Dear Editor and Reviewer:

Thank you for your kind letter of “A novel process for preparation of FePO4 2H2O from LiFePO4 mixed with LiNixCoyMn1-x-yO2 waste material” (ID:8640). We revised the manuscript in accordance with the reviewer’s comments, and carefully proof-read the manuscript to minimize typographical, grammatical, and bibliographical errors.

Here below is our description on revision according to the reviewer’s comments.

EXPERIMENTAL PART

*Materials:*

1. The reviewers’ comment: Give data about materials and chemical used in the experiment, but without results. For example, give sentences from lines 52-56 from the first version of the manuscript. Then, say that chemical and phase compositions, as well as morphology were determined, but do not give results here, but in RESULTS AND DISCUSSION. Also, give some information about chemicals used in the experiment (HCl, H2O2…).

The authors’ Answer: We re-typeset the part based on the suggestion of reviewer, and additionally added some information about chemicals used in the experiment, including HCl, H2O2, EDTA-2Na, NaOH, NH4H2PO4 and NH3 H2O (Lines 75- 87).

1. The reviewers’ comment: It is necessary to explain in the experimental part the leaching experiments (leaching with HCl and H2O2)! Table II from RESULTS AND DISCUSSION should be given in Experimental Apparatus and Procedure. It should be said: “The waste was leached with HCl and H2O2, and the influence of temperature, ratio liquid to solid, reaction time and dosage of HCl and H2O2 on the leaching efficiency, was investigated. Experimental conditions are given in Table II. The leaching efficiency was calculated according Equation:

Leaching efficiency, % = (left side of equation 1 – line 114)

“According to the experiments, the optimal leaching conditions were determined.”

Dosage of HCl and H2O2 should be given per mass of waste material that is leached, not as “mole ratio …times that of FePO4 2H2O”!

The authors’ Answer: According to the reviewer’s comments, the description of the leaching experiment was put into the experimental part, the following part was added according to the reviewer's suggestions: “T The mixed waste was leached with HCl and H2O2, and influence of temperature (30-70 oC), liquid/solid ratio (2-4), reaction time (1-3 h), HCl/raw material ratio (0.98-2.28 ml g-1) and H2O2/raw material ratio (0-0.4 ml g-1) on leaching efficiency was investigated (TABLE I) (Lines 95-97)”. The Table II in the results and discussion was transferred to the experimental apparatus and procedure (Lines 98-102).

For the expression of dosage of HCl and H2O2, we modified “mole ratio of hydrochloric acid 3.6 times that of FePO4 2H2O, mole ratio of hydrogen peroxide 0.75 times that of FePO4.2H2O” to “HCl/raw material ratio and H2O2/raw material ratio (Line 173)” and the axis labels in Fig. 5c and Fig. 5d have corrected (Lines 165-171).

1. The reviewers’ comment: Next, state that leached solution obtained at optimal leaching conditions was diluted to obtain 1M Fe concentration and the resulting solution was used for FePO4 2H2O precipitation, lines 81-83 in new version of the manuscript (give name for the solution, which should be further used, for example “diluted leached solution). Then, give text from lines 83-91 and add explanation that influence of temperature, final pH, mole ratio Fe/P in solution and complexing agent on the characteristics (Fe/P and content of Al, Ni, Co, Mn in the product, precipitation rate) of FePO4 2H2O was determined. Give important details, for example temperature was varied from 55 to 95 ºC, …… (in the same way for other parameters). Give here the equation for precipitation rate.

The authors’ Answer: Based on the reviewer's suggestions, the sentence was modified as“The solution obtained at optimal leaching conditions was diluted to obtain 1 M [Fe] and the resulting solution (diluted leached solution) used for FePO42H2O precipitation (Lines 103-104)”.

Herein, we further define the name of the leached solution obtained at optimal leaching conditions was diluted to obtain 1M Fe concentration, called “diluted leached solution” (Line 104). At the end of this paragraph, a detailed description of the experimental conditions was added during the precipitation process, as follows: “Effects of temperature (55–95 oC), final pH (1.5–2.5), Fe/P mole ratio in solution (from 1:1 to 1:1.65) and complexing agent (0–40 g L-1) on the characteristics (Fe/P ratio, Al, Ni, Co, Mn content in the product and precipitation rate) of FePO4 2H2O was determined (TABLE II)” (Lines 113-120).

The experimental conditions of the precipitation process were summarized in the form of a table (Line 116). In addition, the equation for precipitation rate is also given here (Line 118).

1. The reviewers’ comment: Remove Fig.2 (flow diagram) from the manuscript because it isn’t informative enough and all the information will be given in the text.

The authors’ Answer: Fig. 2 (flow diagram) was removed in the revised manuscript according to the suggestion of reviewer.

*Analytical and testing methods:*

1. The reviewers’ comment: In first sentence say that raw material (not materials, because the mixture was analysed) and products were analysed and remove the sentence from 109-111. Similarly, for XRD and SEM. It is stated that “If there was still insoluble matter after the above treatment, it was filtered and then analysed.” In that case, there will be an error, because the entire sample was not analysed.

The authors’ Answer: The separate descriptions of raw materials and products were combined, and redundant expressions were removed (Line 123). The sentences “If there was still insoluble matter after the above treatment, it was filtered and then analysed” which could cause misunderstanding was deleted.

RESULTS AND DISCUSSION PART

1. The reviewers’ comment: Remove all parts that will be given in Experimental part. For example, start Characterisation and Processing of raw materials from line 127. In Fig. 3 b, the granulometric analysis data are given, but it was not mentioned in Experimental part. All experimental procedures should be given in Experimental part. The results should be discussed in a better way (What it means: the particle sizes (D50) of the raw material were uniform…). It is not necessary to give all D values in the Figure.

The authors’ Answer: All parts that given in EXPERIMENAL part were removed from the RESULTS AND DISCUSSION. The granulometric analysis method was added in the “analytical and testing methods” of the EXPERIMENTAL part as: “Laser particle size analyser (LPSA, Mastersizer 3000) was used to determine granularity (Lines 133-134)”. For the graph of the particle size distribution, the content of the D value was removed (Line 144, Fig 2b).

The sentence "the particle sizes (D50) of the raw material were uniform" is ambiguous actually. It is incorrect to describe D50 here because D50 represents the median diameter instead of the average particle size. Therefore, the “(D50)” was deleted in the revised manuscript. In addition, it can be seen from Fig. 2b that the consistency of the raw materials is superior.

1. The reviewers’ comment: Avoid repetitions! For example, remove sentence from lines 143-144 (Fig 3b and Fig. 4 show…), because it was given previously.

The authors’ Answer: The sentence “Fig. 3b and Fig. 4 show the granularity and SEM images for LiFePO4 / LiNixCoyMn1-x-yO2 waste materials” has been removed from the revised manuscript based on the reviewer’s comments, at the same time, other parts of the manuscript were examined and similar duplicates was modified.

1. The reviewers’ comment: Preparation of solution: Give and discuss results first, then conclude what the conditions were optimal.

The authors’ Answer: The section has been re-typed based on reviewer’s comments (Line163-183).

1. The reviewers’ comment: Preparation of FePO4 2H2O: The information from 182-186, 205-208, 223-228, 249-254, 278-283 and 306-311 should be given in the experimental procedure, in a systematic way. DO NOT REPEAT! Process parameters which are constant can be given in figures title.

The authors’ Answer: For the contents of 182-186, 205-208, 223-228, 249-254 in previous version have been given in the form of table (TABLE II) in the EXPERIMENTAL part (Line116). The process parameters which are constant of the contents of 278-283 and 306-311 were given in figures title (Lines 259-260, 283-284).

1. The reviewers’ comment: Avoid “after a comprehensive series of experiments”!

The authors’ Answer: The sentence of “after a comprehensive series of experiments” was removed in the revised manuscript.

1. The reviewers’ comment: From the Fig. 13, it is not possible to see that “increases in feeding speed lead to decreases in the size of primary particles of FePO4 2H2O, from 100–250 nm at a flow rate of 2–7 ml/min to 50–150 nm at a flow rate of 12–22 ml/min, while the particle size stabilised at 2–4 μm (D50).” The biggest particles are the particles obtained with 17 ml/min and particles obtained with 22 ml/min are smaller. D50 was given. Is the particle size distribution was determined.

The reviewers’ comment: For the figures here, we have modified it to use a higher multiple of the SEM picture, and then, the primary particles in the figure were marked to clearly discern the changes of the primary particles (Lines 279-282). The “(D50)” here was deleted in the revised manuscript.

1. The reviewers’ comment: It cannot be said that the product obtained after drying at 80 ºC “is highly” crystalline (Line 327, fig 14a).

The authors’ Answer: The inappropriate expression of the previous manuscript has been removed in the revised manuscript.

CONCLUSION PART

1. The reviewers’ comment: Remove the sentence “The resulting leached solution was diluted to a 1 M Fe concentration and added to a fabricated baffled 1 L beaker for precipitation experiments.” The concentration of the solution used for precipitation can be added in the following sentence. The part of sentence “The results showed that the optimum conditions for precipitation were….” Replace with “The optimum conditions for the precipitation from 1 M Fe leached solution were….”

The authors’ Answer: We have made correction according to the Reviewer’s comments and the corrected content was as follows: “The optimum conditions for the precipitation from 1 M Fe leached solution were a pH of 2.0, a temperature of 85 oC, and a EDTA-2Na concentration of 20 g L-1” (Lines 309-310).

1. The reviewers’ comment: The conclusion from the last sentence (… the particle size was controlled at 1–5 μm (D50)) is not in accordance with the results from Fig 13 c.

The authors’ Answer: In order to see the particle sizes of product more clearly, we added Fig 12f, and marked the particle size of some particles in red in Figure 12f (Line 282).

SOME OTHER COMMENTS

1. The reviewers’ comment: Check the equation 4! What was oxidized in this reaction with H2O2?

The authors’ Answer: The Ni, Co, and Mn in LiNixCoyMn1-x-yO2 are considered to be trivalent, and H2O2 acts as a reducing agent in this reaction equation, so that trivalent metal ions are reduced to divalent ions.

1. The reviewers’ comment: Authors should be cited in the text in the following way: Li et al., not just Li.

The authors’ Answer: The format of the citation authors appearing in the introduction has been modified. (Line 40, 46, 47, 51).

1. The reviewers’ comment: Check the references cited in the text; for example:
“Kandori prepared a number of ultrafine phosphates with good particle size uniformity and high purity by homogeneous coprecipitation.20 “ There is no Kandori in ref. 20: H. Li, S. Z. Xing, Y. Liu, F. J. Li, H. Guo, G. Kuang, Acs Sustain. Chem. Eng. 5 (2017) 8017 (https://doi.org/10.1021/acssuschemeng.7b01594)

The authors’ Answer: We are so sorry that some errors and omissions in the reference due to our negligence, we have carefully checked the reference and corrected the reference number. The changed literature is as follows: “20 H. Tanaka, A. Yasukawa, K. Kandori, T. Ishikawa, *Colloid Surface. A* **204** (2002) 251 ([https://doi.org/10.1016/S0927-7757(02)00005-5](https://doi.org/10.1016/S0927-7757%2802%2900005-5)) and 21 N. K. Mal, A. Bhaumik, M. Matsukata, M Fujiwara. *Ind. Eng. Chem. Res*. 45 (2006),7748 (<https://doi.org/10.1021/ie060609u>)” (Lines 356-359)

1. The reviewers’ comment: Line 89: What it means: “for 4 hLin”?

The authors’ Answer: We’re really sorry for a typo here and it should be “for 4 h”, it was corrected in the revised manuscript (Line 111).

Many grammatical or typographical errors have been revised.

All the lines and pages indicated above are in the revised manuscript.

Thank you and reviewer for the kind advice.

We tried our best to improve the manuscript and made some changes in the manuscript and marked in red in revised paper.

We appreciate for Editor/Reviewers’ warm work earnestly, and hope that the correction will meet with approval.

Once again, thank you very much for your comment and suggestions.

Sincerely yours,

Xi Dai