbon by using non-thermal plasma for enhancement of Cu(II) adsorption from aqueous solutions[J]. Separation & Purification Technology, 2018, 197.

**Comment 5**: Line 218'The higher becomes the temperature, the better are adsorption properties.'should be 'The adsorption properties of samples for Cu(II) become better with the temperature increasing.'；Line 228-229'The above results confirm that the adsorption of MBAC-N on copper is chemisorption and it has a promising ability to remove copper ions.'should be'The above results confirm that the adsorption of Cu(II) on MBAC-N is chemisorption and MBAC-N is an promising absorbent to remove Cu(II).'；

**Response: Thank you very much for your suggestion. Revised as the reviewer’s advice.**

**Comment 6**: Words in this paper should keep consistent, as'Cu(II)'in Line 222 and Line 223 ；'copper ion'in Line 158 and Line 224.

**Response: Thank you very much for your suggestion. All terms “copper ion” have been replaced by “Cu(II)” as the reviewer’s advice.**

**Comment 7**: The references should be quoted as far as possible for nearly three years. For example, the following papers with relative topics about activated carbons prepared or modified by microwave pyrolysis, such as Chemical Engineering and Processing, 2015, 91: 67-77; Journal of Industrial and Engineering Chemistry, 2016, 39:27-36; Korean Journal of Chemical Engineering, 2015, 32(6): 1129-1136, Bioresource Technology, 2016, 201: 41-49, etc. which are valuable for being referred for analysis the results in manuscript.

**Response: Revised according to the reviewer’s advices. Some old references have been replaced by recent relevant papers in the revised manuscript. These references have been cited in the revised manuscript.**

**Comment 8**: In Fig. 4, which kind of AC was used to adsorb the Cu(II) ?

**Response: Figure 4 shows representative adsorption property analysis of MBAC-N.**