Response to Reviewer 2 Comments

Comments and Suggestions for Authors

Auvenientlythors have thoroughly respond to most of the reviewer’s comments. However, some points still need to be con addressed:

**Response:** Thank you for your letter and comments concerning our manuscript entitled “Preparation of Fe₃O₄@polyoxometalates nanocomposites and their efficient adsorption of cationic dyes from aqueous solution” (ID: nanomaterials-471180). Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully and have made correction which we hope meet with approval. Revised portion are marked in red in the paper. The main corrections in the paper and the responds to the reviewer’s comments are as follows:

1) If composites are formed by Fe₃O₄ cores and POM-hybrid shells, the presence of Mo in the particles should be confirmed EDX. I do not see a direct connection between this work and the references given in the response (R1 to R3), except for R2, which is a similar work carried out by the same authors on related POM hybrid-iron oxide composites for the removal of dyes from water. Please add this citation.


It would also be convenient to include the thermogravimetric analyses as supplementary information and a short discussion within the main text.

**Response 1:** Thank you for your question. The presence of Mo in the particles can be found in the XPS spectra of Fe₃O₄@I (Figure 12f). The reference RSC Adv. 2017, 7, 25325–25333 has been cited. The thermogravimetric analyses as supplementary information and a short discussion within the main text have been added as well.

3) The general formula given for elemental analyses should be: C, H and the symbol of the remaining elements following the alphabetic order.

**Response 3:** Thank you for your question. As suggested, the formula for elements analysis has been revised.

4) Crystallographic table: What do the authors mean with “Data”? Are you referring to “reflections”? Which ones: total, unique, observed? Please indicate all of them, and the Rint and goodness of fit (S) parameters as well. The second values given for the R
indices correspond to R2 values. Please correct. Besides, please indicate the origin of the 363 restraints used for the structure of 1.

**Response 4:** Thank you for your pertinent suggestion. ‘Data’ in crystallographic table refers to observed reflections. In the final crystallographic table, the numbers of total, unique, observed reflections, the Rint and goodness of fit (S) parameters are added in Table 1. Additionally, all the related R and wR residual factors have been also supplemented in Table 1. In addition, in order to remove the ADP and NDP alerts of a lot of C and N atoms, the commands of SIMU and ISOR were used in the final refinement, therefore leading to restraints in the refinement of 1.

5) Response: “Valence bond calculations indicated that there is a one proton in \{P_2Mo_5\} of each composite”. Which POM oxygen atom is protonated? it usually belongs to the phosphate group Where is the second proton in 1? Thermogravimetric analysis on compounds 1 and 2 could experimentally confirm the presence of those protons and corroborate the number of hydration water molecules per formula unit in the bulk sample.

**Response 5:** Thank you for your question. In 1, O(4) of \{P_2Mo_5\} is protonated. In 2, O(22) of \{P_2Mo_5\} is protonated. The bond valence sum parameters for O(4) of 1 and O(22) of 2 is 1.376 and 1.082, respectively. The second proton in 1 was determined based on the charge balance consideration [R1].


9) As stated by the authors “However, 1 in Fe_3O_4@1 will decompose at alkaline environment. Therefore, Fe_3O_4@1 becomes non-active above pH 9.” Please add this information in the manuscript.

**Response 9:** Thank you for your pertinent suggestion. The related sentences have been added to the revised manuscript.

10) Please revise the spelling of author’s list in reference 30.

**Response 10:** Thank you for your pertinent suggestion. The spelling of author’s list in reference 30 has been revised.

11) Although it has improved considerably, English still needs to be polished.

**Response 11:** Thank you for your good suggestion. We have tried our best to polish the language in the revised manuscript. These changes have been marked in red in revised paper. We appreciate for reviewers’ warm work earnestly, and hope that the correction will meet with approval.