Response to Decision Letter

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Title: Improved catalytic propylene epoxidation for extruded micrometer TS-1: introducing meso and macro pores insides the crystals

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Reviewer(s)' Comments to Author:

Reviewer: 2

Question 1: TS-1 was first reported by Taramasso et al. in 1983 (US Patent 4410501). The epoxidation of propene to produce propene oxide was widely studied and commercialized (See the review of F. Cavani et al., ChemSusChem, 2009, 2, 508). Why did the authors use 5 pages of 11 to describe the synthesis, characterization, and textural properties of TS-1 and HPTS-1? This could be added in the supporting information. Table 2 (catalytic performance) and references could be enough.

Our response: Thanks for your good advice. The synthesis, characterization, and textural properties of TS-1 and HPTS-1 have been supplemented in the Supporting Information in the revised manuscript.

Question 2: Selection of the binder.

The role of the inorganic binders is to provide the extrudate with satisfactory physical strength. However, binders this component may interfere with the catalytic properties of the final extruded particles since additional species and phases are introduced to the composition of the final catalytic system. Consequently, a study of the non-efficient decomposition of hydrogen peroxide could be helpful. Why silica sol, sesbania powder was used? Why did the authors add starch and PVA? No characterization of

the products is given by the authors, no commercial references, no information (MW of PVA, % of hydrolysis, …)

Our response: Thanks for your professional advice. The blank experiment has been carried out according to the formula of E-HPTS-1-8 with the exception of HPTS-1 and the results are supplemented in the Supporting Information. The non-efficient decomposition of hydrogen peroxide is 5.5%, so binders have little effect on the catalytic properties of extruded catalysts.

Table S3 The blank experiment results in propylene epoxidation

$X(H_2O_2)$	Y(PO)	S(PO)	$U(H_2O_2)$
5.5%	2.2%	89.6%	45.3%

In the extrusion molding, the silica sol acts as an adhesive. Starch is a pore-making agent. Sesbania powder and PVA have certain viscosity and can play the role of adhesion and lubrication. The type of PVA is 2488, Mw=105600, 88% of hydrolysis. These materials were purchased commercially and their manufacturers are provided in Supporting Information.

Question 3: Catalytic performance

Furthermore, could the authors give the amount in mmol of reactant (Section 4.3). What is the ratio of H_2O_2 /propylene? Did this ratio has an impact on yield and selectivity?

Our response: In the reaction process, propylene was charged at constant pressure (0.70 Mpa). The ratio of propylene / H_2O_2 is 1.95. The effect of $n(C_3H_6)/n(H_2O_2)$ on propylene epoxidation in fixed bed reactor had been investigated and published by our group (Ind. Eng. Chem. Res. 2015, 54, 5416−5426). Although the batch reactor was used to evaluate the propylene epoxidation reaction in the manuscript, the effect trend of $n(C_3H_6)/n(H_2O_2)$) on propylene epoxidation was similar. When $n(C_3H_6)/n(H_2O_2)$) was low, the conversion of H_2O_2 and yield of PO was low. Increasing the ratio of propylene / H_2O_2, the selectivity and yield of PO obviously were improved.

Question 4: Did the authors study the recycling of their catalyst?

Our response: The stability and regeneration properties of E-HPTS-1-8 were both

investigated. The stability of E-HPTS-1-8 was carried out in the fixed bed reactor for 250 h (Figure A). The regenerated E-HPTS-1-8 was carried out in the batch reactor (Table A). The PO yield decreased slightly after E-HPTS-1-8 was run for 250 hours, and the activity of E-HPTS-1-8 was recovered after regeneration.

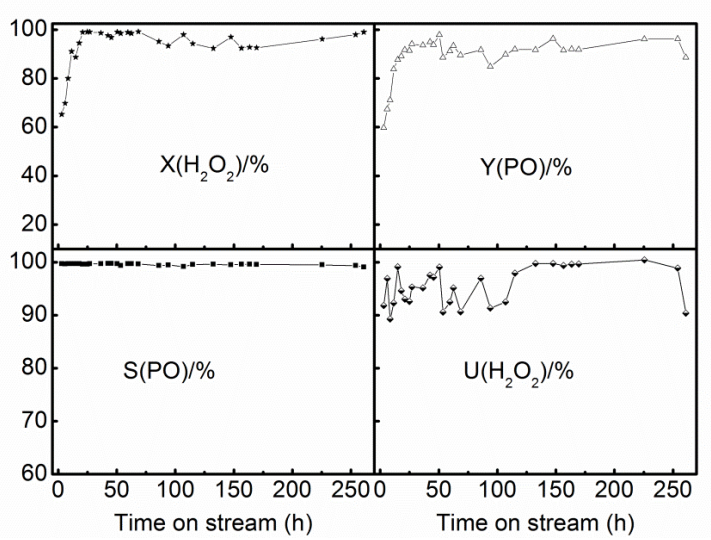

Figure **A** Catalytic performance of E-HPTS-1-8 in fixed-bed reactor

Catalyst concentration: 0.183 g/mL, residence time: 30 min, reaction temperature: 42 °C, the pH of reaction solution: 9.10

Table A Catalytic performance of fresh and regenerated E-HPTS-1-8 in batch reactor

样品	*X*(H_2O_2)/%	*Y*(PO)/%	*S*(PO)/%	*U*(H_2O_2)/%
E-HPTS-1-8	99.14	92.49	98.72	94.51
Re-E-HPTS-1-8	99.60	96.44	98.95	97.86

Question 5: I don't feel qualified to judge the English language and style. However some sentences are not “chemically” correct: line 158, 259 among others

In conclusion, the paper presents interesting results but should be strongly reorganized and the catalytic evaluation should be strengthened.

Our response: Thank you gratefully. Some sentences have been revised in the manuscript in yellow.