Dear Editor and Reviewer:

Thank you for your letter and for the reviewers’ comments concerning our manuscript entitled “A novel process for preparation of FePO4 2H2O from LiFePO4 mixed with LiNixCoyMn1-x-yO2 waste material” (ID:8640). Those comments are all valuable and helpful for revising and improving our paper, as well as the important guiding significance to our research. We have studied comments carefully and revised the manuscript in accordance with the reviewers’ comments, and carefully proof-read the manuscript to minimize typographical, grammatical, and bibliographical errors. Here below is our description on revision according to the reviewers’ comments.

1. The reviewers’ comment: In the introduction, raw materials contained two catodic materials, LiFePO4 and LiNixCoyMn1-x-yO2, not LiFePO4 waste material containing Ni, Co, Mn and Al (lines 45-46), the expression was wrong and inconsistent. It should be clearly stated in the Introduction, where just LiFePO4 was mentioned as catodic material (Lines 33-34) and in the introduction only LiFePO4 is mentioned as the material of the positive electrode

The authors’ Answer: We rewrote the introduction based on the reviewer's suggestion, and correcting the incorrect expression of “LiFePO4 waste material containing Ni, Co, Mn and Al”. The two cathodic materials of LiFePO4 and LiNixCoyMn1-x-yO2 were introduced in detail (Lines37-44).

1. The reviewers’ comment: The typical recycling technologies for LiFePO4 waste material (and also LiNixCoyMn1-x-yO2) should be much better presented.

The authors’ Answer: We have added the typical recycling technology of LiFePO4 and LiNixCoyMn1-x-yO2 in the introduction (Lines 37-44).

1. The reviewers’ comment: What is echelon use?

The authors’ Answer: The application of a lithium battery in the original product could not meet the power and energy requirements, found another way to continue to use it to realize the full value, which called echelon use. Duke Energy cooperated with ITOCHU Corporation to conduct a series of tests on waste batteries that could not be used as automobile power batteries because their capacity was less than 80% of original, and they analysed the service data to determine possible fields of application for the products. We added the above description to the revised manuscript (Lines 38-41).

1. The reviewers’ comment: The principles of regeneration, lithium recovery and material preparation cycle should be explained! It is stated that “Different material preparation cycle processes of LiFePO4 were carried out to prepare FePO4 2H2O and Li2CO3 for the synthesis of LiFePO4 materials” (lines 41-42). Give some information!

The authors’ Answer: Regeneration was a method that analyzed the content of each component, added materials in a certain proportion, and processed by a certain method to directly regenerate into a new battery material. Li added Li2CO3 to the waste materials and the resultant mixture was regenerated at 650 oC, after which the discharge specific capacity and coulombic efficiency were 140.4 mA h/g and 95.32% after 100 weeks of cycling, respectively; this would meet the requirements for lithium-ion batteries. We added the above description to the revised manuscript (Lines 41-44).

One of the methods of material preparation cycle of LiFePO4 meant that fail LiFePO4 materials recovered in the form of Li2CO3 and FePO4 2H2O, and the new LiFePO4 was re-synthesized directly using the recovered Li2CO3 and FePO4 2H2O. Zhou did the corresponding research (<https://doi.org/10.3969/j.issn.1007-7545.2019.04.017>). Since the document was not included in the revised manuscript, it was not reflected in the manuscript, the DOI of the document was given above, which could be viewed.

1. The reviewers’ comment: It is necessary to give some information about the proposed method for the preparation of FePO4 2H2O.

The authors’ Answer: Many methods for preparing FePO42H2O have been reported, including coprecipitation, hydrothermal, spray drying, homogeneous precipitation, and oxidation-liquid phase precipitation. Kandori prepared a number of ultrafine phosphates with good particle size uniformity and high purity by homogeneous coprecipitation. Mal used ferric chloride and sodium phenyl phosphate as the iron source and phosphorus source and sodium dodecyl sulphate as the surfactant, and successfully prepared composite medium FePO4. We added the above description to the revised manuscript (Lines 45-50).

1. The reviewers’ comment: The authors should explain what is new in the proposed process (a novel process for preparation of FePO4 2H2O….)

The authors’ Answer: There are a few reports on the preparation of FePO42H2O from waste LiFePO4; Bian prepared an FePO42H2O precursor by a heated crystallization and solvothermal method after dissolving LiFePO4 waste with acid, supplemented with Li2CO3 and glucose, LiFePO4/C cathode material was re-synthesised after heat treatment and resulted in a new material that meets application requirements. The LiFePO4 waste used for research must not contain impurities.

However, impurities of Ni, Co, Mn are often detected in LiFePO4 waste, some of them doped into the material and some present as a mixture of LiNixCoyMn1-x-yO2 and LiFePO4. To our knowledge, there are few papers that report processes for the preparation of FePO42H2O from LiFePO4 / LiNixCoyMn1-x-yO2 waste material.

In this work, we artificially incorporated a small amount of LiNixCoyMn1-x-yO2 waste into LiFePO4 waste to simulate the composition of lithium iron phosphate waste. And the mixed waste was used as a raw material to recover battery-grade FePO4 2H2O, which was identified in accordance with the HG/T 4701-2014 standard. We added the above description to the revised manuscript (Lines 52-73).

1. The reviewers’ comment: At the end of the Introduction, the goals of the investigation should be indicated, not some of the results (By adding EDTA-2Na, the impurities of Ni, Co, Mn, and Al in the FePO4 2H2O were reduced and thereby battery level FePO4 2H2O was obtained).

The authors’ Answer: We have re-written this part according to the Reviewer’s suggestion:

In this work, we artificially incorporated a small amount of LiNixCoyMn1-x-yO2 waste into LiFePO4 waste to simulate the composition of lithium iron phosphate waste. The purpose was to prepare FePO42H2O from LiFePO4 / LiNixCoyMn1-x-yO2 waste material, and we report the effects of temperature, pH, Fe/P molar ratio, complexing agent, ammonia addition rate, and feeding mode on the purity, composition, morphology and particle size of the resultant battery-grade FePO42H2O, which was identified in accordance with the HG/T 4701-2014 standard (Lines 68-73).

1. The reviewers’ comment: The desirable properties of FePO4 2H2O (particle size, composition, morphology) should be also mentioned in the Introduction.

The authors’ Answer: Due to the many indicators of battery-grade iron phosphate, the addition of the part of indicator would greatly increase the length of the paper. In addition, the corresponding standard was given in the later part of the paper, which would be repeated. We have not insert the contents of the form into the paper, but gave the name of the standard (Line 73). The content of the HG/T4701-2014 industry standard was as following:

TABLE I. Technical requirements for battery-grade iron phosphate

|  |  |
| --- | --- |
| Project | Index |
| Fe, w / % | 29.0~30.0 |
| P, w / % | 16.2~17.2 |
| Fe:P (mole ratio) | 0.97~1.02 |
| Ca, w / % | 0.005 |
| Mg, w / % | 0.005 |
| Na, w / % | 0.01 |
| K, w / % | 0.01 |
| Cu, w / % | 0.005 |
| Zn, w / % | 0.005 |
| Ni, w / % | 0.005 |
| SO42-, w / % | 0.01 |
| Cl-, w / % | 0.01 |
| Moisture, w / % | 19.0~21.0 |
| Tap density / (g cm-3) | 0.7 |
| Granularity(D50) /μm | 2~6 |

If it was necessary to add to the Introduction, please come it up, thank you very much.

1. The reviewers’ comment: At the beginning of Experimental part, the removal of Al was mentioned. It is necessary to explain (in the introduction) the origin of Al in the waste.

The authors’ Answer: An aluminium piece was used to support the cathodic material and was extremely difficult to remove during the recycling process. Therefore, Al was generally contained in the waste catodic materials. We have added this section to the Introduction (Lines 63-65).

1. The reviewers’ comment: Experimental part should be reorganized. All steps should be defined more precisely. Do not give results in the experimental part (Table I, Figs. 1-3), but in the Results and discussion.

The authors’ Answer: Experimental part have reorganized as two part: Experimental Apparatus and Procedure, Analytical and testing method (Line 76, 95). The part of Materials have moved to the later section of Results and Discussion and called:Characterization and Processing of raw materials (Line 120).

1. The reviewers’ comment: Chemical analysis method for the analysis of raw material and the final product should be explained in more details (dissolution of the samples?).

The authors’ Answer: We have included specific methods of chemical analysis (Lines11-105) and the calculations of “Leaching rate” and “Precipitation rate” (Lines 112-117) in this section:

In the analysis of raw materials, chemical analysis and inductively coupled plasma optical emission spectrometry (Thermo Scientific iCAP 7200 ICP-OES) were used to determine the components via the following method: a 0.2000 g sample was dissolved in a small amount of aqua regia, the resulting solution was then cooled and brought to a volume of 250 mL and then diluted to a concentration within the measurable range of the sample curve, and the concentration of the element in question was detected by ICP-OES. If there was still insoluble matter after the above treatment, it was filtered and then analysed. Liquid samples were acidified to pH <2.0 with dilute nitric acid, the solution diluted to within a measurable range, and the corresponding element concentration detected by ICP-OES. X-ray diffraction (XRD, Ultima IV, step: 0.004°, step time: 23.8 s, start: 5.000°, end: 70.008°) and scanning electron microscopy (SEM-EDS, Nova NanoSEM450) were used to determine the mineralogical composition, micromorphology, and distribution of elements. The chemical composition of the products was analysed by inductively coupled plasma optical emission spectrometry and chemical analysis. XRD and chemical analysis were used to confirm the identity of the final product.

The calculation method of leaching rate as shown in Equation 1: (1)

The m1 was the total amount of a certain element in the leached solid after leaching, and m2 was the total amount of this element in the solid before leaching. The m3 was total amount of a certain element remaining in the liquid after precipitation and the m4 was total amount of this element in the liquid before precipitation.

1. The reviewers’ comment: It was not clear what is specific in beaker with a baffle which was used as the reactor. It stated at several places that it is self-made. What was the function of the baffle?

The authors’ Answer: When the material was stirred in the beaker, the stirring paddle rotates at a high speed, and a circumferential flow was generated under the action of centrifugal force, the liquid was sucked around the beaker and rose along with the periphery of the beaker wall, the center liquid naturally dropped, and a large cavity was formed in the beaker, a swirling phenomenon was formed. The swirling material would rotate along the wall of the beaker and the mixing would be uneven, the baffle was present on the peripheral wall with the purpose of circumferential flow into converting radial or axial flow. The spiral concave flow in the longitudinal section of the liquid was blocked by the baffle, forming a vertical shunt in the vertical liquid direction, the liquid was forced to an axial flow and the cavity was disappeared, enhanced the stirring effect, and increased the transfer efficiency. Due to space issues, we have mentioned it in the revised manuscript rather than specifically description (Lines 78-79).

1. The reviewers’ comment: Just the amount of leaching liquid is given, not the composition of the leaching liquid and the quantity of the material that was leached (LiFePO4 with LiNixCoyMn1-x-yO2).

The authors’ Answer: The “leaching solution” was a solution for precipitating FePO4 2H2O, not a solution for leaching raw materials. Therefore, the quantity of the material that was leached (LiFePO4 with LiNixCoyMn1-x-yO2) was not given here, the composition of the “leached solution” was given in the section of Characterization and Processing of raw materials (Lines 129-130). The definition of the leached liquid was not clear that caused ambiguous, and we were very sorry about it.

1. The reviewers’ comment: What was the time of leaching and if completely sample was dissolved?

The authors’ Answer: The precipitation experiment started from the time of the end of the aqueous ammonia was added. It was mentioned in the manuscript (Line85).

1. The reviewers’ comment: What is diluted leaching solution (lines 90 and 91)?

The authors’ Answer: The leaching solution was a solution after leaching which should be called “leached solution”, and the “diluted leaching solution” was the solution that leached solution diluted to obtain 1 M Fe concentration. The definition of the “leached solution” was not clear that caused ambiguous, and we were very sorry about it.

1. The reviewers’ comment: Co-current feeding is not clear. What is the base solution? What was pH of the solution obtained by the sample leaching?

The authors’ Answer: Co-current feeding: the base solution (pure water) was added to the beaker and stirred. Then, the leached solution and the aqueous ammonia were pumped into the beaker simultaneously using a peristaltic pump. The base solution was pure water and the pH was deoended to each experimental condition. The word of “Co-current” have been corrected as “Concurrent” (Line 85, 302, 341).

1. The reviewers’ comment: Remove “Recovery of Al” and “Recovery of Li+/Ni2+/Co2+/Mn2+ from the flow diagram (Fig. 5), because these recoveries were not done in the manuscript.

The authors’ Answer: We have made correction according to the Reviewer’s comments and the corrected flow diagram was as follows (Lines 93-94):



Fig 1. Flow diagram for recycling LiFePO4 waste mixed with LiNixCoyMn1-x-yO2

1. The reviewers’ comment: According to XRD analysis, the presence of LiNixCoyMn1-x-yO2 is confirmed, although the peaks are very weak. Therefore, the authors should explain if Ni, Co and Mn are present in LiFePO4 as impurities (which might be introduced by mixing or other methods?!, lines 72-73) or as constituents of another phase (LiNixCoyMn1-x-yO2).

The authors’ Answer: Considering the complexity of the raw materials in the battery recycling process in the future, LiNixCoyMn1-x-yO2 was added as a separate substance to LiFePO4 waste to explore the presence of Ni, Co and Mn for the subsequent preparation of FePO4 2H2O. The content was also reflected in our revised introduction (Lines 68-70).

1. The reviewers’ comment: The presence of Li3Fe2(PO4)3 is also confirmed by XRD, but it is difficult to detect the presence of a phase according to just one peak (3 in the Fig. 1). What it means: Li3Fe2(PO4)3 was oxidized by LiFePO4 (Lines 74-75).

The authors’ Answer: We are really sorry for our negligence of that only one peak of Li3Fe2(PO4)3 was marked in the XRD pattern, and other peaks were forgotten to be labeled. It has been supplemented in the figure (Line 132) and please check it. The “Li3Fe2(PO4)3 was oxidized by LiFePO4” originally meant to be expressed that Li3Fe2(PO4)3 was a product of oxidation of LiFePO4 and the description was corrected as “The Li3Fe2(PO4)3 resulted from oxidation of LiFePO4” (Line 142).

The corrected flow diagram was as follows:

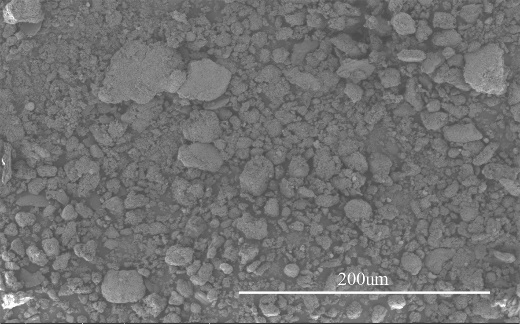
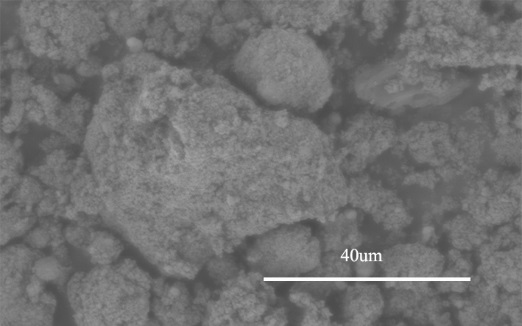


Fig 2. XRD of LiFePO4 mixed with LiNixCoyMn1-x-yO2 waste materials

1. The reviewers’ comment: According to SEM micrograph in Fig. 2a, particles are smaller than 2 μm, while according to Fig. 2b, particles are about 50 μm! Explain! If the particle size distribution was determined, having in mind that D50 is mentioned (line 76)?

The authors’ Answer: The SEM micrograph in Fig. 2a only showed a part of the particle, not the whole one. The surface of the particle was uneven, and it was looked like a particle, the particle in the figure was not less than 2μm actually. We re-submit the new SEM photo (Line 136) and give our results of the particle size test (Line 132), please check it.

**a b**



**c**



Fig 3. SEM images and Granularity of LiFePO4 mixed with LiNixCoyMn1-x-yO2 waste materials: a)5000×; b)1000×; c) Granularity

1. The reviewers’ comment: Explain (in the introduction) the presence of carbon at the surface!

The authors’ Answer: In the production of batteries, acetylene black (as a conductive agent), PVDF (as a binder), and a cathodic material were generally mixed and pulped, while LiFePO4, a poor conductor, was coated with carbon to improve electrical conductivity and low-temperature performance; therefore, it is inevitable that carbon would be present in the cathodic material. It was mentioned in the Introduction (Lines 59-62).

1. The reviewers’ comment: The optimum conditions for the leaching are given at the beginning of the Results and discussion. It is not clear how these conditions were determined? There is no information in the experimental procedure about these experiments! How the leaching efficiency was determined?

The authors’ Answer: The experiments of leaching portion of the raw material was originally written in the initial manuscript. Since the requires of the journal that the length of the article was up to 15 typewritten pages, including Figs, Tables and References, we have removed the detailed description of this part in the paper, only the results were given, however, we have done a lot of research on this part. In the revised manuscript, we have condensed this part of the content as much as possible, and added it to the manuscript (Lines 159-169).   
The content inserted in the manuscript was as follows:

TABLE II. Experimental conditions of the leaching experiment

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Experimental group | *T* / oC | Ratio of liquid to solid | | Dosage of HCl, times | | Dosage of H2O2, times | *t* / h | Variable |
| A | - | | 3:1 | | 3.6 | 0.75 | 1.5 | Temperature |
| B | 50 | | - | | 3.6 | 0.75 | 1.5 | Ratio of liquid to solid |
| C | 50 | | 3:1 | | - | 0.75 | 1.5 | Dosage of HCl |
| D | 50 | | 3:1 | | 3.6 | - | 1.5 | Dosage of H2O2 |
| E | 50 | | 3:1 | | 3.6 | 0.75 | 2.0 | Reaction time |

The calculation method of leaching rate as shown in Equation 1:

 (1)

The m1 was the total amount of a certain element in the leached solid after leaching, and m2 was the total amount of this element in the solid before leaching.

The experimental results are shown in Figure 6.

**a b c**



**d e**



Fig 6. a) Effect of reaction temperature on the leaching rate of the raw materials; b) Effect of the ratio of liquid to solid on the leaching rate of the raw materials; c) Effect of the dosage of HCl on the leaching rate of the raw materials; d) Effect of dosage of H2O2 on the leaching rate of the raw materials; e) Effect of the reaction time on the leaching rate of the raw materials

1. The reviewers’ comment: It should be stated in the experimental procedure that the solution obtained by waste material leaching (do not call it “leaching solution”, because the leaching solution is the solution used for leaching) was diluted to obtain Fe concentration 1 mol/L and so obtained solution was used in the experiments where FePO4 2H2O was precipitated.

The authors’ Answer: We are very sorry for the imprecise wording of “leaching solution”, in the revised manuscript, we call it of “leached solution” Which means the solution after leaching. In addition, we have added each set of experimental conditions of “the solution obtained from waste material leaching was diluted to obtain a 1 M Fe concentration and the resulting solution was added into the experiments where FePO4 2H2O was precipitated” in the revised manuscript (Line 181, 204, 222, 248, 277, 305), please check it.

1. The reviewers’ comment: Crystal lattice integrity? Binding force? The lattice was found to become complete?

The authors’ Answer: We are so sorry for the inaccurate description in the manuscript and have re-written this part in the revised manuscript (Lines 190-197):

As shown in Fig. 7, the crystallinity of the product obtained at 95 oC was higher than that obtained at 55 oC. The 55 oC product was largely amorphous, with virtually no diffraction peaks in the spectrum; presumably, it could adsorb more impurities due to the high activity of the amorphous structure, and the percentages of Ni, Co, Mn, and Al in this FePO4 were high. The FePO42H2O resulting at 95 oC had sharp peak shapes and good crystallinity, and the amount of Ni, Co, Mn, and Al impurities entering the crystal lattice were reduced because of the high crystallinity.

1. The reviewers’ comment: What is “precipitation rate (%)”?

The authors’ Answer: The calculation of precipitation was as follows:  (2)

The m3 was total amount of a certain element remaining in the liquid after precipitation and the m4 was total amount of this element in the liquid before precipitation. The formula was described in the Analytical and testing methods (Lines 112-117).

1. The reviewers’ comment: Line 167: Replacing NH4+?

The authors’ Answer: We are very sorry for our incorrect writing of the word and re-written this part as: “ Increasing the pH with aqueous ammonia did not precipitate Fe and P in a ratio of 1:1, which would have indicated the production of Fe(NH4)x(OH)y(PO4)z.nH2O. Also, increasing pH would precipitate more Fe3+ as Fe(OH)3, resulting in more free H2PO4-/HPO42- and then substituted NH4 or OH in Fe(NH4)x(OH)y(PO4)z.nH2O and thereby reducing slightly the deviation of the Fe/P molar ratio. However, this process is less significant than is the Fe(OH)3 precipitation process, and the molar ratio of Fe/P increased as a result of the above factors.” The word of “replace” was corrected to “substitute” (Line 216).

1. The reviewers’ comment: Line 168: pH control condition?

The authors’ Answer: Considering the Reviewer’s suggestion, the statements of “Therefore, pH 2.00 was chosen as the optimum pH control condition” were corrected as “Therefore, the optimum pH was identified as 2.00” (Lines 219-220).

1. The reviewers’ comment: Line 187: What the equation means? Where from the H3PO4?

The authors’ Answer: Because of the exists form of P element in the solution was H2PO4-/HPO42-, the equation of “Fe(OH)3 + H3PO4 → FePO4 2H2O + H2O” should be corrected as “H++Fe(OH)3 + H2PO4- → FePO4 2H2O + H2O” (Line 238) and the description of the experimental result should be corrected as “From Fig. 9 it can be seen that the molar ratio of Fe to P in the product decreased with increases in the molar ratio of Fe to P in the solution, because the Fe(OH)3 produced in the solution would form FePO42H2O in the presence of excess H2PO4-/HPO42- and the excess H2PO4-/HPO42- would partially substitute the NH4 or OH of the Fe(NH4)x(OH)y(PO4)z.nH2O and further reduce the Fe/P molar ratio.

H++Fe(OH)3 + H2PO4- → FePO4 2H2O + H2O (2)

An increase in the Fe/P ratio led to an increase in the content of Ni, Co, Mn and Al in the product. This is because the increase in the solution concentration of H2PO4-/HPO42- results in precipitation of greater quantities of insoluble phosphate salts of Ni, Co, Mn, and Al, and leads to an increase in the content of Ni, Co, Mn and Al in the product.

After a comprehensive series of studies, an Fe/P molar ratio 1:1.2 was selected as the optimal condition.

H2PO4- / HPO42- + Me2+ + OH- → Me3(PO4)2 ↓ + H2O (Me=Ni, Co, Mn) (3)

H2PO4- / HPO42- + Al3+ + OH- → AlPO4 ↓ + H2O (4)” (Lines 233-246)

1. The reviewers’ comment: Line 240: “easy to cause insufficient sintering” → not sintering, but “solid-state reaction”.

The authors’ Answer: We have made correction according to the Reviewer’s comments. The statements of “Too large particle size was easy to cause insufficient sintering” were corrected as “Too large particle size was easy to cause insufficient solid-state reaction” (Lines 291-292).

1. The reviewers’ comment: Line 241: Give reference and explain “low density of vibration and low specific capacity”

The authors’ Answer: Too small particle size would cause agglomeration tends, and the primary particles first bond to each other to become loose secondary particles, and these secondary particles were stacked in the closest packing way of the hexagons, the tap density was smaller than in the case of dispersed particles. Moreover, particles produced by secondary agglomeration might be further agglomerated into larger particles, resulting in lower tap density. The low tap density would result in a low volumetric capacity. The smaller the tap density of the material, the thicker the material to be coated to reach the corresponding capacity when the battery was coated and fabricated, in a word, it would result in a lower volumetric capacity. It was mentioned in the revised manuscript (Lines 293-298).

1. The reviewers’ comment: Lines 242-246: It is not clear: one-way feeding method and concurrent (co-current?) method!!! Explain!

The authors’ Answer: One-way feeding: the diluted leached solution was added to the 1L beaker and then aqueous ammonia was added gradually into the beaker with heating and stirring to adjust the pH, and the reaction was run for 4 h after the addition of ammonia.

The word of “Co-current” have been corrected as “Concurrent” (Line 85, 302, 341). The base solution (pure water) was added to the beaker and stirred. Then, the leached solution and the aqueous ammonia were pumped into the beaker simultaneously using a peristaltic pump. The pH was determined and the reaction was run for 4 h (Lines 86-88).

1. The reviewers’ comment: The conclusion and abstract should be re-write after the whole manuscript is corrected.

The authors’ Answer: The conclusion and abstract have been re-written in the revised manuscript.

Abstract: A method for preparing battery-grade FePO42H2O from LiFePO4 and LiNixCoyMn1-x-yO2 mixed waste is proposed. The best leaching conditions consisted of a temperature of 50 oC, a liquid-solid ratio of 3:1; a 3.6 mole ratio of HCl / FePO42H2O, a 0.75 mole ratio of H2O2 / FePO42H2O, and a reaction time of 2 h. The solution obtained upon leaching waste material was diluted to a 1 M Fe concentration, added to a 1 L beaker, and temperature, pH, complexing agent, and ammonia addition rate and feeding mode were studied to determine their effects on the precipitation, particle size and morphology of FePO42H2O obtained. It was found that a high precipitation rate of Fe with low percentages of Al, Ni, Co, Mn in the FePO42H2O could be achieved when precipitation was carried out at a temperature of 85oC, with pH 2.0, and with a complexing agent concentration of 20 g/L. Further, it was observed that slow addition of ammonia and a flow feeding method contributed to the production of FePO42H2O with small particle sizes and a flake morphology (Lines 9-21).

Conclusion: Proposed herein is a new method for recovering battery-grade FePO42H2O from LiFePO4 / LiNixCoyMn1-x-yO2 mixed waste. The waste was leached with HCl and H2O2 first, and the optimum leaching occurred at a temperature of 50 oC, a liquid-solid ratio of 3:1, a 3.6 molar ratio of HCl to FePO42H2O, a 0.75 molar ratio of H2O2 to FePO42H2O, and a reaction time of 2 h. 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 results showed that the optimum conditions for precipitation were a pH of 2.0, a temperature of 85 oC, and a EDTA-2Na concentration of 20 g/L. The results also showed that a relatively slow ammonia addition rate of 12 ml/min and the co-current feeding method favoured the formation of FePO42H2O. Under these conditions, the percentages of Ni, Co, Mn, and Al in the obtained products were less than 50 ppm, and the particle size was controlled at 1–5 μm (D50) (Lines 333-343).

Special thanks to your good comments.

Other changes:

1. Many grammatical or typographical errors have been revised.
2. The axis labels in Fig 9a, 9b have corrected (Line 209).
3. We have added or deleted some of the references based on the revised manuscript (References 13, 14, 15, 18, 21, 23-28).

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