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Response to Reviewer Comments

Please refer to track-change version appended below to spot the location and changes made in response to point-by-point of reviewer comments. The track-change version of the revised manuscript is appended in this response document.

Reviewer #1: General opinion

Comment 1: The analysis of the presented measurements are incomplete, in most cases the conclusions are missing.

Response 1: We improved the manuscript. Please refer to Section 3.1 and 3.2. We added some more discussion of the XRD patterns, nitrogen physisorption, SEM, and the sample testing for ammonium removal.

Comment 2: The greatest weakness of the article is the continuous misuse of English grammar, which requires major editing.

2: We improved the English for the whole manuscript as indicated by blue color in the manuscript.

Reviewer #2:

The authors obtained a zeolite sample from the Bayah region of Indonesia that contained impurities and had a low surface area. The aim of the paper was to improve the textural properties of the natural zeolites and remove the impurities using a hydrothermal treatment.

Comment 1: The authors show that they have managed this, by treating the parent zeolite for 8 hours and for 24 hours, however more work should be performed to prove that the 8-hour time for hydrothermal treatment is the most appropriate. Only selecting two time periods means that the optimal could be significantly different and the whole image is not seen.

Response 1: We treated samples for three time periods of hydrothermal treatment, i.e. 8, 24 and 32 hours. All samples including the parent were tested for ammonium sorption. The effect of hydrothermal time on ammonium removal are shown in Figure 5. The effect of hydrothermal time is clearly seen. We agree that the optimum point could be different. Perhaps the optimum condition lay in between 8 and 24 h of hydrothermal time. However, our aim in this study is not to optimize the conditions. We highly appreciate your suggestions.

Comment 2: The characterisation is performed well but the discussion must be improved. For example, the SEM discussion is poor and lacks scientific descriptions, i.e. the use of 'bigger in quantity'.

Response 2: The manuscript has been revised. Please refer to Section 3.1.

Comment 3: Additionally, the reflections in figure 1 should be assigned to the correct phases.

Response 3: We assigned the peaks with m for mordenite, c for clinoptilolite and q for quartz. Please refer to Figure 1.

Comment 4: In general, the English used in the manuscript is not of publication quality. For example, line 184, 'has no pore' is used, what is meant by this?

Response 4: The sentence "has no pore" replaced with "has no porosity". Please refer to the manuscript. We improved the English for the whole manuscript as indicated by blue color in the manuscript.

Comment 5: For this paper to be published further work is required, extensive English editing, improved discussion, and more aging times.

Response 5: We improved discussion on almost all results and discussion parts such as the XRD analysis, FTIR, SEM, BET and the ammonium testing.

Reviewer #3:

Comment 1: This paper presents results of experimental treatment to recrystallize a natural zeolite (mordenite) sample from Bayah in Java. The experiments were designed to assess the optimum duration of hydrothermal treatment to maximize ammonium adsorption of the product. Appropriate methods and analytical approaches were deployed, and the results are interpreted correctly.

Response 1: Thank you very much.

Comment 2: The English language needs improvement. In the attached file I have indicated necessary improvements to make the text understandable. Further improvements are advisable to achieve a higher quality, but this should not be the reviewer's responsibility.

Response 2: The manuscript has been revised according to reviewer suggestion. Please refer to the manuscript. Other improvements of the English language were also performed. The modified sentences and words are in blue color.

Comment 3: (Figure 4) The scale bars are too small to be visible. Add visible scale bars to all 6 images.

Response 3: We inserted new scale bars. Please refer to Figure 4.

Comment 4: In my opinion, the topic of the paper is of regional interest, specifically to Indonesian readers and perhaps to industries that use zeolites for ammonium adsorption. Although a brief mention is made of zeolite from another location in East Java (lines 210-215), apart from this, no comparisons of the results are made with other natural zeolite occurrences. The authors are advised to expand this part of the paper by providing comparisons of their results (including optimum duration of hydrothermal reactions) with zeolite occurrences globally, not only occurrences in Java. The comparisons should include other zeolite species in addition to mordenite. This would greatly increase the scientific value and interest to readers of the journal.

Response 4: Thank you for your suggestions. We have added a table of comparison between Bayah natural zeolites with other natural zeolites which is various in zeolite phases and the method of modification. We also compared Bayah Natural zeolite with synthetic natural zeolite. Please refer to Table 2 and discussion in Section 3.2

Table 2. Comparison of hydrotreated Bayah natural zeolites with other zeolites

No	Origins of Zeolites	Phase	Treatment	Initial [NH ₄ ⁺] (mg/L)	% Ammonium removal		Ref
					Before modification	After modification	
1	Bayah, Indonesia	Mordenite, clinoptilolite, quartz	Hydrothermal	100	48.9	62.6	This work
2	Shimane Prefecture, Japan	Mordenite	Hydrothermal	180	32	64	(16)
3	Lingshou, China	Clinoptilolite-Ca, stilbite-Ca	NaNO ₃ impregnation followed by calcination	5	41.8	81.7	(21)
4	Kardzhali, Bulgaria	Clinoptilolite	Washing followed by drying	100	95	-	(22)
5	Semnan, Iran	Clinoptilolite	No modification	100	87	-	(23)
6	Ponorogo, Indonesia	Mordenite	NaOH	10	30	50-80	(11)
7	Synthetic	NaY	No Modification	100	70	-	(24)

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Reviewer 1

Comments and Suggestions for Authors

The author(s) made a number of efforts to increase the content of the article and improved the grammar accordingly. In this form the article is acceptable.

[Response: Thank you for your valuable comments.](#)

Reviewer 2

Comments and Suggestions for Authors

[Comment 1:](#) The authors have taken some of the comments on board, however most of my concerns still exist. The authors have highlighted that they tested a 32 h HT zeolite. However, no characterisation is provided for this zeolite. This is a significant flaw of the study. How have conclusions been drawn when the textural properties are not known. The characterisation should be added to the paper before the paper is considered for publication.

[Response 1:](#) Thank you for your suggestions. We agree that the 32 h sample characterization would be improve the quality of our manuscript. From our previous work, the longer hydrothermal treatment to zeolites from Klaten, Indonesia lead to quartz phase (1). The quartz as competitor phase growth considerably after 24 h hydrothermal treatment zeolite as seen in the XRD patterns (Figure 1). The quartz reduces ammonium sorption as seen in sample after 24 h hydrothermal treatment (Figure 5). The ammonium sorption of HT 32 h is lower than 24 h, hence, we assumed that the HT 32 h sample would have higher quartz phase.

[Comment 2:](#) Some of the scientific discussion has been improved, however there are still spelling mistakes in the manuscript, such as “precent” in line 234 and “syntetic” in line 237.

[Response 2:](#) Manuscript has been revised.

[Comment 3:](#) The addition of the literature comparison table is good.

[Response 3:](#) Thank you.

Reviewer 3

[Comment 1:](#) The manuscript is considerably improved, both in terms of English language and in the presentation and discussion of the data. Of particular value is the incorporation of a table and text comparing ammonium adsorption of the Bayah treated

mordenite with mordenite and clinoptilolite from other locations in similar studies. This favourable comparison needs to be mentioned in the abstract and conclusions.

[Response 1: Thank you for your suggestions. The manuscript has been revised. Please refer to abstract and conclusions.](#)

Comment 2: Below is a list of grammatical and spelling corrections that I noted while reading the revised manuscript, plus a suggested concluding sentence. With these corrections implemented, I would consider the manuscript to be suitable for publication.

Line 19 - change "its" to "their"

Line 30 - add a final sentence to the abstract, the same of similar to that suggested for the Conclusions: see note regarding line 295.

Line 79 - change "grounded" to "powdered"

Line 94 - change "performed" to "used" and delete "by using"

Line 167 - change "Quartz as phase competitor was pronounce" to "As a phase competitor, quartz was pronounced"

Line 169 - delete the first occurrence of "of"

Line 198 - delete "It" and replace the preceding full stop with a comma (i.e. join the sentences)

Line 199 - change "reduce" to "reducing"

Line 207 - change "The microimages of SEM of" to "SEM micro-images of"

Line 209 - replace "poses" with "displays"

Line 213 - replace "needle shape" with "needle-like shape (arrowed)"

Line 230 - spelling should be "phillipsite"

Line 233 - spelling should be "conversion"

Lines 234-235 - this sentence is ambiguous, should it be joined to the previous sentence with a comma?

Line 250 - change "percent removal ammonium eventhough without" to "percentage of ammonium removal even without"

Line 253 - delete "much"

Line 271 - delete "the"

Line 295 - Add another sentence to the conclusions, commenting on the comparative values shown in Table 2. Suggested sentence: "In terms of ammonium removal efficiency, hydrothermally treated Bayah mordenite compares favourably with treated mordenite from other locations worldwide, although clinoptilolite provides higher removal capacities than mordenite."

[Response 2: The manuscript has been revised. Thank you very much for your valuable comments.](#)

References

1. Kurniawan T, Muraza O, Bakare IA, Sanhoob MA, Al-Amer AM. Isomerization of n-Butane over Cost-Effective Mordenite Catalysts Fabricated via Recrystallization of Natural Zeolites. *Industrial & Engineering Chemistry Research*. 2018;57(6):1894-902.