Hasanudin Hasanudin <hasanudin@mipa.unsri.ac.id>

Your submission has been assigned (#IJC-2208-1961)

1 pesan

Iranian Journal of Catalysis <journals@iau.ir> 13 Agustus 2022 pukul 21.49
Balas Ke: Iranian Journal of Catalysis <ijc@iaush.ac.ir>
Kepada: hasanudin@mipa.unsri.ac.id, hasanudinkf@gmail.com
Cc: massah@iaush.ac.ir

Dear Prof. **Dr. Hasanudin Hasanudin**

Your submission entitled "**Diisopropyl Ether Production via Isopropanol Catalytic Dehydration Over Zirconium Phosphate Modified Natural Zeolite**" has been assigned the following manuscript number: IJC-2208-1961.

You may check on the progress of your paper by logging on to the Journal Editorial System as an author. The URL is http://ijc.iaush.ac.ir.

Thank you for submitting your work to this journal.

Kind regards,

Ahmad Reza Massah

Editor-in-Chief of Iranian Journal of Catalysis

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Hasanudin Hasanudin <hasanudin@mipa.unsri.ac.id>

Manuscript Needs Revision (Major Revision) (#IJC-2208-1961 (R1))

1 pesan

Iranian Journal of Catalysis <journals@iau.ir> 30 Oktober 2022 pukul 11.17
Balas Ke: Iranian Journal of Catalysis <ijc@iaush.ac.ir>
Kepada: hasanudin@mipa.unsri.ac.id, hasanudinkf@gmail.com

Dear Prof. Dr. Hasanudin Hasanudin

I can now inform you that the reviewers have evaluated your manuscript. Major revision has been requested.
Attached you can find comments from the reviewers. Please, submit the revised manuscript with blue marking on the revised parts. The revised manuscript should include Figures, Tables, and other files that you mentioned in the manuscript. With the resubmission, please enclose a detailed description of your revision in response to the reviewers' comments.

I hope you will find the comments to helpful and informative enough. I am looking forward to receiving your revision at your earliest convenience.

Thank you for submitting your work to **Iranian Journal of Catalysis**.

Kind regards,

Ahmad Reza Massah

Editor-in-Chief of *Iranian Journal of Catalysis*

Reviewers Recommendation:

Reviewer 1:

File Sent by Reviewer:
https://ijc.shahreza.iau.ir/jufile?__file=JoynnYHcJnrv0aylQwlyDEhRb7ZLG6B5vz_pgoT0TbM.yyUaAEwSOYniONV2rN8.FRVi.UaPvca0A2XjkG8Do2h3xQ1s_yrKHyruZIcdWIXu8sm7gfDlcdRlfg7oEF4I1KYtlzJJH4m0PNQUDV_WeQK7qIAWB0tTdy8YBXPQlmfx8OwWAkhZo0dnGxrImM9JxrDbaIYG98_CbArjsykd_I58dyLsEDKHcq.1b7w2A88-

Reviewer Comment For Author:

Dear Author(s)
The manuscript has a well idea and discuss a very important environmental issue. But there are many of requirements that must be conducted to show the importance of the paper. all the required modifications are pointed in the attachment.

Reviewer 2:

Reviewer Comment For Author:

Research work focused on the synthesis and characterization of Zr(H2PO4)4 zeolites as isopropanol conversion catalysts.
The authors obtained conclusive results, but many missing remarks were noted in order to complete and publish this work in the journal, such as:

1. The introduction is very long, and it contains the majority of the number of references in the manuscript (44 references). The authors must specify the recent and important references in relation to the work, and minimize the introduction.

2. The authors must add in detail the experimental methods: EDX, N2 adsorption/desorption, IFTR (pyridine), in the experimental part.

3;The results obtained in the table show values on the pore diameter, specific surface, and structural volume of the catalysts. The authors should add the experiments of these results in the manuscript as an example. BJH, BET (diameter and specific surface), atomic % (method ???)

4;The catalytic application part is not clear. The authors must add information on the catalytic application with a diagram and text on the device used in this reaction, as well as the reactor used in this reaction (diameter, volume ...), type of GC (the column..), type MS, flow rates after the reaction,..

5;The results of IR using pyridine are not clear compared to previous research. The method IF (pyridine) provides a qualitative analysis of the acid sites on the surface of the catalysts, contrary to the results obtained by the authors, which only show the bands and frequencies (IFTR).

6;The authors should find an explanation of the relationship between the degree of impregnation of the metal with the acidity and the catalytic activity of the catalysts, the selectivity, and the rate of conversion with previous research references.

Reviewer 3:

Reviewer Comment For Author:

The paper reports "Synthesis of SO4/ZrO2 Catalyst and its Application in the Conversion of Ethanol to Diethyl Ether ". I recommend a revision of the present manuscript.
1- Keywords must be revised:
(Natural zeolite, modified zeolite) change to zeolite, modification,
diisopropyl ether production change to diisopropyl ether
2- Formula must be presented with parameters (NOT with the expressions), and the parameters pointed in formula should be explained in the text.
3- Section 3.2: In relation to similar works done, it is better that the reffernced works be presented in a Table, and type of the catalyst, process conditions, selectivity and yield be reported in the Table.

4- The reference of the zeolite preparing method should be mentioned.

5- Section 2.4: authors said that the amount of the used catalyst was 0.5 g. What is the base of this selection?
6- The name of the country of analysis devices should be mentioned.

7- Authors referred to researches for dehydration reaction’s catalyst. Some of them are old, such as references of [24, 26, 28,…]. It is better these references are revised. For example, reff. [28] can be substituted with Korean J. Chem. Eng., 28(7), 1593-1598 (2011).
8- Table 2: “Surface area (m2/g)” must be revised to Specific surface area (m2/g)

9-The conclusion and the Abstract are the same. They must be revised.
10- Fig. 4: the important peaks must be assigned in the Fig., with the related wave number.

Title : Diisopropyl Ether Production via Isopropanol Catalytic Dehydration Over Zirconium Phosphate Modified Natural Zeolite Manuscript ID : IJC-2208-1961 (R1)		
	Thank you for giving us the opportunity to submit a manuscript titled Diisopropyl Ether Production via Isopropanol Catalytic Dehydration Over Zirconium Phosphate Modified Natural Zeolite" for publication in the Iranian Journal of Catalysis. We appreciate the time and effort that you dedicated to providing feedback on our manuscript and are grateful for the insightful comments and valuable improvements to our paper. We have incorporated the suggestions made by the reviewers. Those changes are written in yellow highlight text within the manuscript.	
	Reviewer 1 Dear Author(s) The manuscript has a well idea and discuss a very important environmental issue. But there are many of requirements that must be conducted to show the importance of the paper. all the required modifications are pointed in the attachment.	
No	Comment from Reviewer	Responses
1	Replaced by "results"	Thank you for pointing this out. We have revised the text from "research" to "results" accordingly.
2	This sentence must be removed because there is no need to mention a details conclusion in abstract	Thank your for pointing this out. We have removed the details as suggested by the reviewer.
3	Long sentence, need to reformulated	Thank you for pointing this out. We have shortened the sentences.

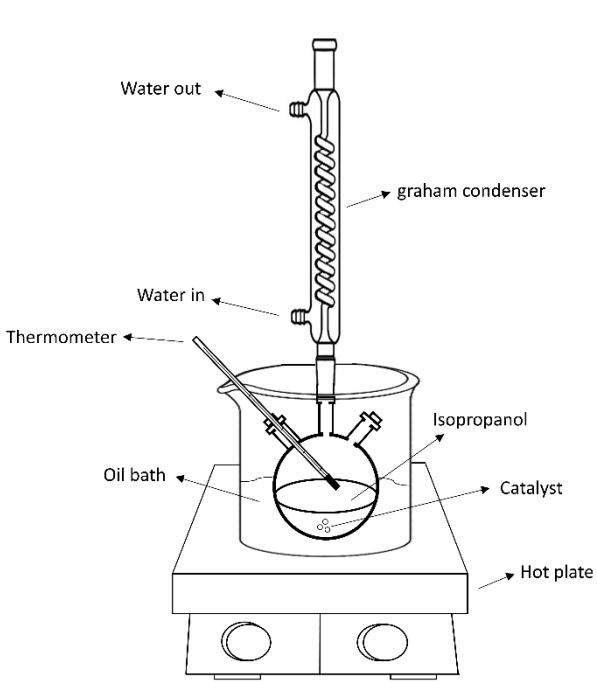
		The revised text as follows: "The highest isopropanol conversion (66.73%) was attained by 8 mEq/g zeolite-Zr(H2PO4)4 followed by the DIPE yield and selectivity up to 35.81% and 47.8%, respectively."
4	Again, this sentence represents a conclusion, so must be deleted from the abstract	Thank you for pointing this out. We have revomed the sentences.
5	General explanation. Need re-write and focus on the importance of the chemical additives to improve the fuel and decrease the toxic emission (in numbers)	Thank you for pointing this out. We have rearranged the sentences, deleted the general explanation, and revised the sentences. The revised text as follows: "Kale et al. [6] reported that the NO_x emision on HCCI combustion could reduce from 96 ppm to 81 ppm as the increase of DIPE load from 10 to 60% on gasoline blends. Uyumaz et al. [7] reported that the DIPE with 40% loaded could increase the power output to 24.7% at a lambda of 2 and 1000 rpm with a maximum indicated thermal efficiency of 23.4% at a lambda of 2.33 on HCCI combustion, which suggested that DIPE may be able to increase the range at which HCCI can operate while preventing knocking."
6	Please, modify this paragraph by adding a suitable part about the cost and economy of using DIPE.	Thank you for pointing this out. According to the study conducted by Uyumaz et al. [7], the effects of diisopropyl ether on cylinder pressure and heat release rate with different lambda values showed that For HCCI combustion, diisopropyl ether is regarded as a cost-effective and environmentally friendly addition because the test engine could not be operated at 1200 rpm with fuel blends with lambda values of 2. The revised text as follows:

		"Further, diisopropyl ether is regarded as a cost-effective and eco-friendly fuel additive based on the cylinder pressure and heat release rate study on HCCI combustion."
7	The mention of these references is very good, but not enough. More details about the limitations, conversion, selectivity, etc. must be mention to show the importance of this work.	Thank you for pointing this out. The catalysts that have been mentionted exhibited high conversion at particular conditions. However, some of these catalysts have drawbacks due to low selectivity and yield towards DIPE and require high-cost precursors. The revised text as follows: "These catalysts exhibited high conversion at particular conditions. However, some of these catalysts have drawbacks due to low selectivity and yield towards DIPE and require high-cost precursors."
8	If the alumina-silicates has these properties, What is the difference between it and Zeolite in using? i.e. why you use Zeolite in this work?	In this study, we have employed low-cost catalyst supported, i.e., activated natural zeolite from Bayan, Central java, using HF and HCl. Since it was taken from nature, their properties are relatively different depending on where they took and prepared. We used zeolite in this work since it has tuneable properties with high acidity value, thus promoting the dehydration reaction.
9	Experimental work details, no need to mention it in the introduction. Please, delete this sentence	Thank you for pointing this out. We have deleted this sentence as suggested by the reviewer.
10	Please, mention the times and the time required for washing	Thank you for pointing this out. The washing process took 8 times and required 24 h for each washing. The revised text as follows:

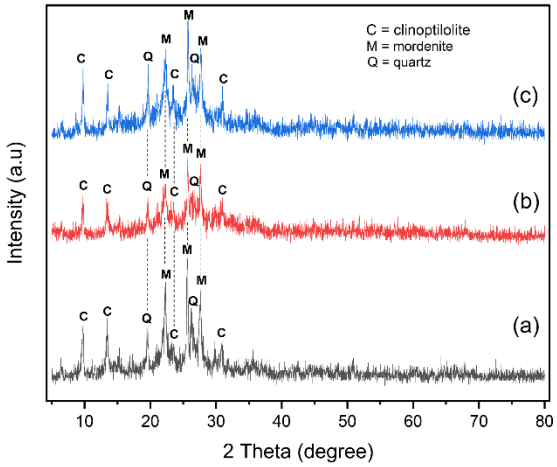
		"Afterward, 100 g of natural zeolite was immersed in a 1% HF solution, subsequently stirred for 1 hour, and then washed with distilled water 8 times each for 24 h"
11	What is the volume of HCl used? Please, clarify it	Thank you for pointing this out. We used 6 N HCl (125 mL) for 4 hours to activate the natural zeolite. The revised text as follows: "The natural zeolite was later immersed in a 6 N HCl solution (125 mL) for 4 hours, filtered, and washed using distilled water until the pH was close to neutral."
12	Please, clarify the volume used clearly (i.e. by numbers).	Thank you for pointing this out. We used 25, 50, 70, 75, 100, and 125 mL of Zr4+ precursors, which correspond to 2, 4, 6, 8, and 10 mEq/g, respectively. The revised text as follows: "Firstly, 5 g of as-prepared natural zeolite was dispersed on 0.1 M ZrOCl2.8H2O solution by varying the volume of Zr4+ precursor (25, 50, 70, 75, 100, and 125 mL corresponding to 2, 4, 6, 8, 10 mEq/g, respectively) and stirred for 1 hour utilizing a magnetic stirrer (SH-2 Corona) at an ambient temperature."
13	Mention the name and type of the stirrer i.e. if the stirring performed via magnetic or ordinary stirrer	Thank you for pointing this out. We used a magnetic stirrer (SH-2 Corona) for stirred the solution. The revised text as follows: "……for 1 hour utilizing a magnetic stirrer (SH-2 Corona) at an ambient temperature."

14	What is the equipment used to keeping the rate is constant?	Thank you for pointing this out. We used a burette to keep the constant rate. The revised text as follows: "….solution was gradually dropped using a burette into the mixture at a rate of 1 mL/min until it reached….."
15	Mention it clearly by number in (ml)	Thank you for pointing this out. We used 10, 20, 30, 40, and 50 mL of 1 M NH4H2PO4 solution, corresponding to the 2, 4, 6, 8, and 10 mEq/g zirconium phosphate solution, respectively. The revised text as follows: "Afterward, the 1 M NH4H2PO4 (≥ 99% purity, Merck) solution was gradually dropped using a burette into the mixture at a rate of 1 mL/min until it reached 10, 20, 30, 40, and 50 mL volumes, corresponding to 2, 4, 6, 7, 10 mEq/g of zirconium phosphate, respectively, and stirred for one day."
16	Give a suitable reason for increasing the temperature	Thanks for pointing this out. The reason for increasing the temperature is to increase the evaporation of water, thus the paste forms rapidly.
17	How can you know removing all Cl- from the paste? Clarify that and mention the name of equipment used	Thank you for pointing this out. The free Cl- ions can be indicated by no white precipitation on the filtrate formed after being tested by the $AgNO_3$ solution (0.01 M). The revised text as follows: "The free Cl- ions can be indicated by no white precipitation on the filtrate formed after being tested by the AgNO3 solution (0.01 M)."
18	What is the time required for drying?	Thank you for pointing this out. The paste was dried in an oven at 375.15 K for 24 hours.

		The revised text as follows: “The paste was later washed with distilled water until free from Cl- ions and dried in an oven at 378.15 K for 24 h.”
19	This paragraph needs to add several parts about the important of each test	Thank you for pointing this out. The characterizations such as XRD, FTIR, SEM-EDX, N2 adsorption-desorption, and acidity features were critical either to confirm the success of synthesis or to see their relation to the catalysis study. The XRD analysis was used to determine the crystal structure or phase of the catalyst. The FTIR analysis was employed to determine the functional groups of the catalyst. The N2 adsorption-desorption and SEM-EDX were employed to determine the textural properties and the morphological surface along with their constituent element, respectively. Further, the FTIR-pyridine and the acidity analysis using the gravimetric method with pyridine as a probe to evaluate the acidity features of the catalyst. The revised text as follows: “The crystal structure and phase of natural and modified zeolite were assessed using the X-ray diffractometer Rigaku MiniFlex 600. FTIR Shimadzu-Prestige 21 was utilized with the KBr pellet technique for functional group analysis. The textural characteristic was evaluated using N2 physisorption at 77.35 K in a Quantachrome instrument. The catalyst was vacuum degassed to 300 ˚C with a heating rate of 10 ˚C/min for 60 min. The multi-point BET method was employed to determine the catalysts' surface area, the pore features were determined by the BJH method, whereas the external surface area and the micropore area were evaluated using the t-plot method. The catalysts' morphology and elemental composition were inspected using a Tescan Vega 3 scanning electron microscope assisted with X-ray energy dispersive spectroscopy. The pyridine-adsorbed gravimetric method was employed to evaluate their surface acidity features.”

20	The dehydration unit must be clarified by a photographic picture and schematic diagram. Please add this requirements to the section 2.4	Thank you for pointing this out. We have added the schematic diagram of regarding of isopropanol dehydration reaction using reflux system. The revised text as follows:  Fig 1. Schematic diagram of dehydration reaction using reflux system

21	The conditions of GC-MS used to detection the products must be mention clearly	Thank you for pointing this out. The GC-MS (Thermo Fisher Scientific) with TG-5MS columns was used to analyze the product. The initial oven programmed temperature was 32 ˚C for 2.5 min and ramped at 3˚C/min to a final temperature of 45 ˚C for 2 min, with an H_2 as a carrier gas (1 mL/min). The injection temperature was 200 ˚C. The MS transfer line temperature was 230 ˚C, whereas the ion source temperature was 210 ˚C. The revised text as follows: “Reaction products were determined using GC-MS (Thermo Fisher Scientific) with TG-5MS columns. The initial oven programmed temperature was 32 ˚C for 2.5 min and ramped at 3˚C/min to a final temperature of 45 ˚C for 2 min, with an He as a carrier gas (1 mL/min). The injection temperature was 200 ˚C. The MS transfer line temperature was 230 ˚C, whereas the ion source temperature was 210 ˚C.”
22	Figure 1 should be modified to show the slight modification of 2θ	Thank you for pointing this out. We have revised the sentences as well the figure accordingly. The revised text as follows: “Furthermore, there were slight shifts of 2θ from ~20 to 30˚ after natural zeolite modification using zirconium and zirconium phosphate, which was presumably due to the stress formation by the dissimilarity in ionic size between natural zeolite, Zr, and Zr(H2PO4)4 ions”

		
23	Repeated sentence. So, please deleted	Thank you for pointing this out. We have deleted the sentence.
	Reviewer 2 Research work focused on the synthesis and characterization of Zr(H2PO4)4 zeolites as isopropanol conversion catalysts. The authors obtained conclusive results, but many missing remarks were noted in order to complete and publish this work in the journal, such as:	
1	The introduction is very long, and it contains the majority of the number of references in the manuscript (44 references). The authors must specify the recent and important references in relation to the work, and minimize the introduction.	Thank you for pointing this out. We have shortened the introduction content. The references in the indroduction section were reducted to 33 references.

2	The authors must add in detail the experimental methods: EDX, N2 adsorption/desorption, IFTR (pyridine), in the experimental part.	Thank you for pointing this out. We have added the detail of the characterization technique. We also would like to clarify that we did not conduct the ex-situ IFTR pyridine due to a technical issue. Instead, we evaluate the catalyst acidity analysis using the gravimetric method based on other previous studies since we know the surface acidity value quantitatively, which seems more practical. However, it is necessary to consider IFTR pyridine in further research. The revised text as follows: "The crystal structure and phase of natural and modified zeolite were assessed using the X-ray diffractometer Rigaku MiniFlex 600 (Japan). FTIR Shimadzu-Prestige 21 (Japan) was utilized with the KBr pellet technique for functional group analysis (recorded from 4500 to 500 cm^{-1}). The textural characteristic was evaluated using N_2 physisorption at 77.35 K in a Quantachrome instrument (USA). The catalyst was vacuum degassed to 300 ˚C with a heating rate of 10 ˚C/min for 60 min. The multi-point BET method was employed to determine the catalysts' surface area, the pore features were determined by the BJH method, whereas the external surface area and the micropore area were evaluated using the t-plot method. The catalysts' morphology and elemental composition were inspected using a Tescan Vega 3 (Czech Republic) scanning electron microscope (recorded at 5000× magnification with HV of 15 kV) assisted with X-ray energy dispersive spectroscopy (Bruker QUANTAX, US). The gravimetric method was employed to evaluate the surface acidity features of catalysts utilizing a pyridine base [35]."
3	The results obtained in the table show values on the pore diameter, specific surface, and structural volume of the catalysts. The authors should add the experiments of these results in the manuscript	Thank you for your valuable comment. We have revised the text and added the experimental details regarding the N_2 adsorption-desorption method, as suggested by the reviewer. The revised text as follows:

	as an example. BJH, BET (diameter and specific surface), atomic % (method ???)	"The textural characteristic was evaluated using N2 physisorption at 77.35 K in a Quantachrome instrument. The catalyst was vacuum degassed to 300 ˚C with a heating rate of 10 ˚C/min for 60 min. The multi-point BET method was employed to determine the catalysts' surface area, the pore features were determined by the BJH method, whereas the external surface area and the micropore area were evaluated using the t-plot method."
4	The catalytic application part is not clear. The authors must add information on the catalytic application with a diagram and text on the device used in this reaction, as well as the reactor used in this reaction (diameter, volume ...), type of GC (the column..), type MS, flow rates after the reaction,..	Thank you for pointing this out. We have added the details regarding the GCMS operational condition. In this study, we employed the batch reactor consisting of a reflux system (graham condenser) and a 3-neck round flask (100 mL) as a sample container. The round flask was placed in an oil bath equipped with a thermometer, and the temperature of the reaction was controlled by the hot plate. The schematic diagram of the dehydration reaction has been added accordingly.

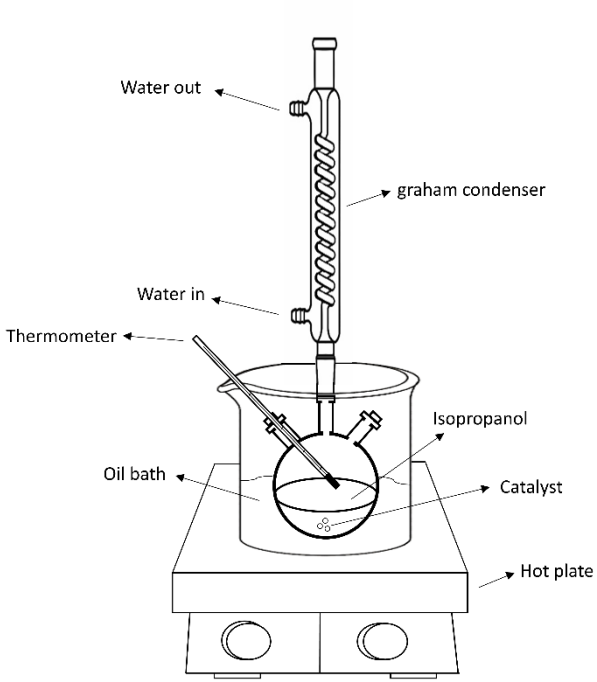


Fig 1. Schematic diagram of dehydration reaction using reflux system

The revised text as follows:

"The activities of the catalysts were evaluated by the dehydration of isopropanol, performed in a batch reactor consisting of a reflux system (graham condenser) and a 3-neck round flask (100 mL) as a sample container. The round flask was placed in an

		oil bath equipped with a thermometer, and the temperature of the reaction was controlled by the hot plate (Fig. 1) using 50 mL of isopropanol and 0.5 g of catalyst." "Reaction products were determined using GC-MS (Thermo Fisher Scientific) with TG-5MS columns. The initial oven programmed temperature was 32 ˚C for 2.5 min and ramped at 3˚C/min to a final temperature of 45 ˚C for 2 min, with an He as a carrier gas (1 mL/min). The injection temperature was 200 ˚C. The MS transfer line temperature was 230 ˚C, whereas the ion source temperature was 210 ˚C."
5	The results of IR using pyridine are not clear compared to previous research. The method IF (pyridine) provides a qualitative analysis of the acid sites on the surface of the catalysts, contrary to the results obtained by the authors, which only show the bands and frequencies (IFTR).	Thank you for pointing this out. In this manuscript, we would like to clarify that the FTIR analysis that we provided was the catalyst itself, not the catalyst pyridine absorbed. Regarding the acidity of the catalyst, we employed the gravimetrical method using pyridine as a probe. In this context, we explain the relation of the surface acidity features (mmol/g) with the zirconium phosphate loading as well as their relation to the isopropanol dehydration to the DIPE. However, it is necessary to explore this aspect in future studies.
6	The authors should find an explanation of the relationship between the degree of impregnation of the metal with the acidity and the catalytic activity of the catalysts, the selectivity, and the rate of conversion with previous research references.	Thank you for pointing this out. The previous report employed the nickel phosphate-zeolite catalyst which showed that the metal loading of 8 mEq/g also exhibited the highest catalytic activity towards DIPE production. The effect of acidity due to the metal phosphate impregnation on the zeolite were dominant in this process. "Based on the previous study [85], the metal phosphate loading during the impregnation process significantly enhanced the dehydration of isopropanol to DIPE, in which high loading of up to 8 mEq/g was found to be sufficient to promote the optimum selectivity and yield towards DIPE. At this condition, the metal-phosphate interaction produced a harmonious effect, which was suggested due to the existence of both Lewis and Bronsted acidic sites, although the textural and the morphological surface might affect the catalytic activity but were slightly dominant."

	Reviewer 3 The paper reports "Synthesis of SO4/ZrO2 Catalyst and its Application in the Conversion of Ethanol to Diethyl Ether ". I recommend a revision of the present manuscript.	
1	Keywords must be revised: (Natural zeolite, modified zeolite) change to zeolite, modification, diisopropyl ether production change to diisopropyl ether	Thank you for pointing this out. We have revised the text and changed the keywords, accordingly. The revised text as follows: “Keywords: isopropanol conversion, diisopropyl ether, zeolite, modification, zirconium phosphate”
2	Formula must be presented with parameters (NOT with the expressions), and the parameters pointed in formula should be explained in the text.	Thank you for ponting this out. We have revised the text as suggested by the reviewer. The revised text as follows: “ $IPA_C = \frac{\alpha_0 - \alpha}{\alpha_0} \times 100$ (1) $DIPE_Y = \frac{\beta \times 2}{\alpha_0 - \alpha} \times 100$ (2) $DIPE_S = \frac{\beta}{\alpha_0} \times 100$ (3)

<table>
<tr><td></td><td></td><td>Where α_0 and α are denoted as isopropanol initial and final moles, respectively, whereas β is denoted as diisopropyl ether product mole. IPA_c, $DIPE_Y$, and $DIPE_s$ are denoted as isopropanol conversion, DIPE yield, and selectivity, respectively.”</td></tr>
<tr><td>3</td><td>Section 3.2: In relation to similar works done, it is better that the reffernced works be presented in a Table, and type of the catalyst, process conditions, selectivity and yield be reported in the Table.</td><td>Thank you for pointing this out. The similar works have been presented in a Table.

The revised text as follows:

“The previous study regarding the dehydration of isopropanol with various catalyst and reaction process are presented in Table 3.

Table 3. Comparison of the previous study on isopropanol dehydration with various catalyst and reaction processes

<table>
<tr><th>Catalyst</th><th>Reaction process</th><th>DIPE selectivity</th><th>DIPE yield</th><th>IPA conversion</th><th>Refs.</th></tr>
<tr><td>SiO_2-ZrO_2 (20-30 mol %)</td><td>T= 180-210 ˚C</td><td>5-13%</td><td>-</td><td>10-50%</td><td>[11]</td></tr>
<tr><td>γ-Al_2O_3</td><td>T= 226.85 ˚C</td><td>12%</td><td>-</td><td>-</td><td>[83]</td></tr>
<tr><td>Fe_3O_4/γ-Al_2O_3</td><td>P= 0.1 MPa, T= 250 ˚C</td><td>55%</td><td>-</td><td>63%</td><td>[13]</td></tr>
<tr><td>ZrO_2</td><td>P= 0.1 MPa, T=250 ˚C</td><td>45%</td><td>-</td><td>4%</td><td>[84]</td></tr>
</table>
</td></tr>
</table>

		NiP-zeolite (8 (mEq/g)	P=150 ˚C t= 3 h catalyst weight= 0.5 g	33%	40%	81.51%	[85]
		ZrP-zeolite (8 mEq/g)	P=150 ˚C t= 3 h catalyst weight= 0.5 g	47.8%	35.81%	66.73%	This study
		"It can be seen that the zirconium phosphate-supported natural zeolite catalysts provided sufficient catalytic activity toward DIPE production compared to the other reports (Table 3). Based on the previous study [85], the metal phosphate loading during the impregnation process significantly enhanced the dehydration of isopropanol to DIPE, in which high loading of up to 8 mEq/g was found to be sufficient to promote the optimum selectivity and yield towards DIPE. At this condition, the metal-phosphate interaction produced a harmonious effect, which was suggested due to the existence of both Lewis and Bronsted acidic sites, although the textural and the morphological surface might affect the catalytic activity but were slightly dominant.					
4	The reference of the zeolite preparing method should be mentioned.	Thank you for pointing this out we have added the reference on the zeolite preparing method.					

		The revised text as follows: "…..Afterward, 100 g of natural zeolite was immersed in a hydrogen flouride solution (1 %), subsequently stirred for 1 hour, and then washed with distilled water 7 times each for 24 h. The natural zeolite was later immersed in a 6 N HCl solution (125 mL) for 4 hours, filtered, and washed using distilled water until the pH was close to neutral [85]."
5	Section 2.4: authors said that the amount of the used catalyst was 0.5 g. What is the base of this selection?	Thank you for pointing this out. In this study, we varied the metal phosphate concentration from 2 to 10 mEq/g. The ratio of catalyst in the reaction was conditioned to no more than 1%. Hence, we employed 0.5 g of catalyst weight for this study.
6	The name of the country of analysis devices should be mentioned.	Thank you for pointing this out. We have added the country of analysis devices, as suggested by the reviewer. The revised text as follows: "The crystal structure and phase of natural and modified zeolite were assessed using the X-ray diffractometer Rigaku MiniFlex 600 (Japan). FTIR Shimadzu-Prestige 21 (Japan) was utilized with the KBr pellet technique for functional group analysis (recorded from 4500 to 500 cm-1). The textural characteristic was evaluated using N2 physisorption at 77.35 K in a Quantachrome instrument (USA). The catalyst was vacuum degassed to 300 ˚C with a heating rate of 10 ˚C/min for 60 min. The multi-point BET method was employed to determine the catalysts' surface area, the pore features were determined by the BJH method, whereas the external surface area and the micropore area were evaluated using the t-plot method. The catalysts' morphology and elemental composition were inspected using a Tescan Vega 3 (Czech Republic) scanning electron microscope (recorded at 5000× magnification with HV of 15 kV) assisted with X-ray energy dispersive spectroscopy (Bruker QUANTAX, US). The

		gravimetric method was employed to evaluate the surface acidity features of catalysts utilizing a pyridine base [34].”
7	Authors referred to researches for dehydration reaction’s catalyst. Some of them are old, such as references of [24, 26, 28,…]. It is better these references are revised. For example, reff. [28] can be substituted with Korean J. Chem. Eng., 28(7), 1593-1598 (2011).	Thank you for your valuable comment. We have added the corresponding reference, as suggested by the reviewer.
8	Table 2: “Surface area (m2/g)” must be revised to Specific surface area (m2/g)	Thank you for pointing this out. We have revised the word “surface area” to “specific surface area (S_{BET})”.
9	The conclusion and the Abstract are the same. They must be revised.	We have revised the abstract and omitted the explanation regarding the results specifically because it has already been mentioned in the conclusion. The revised text as follows: “In this work, diisopropyl ether (DIPE) was produced through catalytic dehydration of isopropanol over zirconium phosphate modified natural zeolite. The catalyst was prepared via the wet impregnation method. They were tested at 150 ˚C for 3 hours under a reflux system. The effect of zeolite-Zr(H2PO4)4 metal loading and zeolite-Zr without phosphate incorporation on dehydration isopropanol was also assessed. The results showed the natural zeolite was successfully modified as confirmed by XRD, FTIR, SEM-EDX, N2 physisorption, and catalyst acidity by the gravimetric technique. The highest isopropanol conversion (66.73%) was accomplished by 8 mEq/g zeolite-Zr(H2PO4)4 followed by the DIPE yield and selectivity up to 35.81% and 47.8%, respectively. Further reusability investigation showed that zeolite-Zr(H2PO4)4

		catalyst provided adequate reusability up to the fourth reused with relatively decreased catalytic activity towards isopropanol dehydration.”
10	Fig. 4: the important peaks must be assigned in the Fig., with the related wave number.	Thank you for pointing this out. We have assigned the corresponding important peaks on the FTIR spectrum.

Hasanudin Hasanudin <hasanudin@mipa.unsri.ac.id>

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