

Confirming submission to Journal of Molecular Liquids

From: Journal of Molecular Liquids (em@editorialmanager.com)

To: sheila_p5@yahoo.com

Date: Saturday, January 30, 2021 at 03:19 PM GMT+7

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Manuscript Number: MOLLIQ_2020_6318R2

Biocomposite hydrogel beads from glutaraldehyde-crosslinked phytochemicals in alginate for effective removal of methylene blue

Dear Dr Santoso,

We have received the above referenced manuscript you submitted to Journal of Molecular Liquids.

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Journal of Molecular Liquids

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Decision on submission to Journal of Molecular Liquids

From: Journal of Molecular Liquids (em@editorialmanager.com)

To: sheila_p5@yahoo.com

Date: Friday, November 6, 2020 at 12:03 AM GMT+7

Manuscript Number: MOLLIQ_2020_6318

Biocomposite hydrogel beads from glutaraldehyde-crosslinked phytochemicals in alginate for effective removal of methylene blue

Dear Dr Santoso,

Thank you for submitting your manuscript to Journal of Molecular Liquids.

I have completed my evaluation of your manuscript. The reviewers recommend reconsideration of your manuscript following major revision. I invite you to resubmit your manuscript after addressing the comments below. Please resubmit your revised manuscript by Jan 04, 2021.

When revising your manuscript, please consider all issues mentioned in the reviewers' comments carefully: please outline every change made in response to their comments and provide suitable rebuttals for any comments not addressed. Please note that your revised submission may need to be re-reviewed.

To submit your revised manuscript, please log in as an author at <https://www.editorialmanager.com/molliq/> and navigate to the "Submissions Needing Revision" folder.

Journal of Molecular Liquids values your contribution and I look forward to receiving your revised manuscript.

Kind regards,
Artur Valente
Editor-in-Chief

Journal of Molecular Liquids

Editor and Reviewer comments:

Reviewer #1: In this paper, authors has studied the role of plant phenolic extracts in improving the adsorption capacity of alginate beads for methylene blue dyes. The authors states the major adsorption occurs due to physio-sorption with significant adsorption capacity, however the beads lack reusability.

Major comments:

The manuscript lacks the relevant scientific discussion regarding mass transfer, intra-particle diffusion and endothermic adsorption phenomenon. Authors must highlight the role of coating in the results obtained as that is the major objective of the study, the beads are well-reported for high adsorption capacity. Authors must re-write the discussion describing the role of plant extract in the entire process. Few other comments are listed below:

1. What were the optimized pH conditions for Alg, Alg/RCE and BHB?
2. Figure 5: Data of unmodified alginate is not shown? The qm value of Alg and BHB are reported as 1055.4 and 1388.5 mg/g, respectively. However, these two systems may have different optimum conditions?
3. How 8 h was optimized for adsorption experiments?
4. Figure 1a: With the coating of RCE, why there is no increase in intensity or presence of new peaks (phenolic or other aromatic compounds in the crude extract used) in the FTIR spectra?
5. Line 223: Authors have not provided the SEM image for all three samples, as stated in the text to make conclusive statements. Verify.
6. Line 238: Authors should explain what is meant by 'functional adsorption groups', and what type of adsorption is involved in the stated case.
7. Line 243: Authors should discuss the importance of 'mass transfer' as driving force with in terms of Co. (Refer DOI:10.1016/j.jece.2016.12.017, 10.1007/s11356-018-2280-z).
8. Provide standard error bars in each figure and add units in the axis title. For example Figure 6.

9. Table 1: From the Table its clearly Redlich-Peterson model, however authors have not described why Langmuir is chosen above all.

10. Since beads are non-recyclable, authors should highlight the cost analysis of the adsorbent, to support its importance over existing biosorbents.

Minor comments:

1. Abstract: Authors should capitalize each keyword initials
2. Line 60: Authors should use water 'bodies' instead of 'body'.
3. Line 72, 195, 201, 283: Authors should check the sentence formation.
4. Line 78: Authors should check the appropriateness of 'calcium' instead of 'sodium'.
5. Line 129-130, 161: Authors should avoid mentioning results in the 'Materials and methods' section.
6. Line 134,156: Authors should use single abbreviation for particular compound. Does the Alg powder stands for sodium alginate? Authors should explain the abbreviation in the first place of occurrence and use the same throughout the manuscript.
7. Line 235: Use 'respectively' while discussing more than one independent results.
8. Line 284: Authors must write the sentence more scientifically.

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Decision on submission to Journal of Molecular Liquids

From: Journal of Molecular Liquids (em@editorialmanager.com)

To: sheila_p5@yahoo.com

Date: Wednesday, February 3, 2021 at 05:14 PM GMT+7

Manuscript Number: MOLLIQ_2020_6318R2

Biocomposite hydrogel beads from glutaraldehyde-crosslinked phytochemicals in alginate for effective removal of methylene blue

Dear Dr Santoso,

Thank you for submitting your manuscript to Journal of Molecular Liquids.

I am pleased to inform you that your manuscript has been accepted for publication.

My comments, and any reviewer comments, are below.

Your accepted manuscript will now be transferred to our production department. We will create a proof which you will be asked to check, and you will also be asked to complete a number of online forms required for publication. If we need additional information from you during the production process, we will contact you directly.

We appreciate you submitting your manuscript to Journal of Molecular Liquids and hope you will consider us again for future submissions.

Kind regards,
Artur Valente
Editor-in-Chief

Journal of Molecular Liquids

Editor and Reviewer comments:

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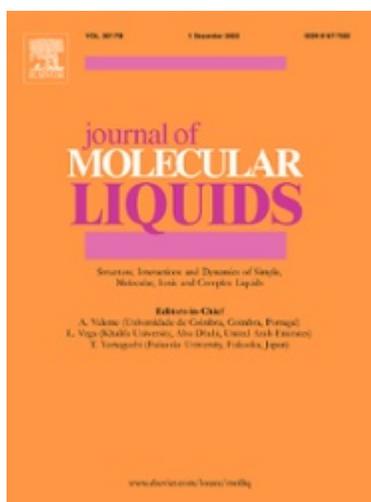
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Corresponding author

Shella Permatasari Santoso

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Andreas Andreas

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Journal of Molecular Liquids

Biocomposite hydrogel beads from glutaraldehyde-crosslinked phytochemicals in alginate for effective removal of methylene blue

--Manuscript Draft--

Manuscript Number:	MOLLIQ_2020_6318R2
Article Type:	Full length article
Section/Category:	Water, aqueous solutions and other hydrogen-bonded liquids
Keywords:	Alginate bead; Red cabbage extract; Phytochemicals; Adsorption methylene blue; Intraparticle diffusion.
Corresponding Author:	Shella Permatasari Santoso Widya Mandala Surabaya Catholic University Surabaya, East Java Indonesia
First Author:	Andreas Andreas
Order of Authors:	Andreas Andreas Zefanya Gerald Winata Shella Permatasari Santoso Artik Elisa Angkawijaya Maria Yuliana Felycia Edi Soetaredjo Suryadi Ismadji Hsien-Yi Hsu Alchris Woo Go Yi-Hsu Ju
Abstract:	<p>Phytochemicals, i.e., flavonoids, phenolics, and anthocyanin, extracted from red cabbage, were crosslinked with alginate to prepare biocomposite hydrogel beads (BHB). The preparation of BHB involved three consecutive steps: (1) extraction and solvent reduction of phytochemicals from red cabbage, (2) crosslinking of phytochemicals into alginate matrix using glutaraldehyde, and (3) formation of the hydrogel beads in CaCl₂ solution. The resulting BHB sorbents were characterized using scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), and X-ray diffraction (XRD) analyses. The cross-section structure of the BHB was confirmed from the SEM images. The alteration of FTIR peaks implied the success of the crosslinking of phytochemical compounds into the alginate. The adsorption equilibrium and kinetic studies of BHB were conducted using basic blue 9 (BB9) as the model adsorbate. FTIR characterization of the BHB post-adsorption reveals the functional groups of the adsorbent involved in the dye adsorption. The calculated adsorption isotherm, kinetics, and thermodynamic parameters show good agreement with the characterization results of adsorbate post-adsorption. The adsorption isotherm is being in congruence with the Langmuir model, and the highest adsorption capacity recorded was 1442.0 mg g⁻¹ at 323 K and pH of 11.0. Adsorption kinetics was better fitted to the pseudo 1st order model than the pseudo 2nd order and Elovich models, which further support the dye physisorption behavior. The initial adsorption rate was influenced by the rapid surface adsorption followed by intraparticle diffusion. The thermodynamic parameters show the spontaneity of the adsorption, and the adsorption proceeds endothermically. The cost analysis shows the economic feasibility of BHB sorbent production for adsorption applications.</p>
Suggested Reviewers:	CH ZHOU clay@zjut.edu.cn Jaka Sunarso jsunarso@swinburne.edu.my

	Alfin Kurniawan alfin_kur@yahoo.com
Opposed Reviewers:	
Response to Reviewers:	<p>Editor and Reviewer comments:</p> <p>Reviewer #1: The authors answered properly to all the comments and questions, and added comprehensive explanations to support their answers. Consequently, the quality of the manuscript was considerably improved. However, few issues are listed below:</p> <p>1. Figure 3: Why there is no change in the removal ability of Alg/RCE, and Alg/RCE removal capacity at pH 11, the optimized pH? Response: We thank the reviewer for this insightful comment. The corresponding discussion has been added in the revised manuscript. Please refers to P16, L285-288.</p> <p>2. Figure 7: Why there is no change in the adsorption capacity of Alg and Alg/RCE? Response: The not significant increase in the adsorption capacity of Alg/RCE compare to Alg can be related to the absence of GA molecules in the preparation of Alg/RCE. The absence of GA in Alg/RCE cause the polyphenols to be easily removed from the Alg matrix, especially during the washing step in the sample preparation procedure. The corresponding discussion has been provided in the revised manuscript, L254-265.</p> <p>3. Authors should rearrange the SEM image based on magnification index. Response: We thank the reviewer for the suggestion. For the SEM images, we tried to show the two sections of the samples, which is the surface morphology and the cross-section morphology; and we would like to arrange the images based on this section instead of their magnification index. We have modified the SEM images in Figure 2 to verify the arrangement of the images.</p>



WIDYA MANDALA CATHOLIC UNIVERSITY SURABAYA
CHEMICAL ENGINEERING DEPARTMENT
FACULTY OF ENGINEERING
#37, Kalijudan Rd., Surabaya 60114, East Java, Indonesia
Tel. +6231 389 1264, Fax. +6231 389 1267

January 30, 2021

Dear Editor:

I am submitting the revised (R2) manuscript entitled “Biocomposite hydrogel beads from glutaraldehyde-crosslinked phytochemicals in alginate for effective removal of methylene blue” for re-consideration of publication in *Journal of Molecular Liquids*. In this revised manuscript, we have carefully addressed all the comments raised by the reviewer and responded to each of the comments in a point-by-point fashion. The changes and additions made in the revised manuscript are highlighted in blue-colored fonts. We hope the editor and the reviewers will be satisfied with our response to the ‘comments’ and the revision for this manuscript for reconsideration by the journal. Thank you for giving us the opportunity to revise this manuscript and I look forward to hearing from you soon.

Sincerely yours,

Shella Permatasari Santoso, Ph.D.
Widya Mandala Surabaya Catholic University
Chemical Engineering Department
Faculty of Engineering