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Judul setelah revisi	Investigation of the influence of crosslinking activation methods on the physicochemical and Cu(II) adsorption characteristics of cellulose hydrogels

A manuscript number has been assigned: JECE-D-21-06186

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Title: Study on the influence of linking activation procedure on the physicochemical properties and adsorption performance of cellulose hydrogels
Journal of Environmental Chemical Engineering

Dear Dr. Santoso,

Your submission "Study on the influence of linking activation procedure on the physicochemical properties and adsorption performance of cellulose hydrogels" has been assigned manuscript number JECE-D-21-06186.

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Ms. Ref. No.: JECE-D-21-06186
Title: Study on the influence of linking activation procedure on the physicochemical properties and adsorption performance of cellulose hydrogels
Journal of Environmental Chemical Engineering

Dear Dr. Santoso,

Thank you for submitting the above paper to Journal of Environmental Chemical Engineering. Your manuscript needs major revisions.

The reviewers comments are included below for your attention. Please carefully address the issues raised in the comments. I invite you to submit your revised manuscript.

The due date for submitting your revised manuscript is Oct 21, 2021

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I look forward to receiving your revised manuscript.

Yours sincerely,

Guilherme Luiz Dotto, Ph.D
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Journal of Environmental Chemical Engineering
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Reviewers' comments:

Reviewer #2: Manuscript Number: JECE-D-21-06186
Title: Study on the influence of linking activation procedure on the physicochemical properties and adsorption performance of cellulose hydrogels
For: Journal of Environmental Chemical Engineering

Overview：
In this study, the authors synthesized cellulose hydrogels with two different cross-linking activation methods, including water bath heating and microwave radiation, and investigated the physicochemical properties, adsorption behavior, and reusability of the cellulose hydrogels produced by these two different cross-linking activation methods. In addition, this manuscript evaluates the economic cost of two crosslink-activated hydrogels and examines the kinetic and isothermal adsorption properties of copper ions on two hydrogels. This paper has a guiding value for the in-depth study of new hydrogels by different activation methods in the future, and the research results have certain scientific practice in engineering applications. However, after careful assessment of the manuscript, the reviewer found that this manuscript

still has much work to do, for example, characterization, data analysis and discussion need to be strengthened. Therefore, I recommend a major revision so
that the quality of this can be improved again to meet the journal's publication requirements.

Specific Comments:
Graphical Abstract
1. It's good.

Highlights section
2. Highlights consist of a short collection of bullet points that capture the novel results of your research as well as new methods that were used during the study (if any). Although the authors provide three highlights, these are responses to synthetic methods or describe what was done, and there are no results or quantitative data. I suggest that the author seriously reconsider and add new highlights.
For detailed information, please refer to https://www.elsevier.com/journals/journal-of-environmental-chemical-engineering/2213-3437/guide-for-authors

Title section
3. The adsorption performance is too vague, and it should be clearly referred to as copper ion.

Abstract section
4. Line 6, "scarcely", is this word used incorrectly? Why do I feel like this sentence is contradictory?
5. The presentation of the results is a little less, if necessary, I suggest that the adsorption mechanism or macroscopic research significance could be added at the end of the abstract.
7. Lines 14~15, How much is this "utility costs", please specify. Lines 17~18, Is this theoretical maximum adsorption of WBH, or MRH, and shouldn't it be 261 mg g-1? The 236 mg g-1 here is an error.
8. The keywords are a little inappropriate, if necessary, I suggest adding "water heat".

Introduction section
9. This abstract is very nice, but I think there are still some necessary descriptions. The last paragraph, pages 5~6, this study objective and the abstract are slightly repetitive, and I think it would be better if they were changed to specific characterization items, adsorption kinetic models and isothermal models.

Experimental section
10. Page.6 Lines 6~13, Have these reagents been further purified? Please add a description.
11. P.7 Line 4, What are the specific specifications of this "cm×cm molds" ? Is it 1 cm×1 cm? Some detailed information about the origin, model and manufacturer of the instrument, please add.
12. I think it would be more attractive to readers if Atomic Force Microscope (AFM), Thermogravimetric Analysis (TGA), and Swelling properties were characterized for the two hydrogels.
13. The reaction temperature is important in the adsorption process, please explain in section 2.4
To better investigate the mechanism of adsorption and adsorption characteristics, the classic isotherms models (Langmuir, Freundlich, Temkin and Dubinin-Radushkevich model) or common kinetic models (pseudo-first-order, pseudo-second-order, Elovich, and Intraparticle diffusion model) should be applied and illustrated. If it is necessary, more model information please read the following literature:
Tümsek, F., Avcı, Ö., 2013. Investigation of Kinetics and Isotherm Models for the Acid Orange 95 Adsorption from Aqueous Solution onto Natural Minerals. J. Chem. Eng. Data 58(3), 551-559.
T. Wang, J. Zheng, H. Liu, Q. Peng, H. Zhou, X. Zhang, Adsorption characteristics and mechanisms of Pb2+ and Cd2+ by a new agricultural waste-Caragana korshinskii biomass derived biochar, Environ. Sci. Pollut. Res. (2020).

Results and Discussion section
14. Pages 9~12, Are these formation mechanisms supported by similar literature, please? If so, please enhance the verification and discussion.
15. Pages 12 Line 6, I think the "3.1.1 part" can be "3.2 part", and the original 3.2 part will follow in turn.
16. The elemental analysis of SEM mapping is a bit inaccurate, but it does show the percentage of elements. Page14 Line 8-17, the XRD analysis is to depend on JADE software or ICDD's crystal database (PDF card) analysis it, rather than citing other people's literature, which belongs to plagiarism of other people's ideas. What is the JCPDS standard card number of FP-cellulose? More detailed information on FTIR spectroscopy can be found on the following websites: the "Spectroscopic Tools" website (http://www.science-and-fun.de/tools/).
17. This Economic cost analysis is very well done, but please tell me which region is the reference for the price standard in Table 1, and what is the reference for the average cost globally? Please add, if possible, a more detailed calculation using the life cycle assessment method.
I give this comment with reference to the following literature

T. Wang, G. Li, K. Yang, X. Zhang, K. Wang, J. Cai, J. Zheng, Enhanced ammonium removal on biochar from a new forestry waste by ultrasonic activation: Characteristics, mechanisms and evaluation, Sci. Total Environ. 778 (2021) 146295. https://doi.org/10.1016/j.scitotenv.2021.146295.
18. The "3.3.1. section" could be discussed with the relevant literature based on the available characterization data after adsorption, and then compile a few sentences on possible adsorption mechanisms of copper ions by hydrogels as part of the conclusion or abstract. I think this will enhance the quality of the paper.
19. Pages 24 Line 4, For the isotherm adsorption model and parameter KL and RL, please refer to the original literature. The essential characteristic of a Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor RL (or equilibrium parameter), originally defined by Hall et al. (1966) and applied by many groups (Mohan et al. 2002 and 2007). Please refer to other literature to enhance the data analysis and writing in this section.
D. Mohan, C.U. Pittman, Jr., M. Bricka, F. Smith, B. Yancey, J. Mohammad, P.H. Steele, M.F. Alexandre-Franco, V. Gomez-Serrano, H. Gong, Sorption of arsenic, cadmium, and lead by chars produced from fast pyrolysis of wood and bark during bio-oil production, J. Colloid Interface Sci. 310 (2007) 57-73.
D. Mohan, K.P. Singh, Single- and multi-component adsorption of cadmium and zinc using activated carbon derived from bagasse—an agricultural waste, Water Res. 36 (2002) 2304-2318.
K.R. Hall, L.C. Eagleton, A. Acrivos, T. Vermeulen, Pore- and Solid-Diffusion Kinetics in Fixed-Bed Adsorption under Constant-Pattern Conditions, Industrial & Engineering Chemistry Fundamentals 5 (1966) 212-223.
T. Wang, D. Zhang, K. Fang, W. Zhu, Q. Peng, Z. Xie, Enhanced nitrate removal by physical activation and Mg/Al layered double hydroxide modified biochar derived from wood waste: Adsorption characteristics and mechanisms, J. Environ. Chem. Eng. (2021) 105184.
20. What do the numbers in parentheses in Table 2 mean?

Conclusion section
21. The conclusion is a bit short and could be enhanced with some future research perspectives.

References section
22. The reference format of this manuscript does not meet the requirements of the journal, please refer to the "guide for authors" for details, or the recently published papers are in this journal.
https://www.elsevier.com/journals/journal-of-environmental-chemical-engineering/2213-3437/guide-for-authors.

Date: Dec 05, 2021
To: "Shella Permatasari Santoso" shella_p5@yahoo.com
From: "Journal of Environmental Chemical Engineering" support@elsevier.com
Subject: Your Submission - JECE-D-21-06186R2

Ms. Ref. No.: JECE-D-21-06186R2
Title: Investigation of the influence of crosslinking activation methods on the physicochemical and Cu(II) adsorption characteristics of cellulose hydrogels
Journal of Environmental Chemical Engineering

Dear Dr. Santoso,

I am pleased to inform you that your manuscript "Investigation of the influence of crosslinking activation methods on the physicochemical and Cu(II) adsorption characteristics of cellulose hydrogels" has been accepted for publication in Journal of Environmental Chemical Engineering.

Below are comments from the editor and reviewers.

Once your paper is entered in our Production system, we aim to provide you with a typeset proof within 24 hours.

Thank you for submitting your work to Journal of Environmental Chemical Engineering.

Your accepted manuscript will now be transferred to our production department and work will begin on creation of the proof. If we need any additional information to create the proof, we will let you know. If not, you will be contacted again in the next few days with a request to approve the proof and to complete a number of online forms that are required for publication.

Yours sincerely,

Guilherme Luiz Dotto, Ph.D
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Investigation of the influence of crosslinking activation methods on the physicochemical and Cu(II) adsorption characteristics of cellulose hydrogels

Article reference
JECE_106971
Journal
Journal of Environmental Chemical Engineering
Corresponding author
Shella Permatasari Santoso
First author
Shella Permatasari Santoso
Received at Editorial Office
25 Aug 2021
Article revised
9 Nov 2021
Article accepted for publication
5 Dec 2021
DOI
10.1016/j.jece.2021.106971

Last update: 12 Jan 2022

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10/1
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Journal of Environmental Chemical Engineering

Investigation of the influence of crosslinking activation methods on the physicochemical and Cu(II) adsorption characteristics of cellulose hydrogels

--Manuscript Draft--

Manuscript Number:	JECE-D-21-06186R2
Article Type:	Research Paper
Keywords:	Cellulose hydrogel; Epichlorohydrin; Crosslinking; Microwave irradiation; Water-bath heating; Adsorption
Corresponding Author:	Shella Permatasari Santoso, Ph.D. Widya Mandala Catholic University: Universitas Katolik Widya Mandala Surabaya Surabaya, INDONESIA
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Abstract:	Hydrogels represent an attractive soft material with promising applications in many fields. Conventional methods for hydrogel preparation typically involve heat curing either by water-bath (WB) heating or microwave (MW) irradiation to facilitate crosslinking. However, a detailed investigation of the influence of WB- and MW-mediated crosslinking process on the characteristics of hydrogels has been rarely reported. This work aims to evaluate the physicochemical properties of epichlorohydrin (ECH) crosslinked cellulose hydrogels prepared by WB and MW heating methods, including pore morphology, chemical composition, crystallinity, thermal stability, and water absorption capacity. The results showed that MW heating could accelerate the crosslinking reaction between cellulose and ECH to produce robust hydrogel, with a 3 min total irradiation time at 400 W compared to 2 h at 60 °C under conventional WB heating. The total utility cost to produce WBH is US$ 0.052, which is 14 times higher than MWH (US$ 0.004). The estimation of the total production cost of MWH on a large scale is US$ 2.86 per kg. Moreover, the as-prepared MWH displayed outstanding performance in Cu(II) removal at 30 °C and pH 7, with a maximum adsorption capacity of 119 mg/g, respectively. The kinetic and equilibrium behaviors of Cu(II) ions on WBH and MWH were best described by pseudo-second order and Langmuir isotherm models, respectively.

Widya Mandala Catholic University Surabaya
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Tel: +62 31 3893933 ext.: 103, 111, Fax: +62 31 3891267

November 10, 2021

Dear Editor,

Attached, please find the revised article JECE-D-21-06186_R2. We have addressed the suggestions and comments by the Reviewer, and the answers are given in a point-by-point fashion. The revised parts have been highlighted in green color font in the revised manuscript. We would like to thank the Reviewers and Editor for the given chance to revise our manuscript, we believe that the revised manuscript now is acceptable for publication in the *Journal of Environmental Chemical Engineering*. Thank you again for giving us the opportunity to revise this manuscript.

Sincerely,

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