

Rebuttals on the comments made on our manuscript entitled “Comparative catalytic performance study of 12-tungstophosphoric heteropoly acid supported on mesoporous supports for biodiesel production from unrefined green seed canola oil”

(Manuscript ID: catalysts-1676411)

The authors thank the Reviewers for excellent comments on the manuscript. The itemized response to each comment is given below and the manuscript is modified accordingly. These modifications helped to improve the quality of the manuscript. The discussion added to the manuscript are based on the Reviewers' comments and are highlighted in the rebuttals below.

Reviewer Comments:

Reviewer 3,

The authors significantly improved their manuscript but still some improvements are needed. Please find my comments below.

1. The authors should provide the quantification of Pyridine-DRIFTS results and include these results in Table 3 with additional columns such as BAS, LAS, LAS/BAS. To present just spectra in Fig. 3 is not enough. Also, these results should be discussed and compared with NH₃-TPD results. How the authors can explain so big discrepancy between Pyridine-DRIFTS and NH₃-TPD analyses for the HPW/Al₂O₃ and HPW/MAS catalysts?

Our response: Regarding the acidic properties, the total acidic quantity ($\mu\text{mol/g}$) of the catalysts were determined using NH₃-TPD analysis. The Pyridine-FTIR was used to indicate the presence of both types of acidic sites (Bronsted and Lewis), however, it was a qualitative analysis in our case as we do not have access to the quantitative analysis equipment to determine the amount of Bronsted and Lewis acidity. Similar procedures were used by Hoo et al. [34], and Sudhakar et al. [35] to determine and discuss catalysts' acidity.

[34] P. Y. Hoo and A. Z. Abdullah, “Direct synthesis of mesoporous 12-tungstophosphoric acid SBA-15 catalyst for selective esterification of glycerol and lauric acid to monolaurate,” *Chem. Eng. J.*, vol. 250, pp. 274–287, 2014, doi: 10.1016/j.cej.2014.04.016.

[35] P. Sudhakar and A. Pandurangan, “Heteropolyacid (H₃PW₁₂O₄₀)-impregnated mesoporous KIT-6 catalyst for green synthesis of bio-diesel using transesterification of non-edible neem oil,” *Mater. Renew. Sustain. Energy*, vol. 8, no. 4, pp. 1–11, 2019, doi: 10.1007/s40243-019-0160-1.

2. Please include more details about the Pyridine-DRIFTS analysis to the 2.6. Catalyst characterization techniques.

Our response: We thank the Reviewer for this comment. Based on the recommendation, more details about Pyridine-FTIR are added to the revised manuscript (please see lines 158-161 on page 4) as follows:

HPW supported catalysts (around 100 mg) were first immersed in pyridine for 6 hours. Then, the samples were heated at 100 °C for 2 hours and the pyridine chemisorbed catalysts were analyzed to determine the types of acidic sites (Bronsted and Lewis) of the prepared catalysts.

3. The authors said: “Larger average pore size of HPW/MAP compared to that of MAP could be due to the occupation or blockage of the mesopores of the support with HPW anions”. But this statement is against the logic. How the average pore size can INCREASE after occupation or blockage of the mesopores of the support? Why the authors did not observe the same effect for the HPW/Al₂O₃ and HPW/MAS catalysts? Maybe the authors just have experimental errors for the HPW/MAP catalyst? According to the reference that the authors provided, there was mentioned about “the development of abnormalities in the pores”. It can be another explanation.

Our response: The BET analyses including the determination of average pore diameters for MAP and HPW/MAP were repeated and showed approximately the same values. Based on the recommendation, the explanation in the revised paper is modified (please see lines 187-189 on page 5), which is given below:

Larger average pore size of HPW/MAP compared to that of MAP could be due to the occupation or blockage of the microporous of the support, which led to an increase in average pore size of HPW/MAP.