

Research Article

Tingting Yan, Shengwen Zhong*, Miaomiao Zhou, Xiaoming Guo, Jingwei Hu, Fangfang Wang, Fantao Zeng, and Sicheng Zuo

High-efficiency method for recycling lithium from spent LiFePO_4 cathode

<https://doi.org/10.1515/ntrev-2020-0119>

received November 23, 2020; accepted December 24, 2020

Abstract: The extraction of Li from the spent LiFePO_4 cathode is enhanced by the selective removal using interactions between HCl and NaClO to dissolve the Li^+ ion while Fe and P are retained in the structure. Several parameters, including the effects of dosage and drop acceleration of HCl and NaClO, reaction time, reaction temperature, and solid–liquid ratio on lithium leaching, were tested. The Total yields of lithium can achieve 97% after extraction process that lithium is extracted from the precipitated mother liquor, using an appropriate extraction agent that is a mixture of P507 and TBP and NF. The method also significantly reduced the use of acid and alkali, and the economic benefit of recycling is improved. Changes in composition, morphology, and structure of the material in the dissolution process are characterized by inductively coupled plasma optical emission spectrometry, scanning electron microscope, X-ray diffraction, particle size distribution instrument, and moisture analysis.

Keywords: LiFePO_4 , Li recovery, battery-grade Li_2CO_3 , extraction

1 Introduction

LiFePO_4 is one of the main cathode materials in rechargeable lithium-ion batteries in China [1]. However, one of the values of LiFePO_4 , compared with other cathode materials such as Li_2CoO_2 [2], is that it does not contain high recovery value elements such as Mn, Ni, and Co [3]. Given this, China will have 9,400 tons of scrap LiFePO_4 batteries by 2021 [4]. The lithium content accounts for about 200 tons, and projections of possible lithium scarcity point to clear environmental and economic benefits from lithium recycling in the future [5].

Currently, there are several methods for treating waste cathode materials [6], including the fire method, the wet method, and a combined method [7]. Many companies used high-temperature method to recycle lithium battery cathode material, such as Toxco Inc. (USA), SONY Corp (Japan), and Umicore (Belgium). These methods are rather inefficient for recovery as mostly lithium is lost in metallurgical slag during refinement and is not recycled [8,9]. These high-temperature processes have high energy consumption and produce waste gas and waste residue pollution and have pushed methods toward wet extraction of lithium in the industry and academic research [10].

Through a direct regeneration process, Li *et al.* [11] obtained high purity cathode material mixture (LiFePO_4 þ acetylene black), anode material mixture (graphite þ acetylene black), and other by-products (shell, Al foil, Cu foil, electrolyte solvent, etc.) from scrapped LiFePO_4 batteries. Subsequently, recycled cathode material mixture without acid leaching is further regenerated with Li_2CO_3 directly. Batteries using recycled LiFePO_4 confronted many problems, such as safety and cycling stability. Based on the concept of zero waste, Dutta *et al.* [12] proposed a complete process for the recycling of LIBs, in which the metals and materials as value added products were obtained. Although the leaching rate of Li^+ ions were 99.9%, the recovery rate of lithium indeed was not high, because the consumption of oxidant H_2O_2 is relatively high, except that the battery grade Li_2CO_3 cannot

* **Corresponding author: Shengwen Zhong**, Jiangxi Key Laboratory of Power Batteries and Materials, School of Materials Science and Engineering, Jiangxi University of Sciences and Technology, Ganzhou, 341000, China, e-mail: zhongshw@126.com

Tingting Yan, Miaomiao Zhou, Jingwei Hu, Fantao Zeng, Sicheng Zuo: Jiangxi Key Laboratory of Power Batteries and Materials, School of Materials Science and Engineering, Jiangxi University of Sciences and Technology, Ganzhou, 341000, China

Xiaoming Guo: Technical Research and Development Department, Ganzhou Huajing Rare Earth New Materials Co. LTD, Ganzhou, 341000, China

Fangfang Wang: College of Chemical Engineering, Fuzhou University, Fuzhou, 350108, China

be obtained. Zhang *et al.* [13] studied the selective recovery of lithium from spent LiFePO_4 batteries. With the help of sodium persulfate ($\text{Na}_2\text{S}_2\text{O}_8$), the LiFePO_4 were oxidized to FePO_4 , and more than 99% of Li in the material can be selectively leached in 20 min at room temperature. But the sodium persulfate ($\text{Na}_2\text{S}_2\text{O}_8$) that has the chemical properties of highly oxidizable and toxic is expensive, and this method is difficult to realize industrial production.

Yang *et al.* [14] also proposed a method to extract lithium from spent LiFePO_4 cathode; the wet extraction of Li from LiFePO_4 cathode typically involves H_2SO_4 and H_2O_2 to dissolve the cathode material, adjusting pH to precipitate Fe^{3+} , filtering the precipitate, and precipitating Li with Na_2CO_3 solution. This approach is marred by relatively low lithium recovery, excessive use of acid and alkali, and high energy consumption with relatively poor Li_2CO_3 purity. Economically, the market value is limited without a more efficient method for the recovery of lithium [15].

The method shown herein is based on the wet process [16]. The refined approach studies the structural change of LiFePO_4 by hydrometallurgy to achieve preferred dissolution of lithium, thereby minimizing the reaction volumes and retaining the solid structure of FePO_4 [17].

Careful control of HCl and NaClO additions establishes a chemical equilibrium, and lithium is efficiently extracted from the structure with a higher yield [18]. This dramatically reduces the consumption of acid and alkali [19,20]. Recycling lithium of the mother liquor by extraction technology reduces energy consumption [21]. The unique reverse sedimentation process prepares battery-grade Li_2CO_3 for the increased economic benefit [22,23]. Keeping in view of stringent environmental regulations, limited natural resources, and energy crisis, adopting recycling will not only protect the environment and pacify the gap between demand and supply but also conserve the natural resources [24,25].

2 Materials and methods

2.1 Materials

The waste batteries used in this experiment were provided by the Lithium Battery Power Research Institute of Jiangxi University of Science and Technology. All batteries were discharged in saturated NaSO_4 solution and

disassembled by hand [26]. The removed positive electrode was washed with DMC (dimethyl carbonate) and was crushed mechanically with a grinder and screened to obtain LiFePO_4 material, with a mass fraction of lithium in the LiFePO_4 positive electrode material of 3.96% [27]. All the chemicals used in the experiment were analytically pure, and the solutions were prepared with ultrapure water.

2.2 Lithium preferential extraction

LiFePO_4 was mixed with water, adding HCl and NaClO slowly over time with heated stirring and careful pH monitoring. Controlled drip rates for HCl and NaClO prevent the production of Cl_2 gas [28,29].

2.3 High-efficiency lithium recovery

2.3.1 Purification

The pH of lithium-containing liquid for a certain volume was adjusted to a range of 9–10 with alkali, stirred at 85°C for 1 h to remove impurities of Fe, Cu, Al, and others in the solution [30]. After filtration, the lithium-containing liquid was purified further by the addition of a sodium salt at a mass ratio of 0.3% and EDTA at a mass ratio of 0.1% as a complexing agent to adjust Ca^{2+} to $60\text{ }\mu\text{g mL}^{-1}$ [31].

2.3.2 Precipitation of lithium

The lithium solution was heated to $70\text{--}80^\circ\text{C}$, and Na_2CO_3 solution was added to the reaction dropwise to produce sediments of Li_2CO_3 [32]. This precipitate was vacuum filtered, washed with high purity water several times, and dried under vacuum to obtain high purity battery grade Li_2CO_3 [33].

2.3.3 Extraction

The solubility of lithium is $1.2\text{--}2.0\text{ g L}^{-1}$ in the precipitated mother liquor and accounts for 20–25% of the total lithium content [34]. Lithium extraction was performed using a mixed extraction agent ($\text{P507:TBP:NF} = 10:2:1$) in this experiment. The residual lithium was less than $50\text{ }\mu\text{g L}^{-1}$ in the final remaining liquid, giving a recovery rate of 98% [35,36].

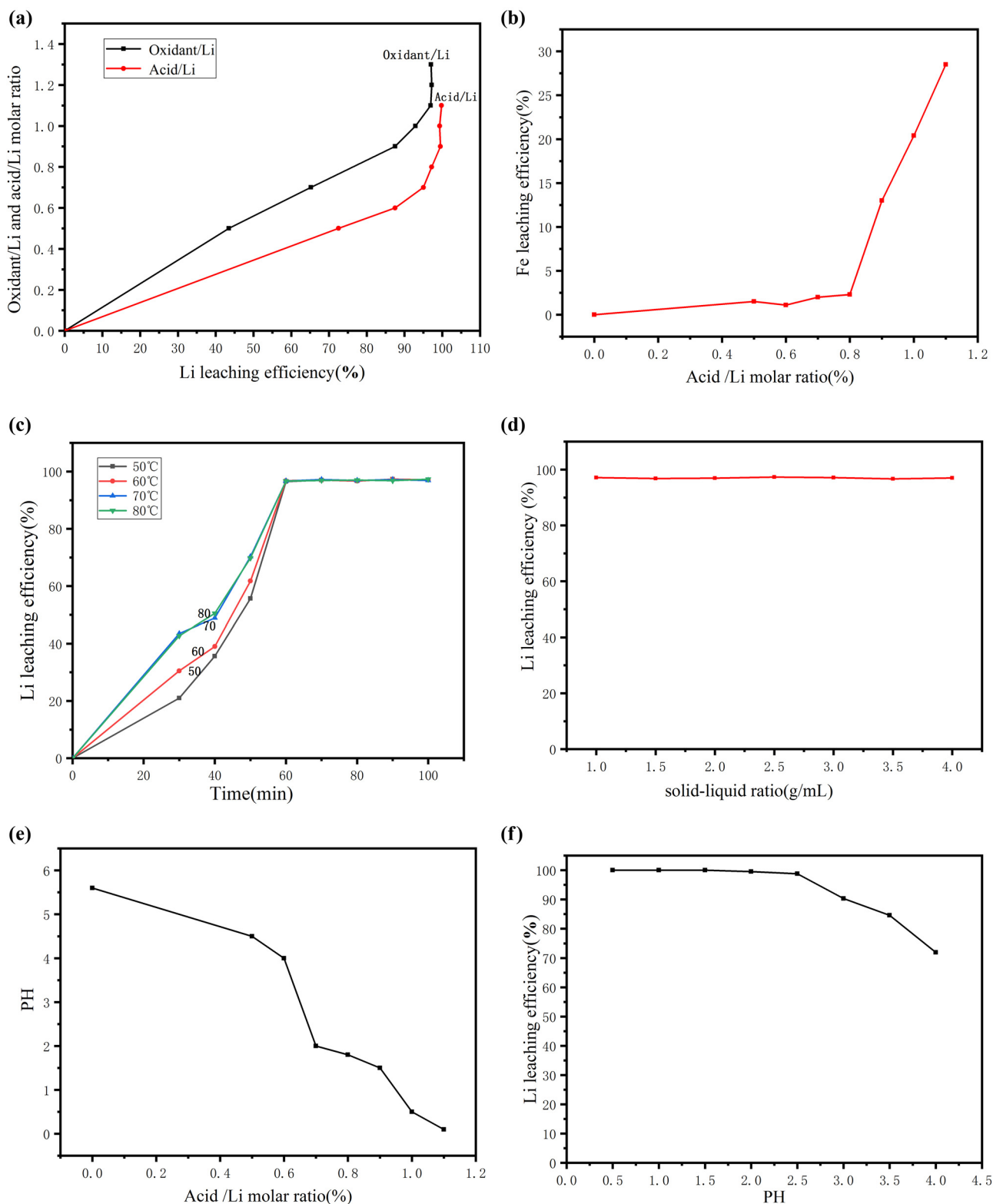


Figure 1: (a) Acid and oxidant molar ratios to Li and the impact on leaching efficiency; (b) HCl : Li ratio effect on unwanted Fe^{3+} leaching; (c) Four (4) temperature condition are plotted against time to measure efficiency showing 60 minutes is optimal for leaching; (d) Leaching efficiency against solid–liquid ratio and shows a minimal effect, (e) Plot of pH against the HCl/Li molar ratio; (f) the relationship between pH of the solution and lithium leaching efficiency.

2.4 Analytical equipment and characterization of materials

The solid samples were digested in concentrated hydrochloric acid for analysis using inductively coupled plasma optical emission spectrometry (ICP-OES, Thermo Fisher ICAP 7400). X-ray diffraction (XRD, D8 Advance, Bruker, Germany) was used to characterize the relative crystal phases during the experimental process and identify any structural changes in the samples in the reaction [37]. Scanning electron microscope (SEM) (Flex SEM 1000, Hitachi, Japan) was used to image the morphology of the products [38,39]. Particle size and the water content of the lithium products were measured by a particle size analyzer and moisture meter to determine if the Li_2CO_3 product meets the standards for cathode material synthesis [40,41].

3 Results and discussion

Figure 1 shows the influence of different parameters on the leaching efficiency. Meaning of each picture: (a) acid and oxidant molar ratios to Li and the impact on leaching efficiency; (b) $\text{HCl}:\text{Li}$ ratio effect on unwanted Fe^{3+} leaching; (c) four temperature conditions are plotted against time to measure efficiency showing 60 min is optimal for leaching; (d) leaching efficiency against solid–liquid ratio and shows a minimal effect; (e) plot of pH against the $\text{HCl}:\text{Li}$ molar ratio; (f) the relationship between pH of the solution and lithium leaching efficiency.

The addition of NaClO oxidizes $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$ and facilitates the extraction of lithium from its structure. From simple stoichiometry, the moles of NaClO should equal the moles of lithium extracted, and the moles of HCl should be less than the moles of lithium if Fe^{3+} remains were intact. The rate of addition for HCl and NaClO should maintain a pH of 1.5–2; otherwise, Fe^{3+} will leach into the solution. Figure 1(a) shows the influence that the amount of NaClO and HCl has on the lithium dissolution rate, and “oxidant/Li and acid/Li molar ratio” expresses the ratio of NaClO and HCl to the molar amount of lithium, respectively. The concentration of Li^+ ion in solution was measured by ICP. The calculated recovery rate of lithium was 99% using excess NaClO . The recovery rate was 100% with excess HCl ; however, unwanted Fe^{3+} also dissolved. Figure 1(b) clearly shows that when the molar ratio of HCl and Li is greater than 0.8 (pH \sim 0.5), the solution rate of Fe^{3+} increases linearly and further introduces impurities during the extraction of lithium. “Fe leaching efficiency” refers to the mass of

the dissolved Fe ion to total iron in LiFePO_4 , again measured in solution by ICP. Figure 1(c) shows the influence of temperature and time on extraction. Exceeding 60 minutes, the temperature has no effect and the leaching effect of lithium is the best. Figure 1(d) shows that with optimal conditions, the size of the solid–liquid ratio has little influence on extraction above a 1:1 ratio. Figure 1(e) shows the influence of pH during the experiment by HCl addition and the effect on lithium yield. It can be seen from Figure 1(f) that when $\text{pH} \leq 2.0$ (the value measured with a pH strip and related to the amount of HCl added), marking a small amount of HCl , the lithium yield is greater than 99% [40].

Figure 2 shows the diffraction patterns phase after the extraction of lithium. LFP refers to LiFePO_4 , LFP-1 refers to the slag that NaClO is used only 70% in the process of extracting lithium, LFP-2 and LFP-3 refers to the slag that the addition amount of NaClO and HCl is optimal. The two reflections (121) and (131) are intense peaks seen in LFP, whereas the leached samples show a significant reduction or absence. The outlined sections A and B of complete patterns correspond to Figure 2(a and b). There are new peaks near the (131) peaks in the XRD pattern of LFP-1 and still retains intensity at (131). However, there are no (131) peaks in the XRD pattern of LFP-2/3.

These patterns indicate that a small amount of LiFePO_4 remains in the LFP-1 but is completely nonexistent in the LFP-2/3 samples. It can be seen that the structure of LiFePO_4 changed in the process of preferential extraction of lithium in the XRD pattern. The main peak (011) and (111) of LiFePO_4 did not change with the dissolution of lithium, indicating that only part of the raw material structure changed in the optimal solution process to realize the preferential extraction of lithium.

Figure 3(a–f) shows the morphology of the LiFePO_4 cathode material and FePO_4 residues. It comprises many primary and secondary particles (Figure 3a), and magnified images in Figure 3(b and c) show spherical agglomerates and much smaller spherical primary particles. Figure 3d shows the particle morphology of post-extraction FePO_4 residues. There is an absence of the previously seen secondary agglomerated particles. In Figure 3(e and f), the primary particles are much smaller irregular shapes with some agglomeration. The extraction caused the secondary particles of LiFePO_4 to break apart, possibly explained by the structural stress as Li^+ ion is extracted and the crystal structure has volumetric changes from $\text{LiFePO}_4 \rightarrow \text{FePO}_4$.

Figure 4 shows the XRD diagrams of battery-grade Li_2CO_3 products; compared with the standard card of Li_2CO_3 , each peak of the two products is perfectly

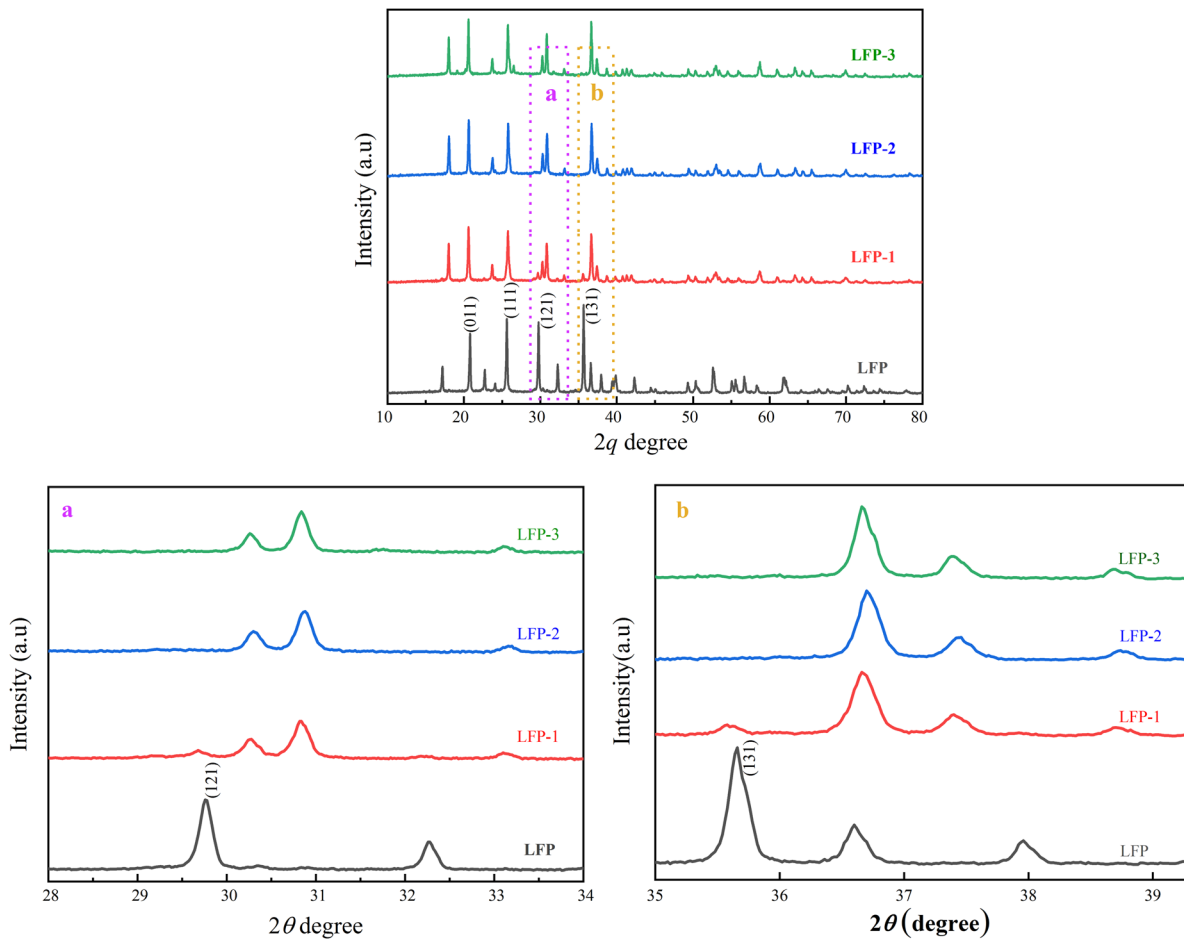


Figure 2: XRD patterns for different NaClO, and the outlined sections A and B of complete patterns correspond to Figure 2a and 2b.

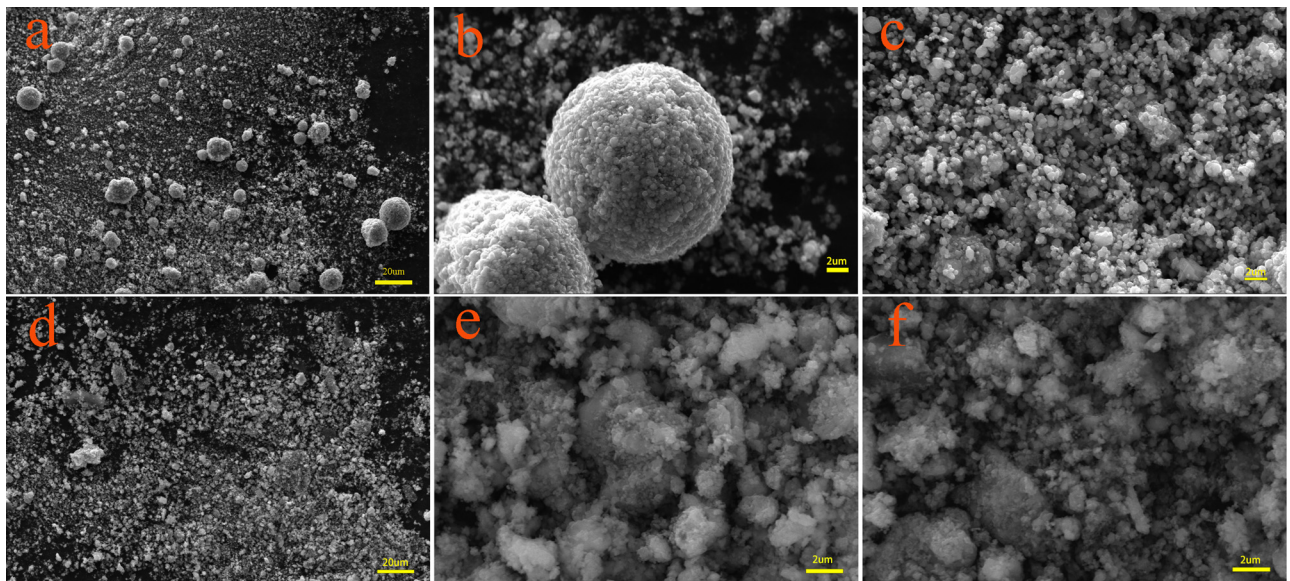


Figure 3: SEM Graph: LiFePO_4 materials, (b) the primary particles of LiFePO_4 materials, (c) the secondary particles of LiFePO_4 materials, (d) the acid slag, (e) the secondary particles of the acid slag are absent, (f) the primary particles of the acid slag broken apart into smaller irregular shapes.

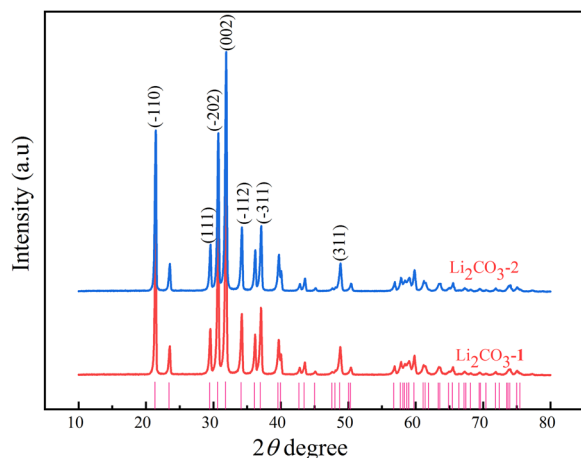


Figure 4: XRD diagrams of battery-grade Li_2CO_3 products.

consistent with the standard peak, indicating that Li_2CO_3 -1 and Li_2CO_3 -2 are both Li_2CO_3 products.

Figure 5 shows the morphology of battery-grade Li_2CO_3 -1 and Li_2CO_3 -2 of Figure 4. The lithium-extracted solution was reacted with Na_2CO_3 after impurity removal. The dropwise addition of the Na_2CO_3 solution permits particle ripening over nucleation to produce larger robust particles. This can be seen from the two batches of battery-grade Li_2CO_3 products in Figure 5 (Li_2CO_3 -1 and Li_2CO_3 -2).

Figure 6 plots the impurity fractions in the Li_2CO_3 products measured by the ICP analysis. Line A represents the impurity standard for battery-grade Li_2CO_3 , and Line B and Line C represent the impurity content of two batches of Li_2CO_3 products respectively.

Table 1 presents the purity, moisture content, and particle size ranges for battery-grade Li_2CO_3 compared with the products from our Li extraction and precipitation methods. The products meet or exceed the standards for Li_2CO_3 in battery material synthesis.

The lithium concentration ranged from 1.2 to 2.0 g/L in the mother liquid, and this concentration accounts for

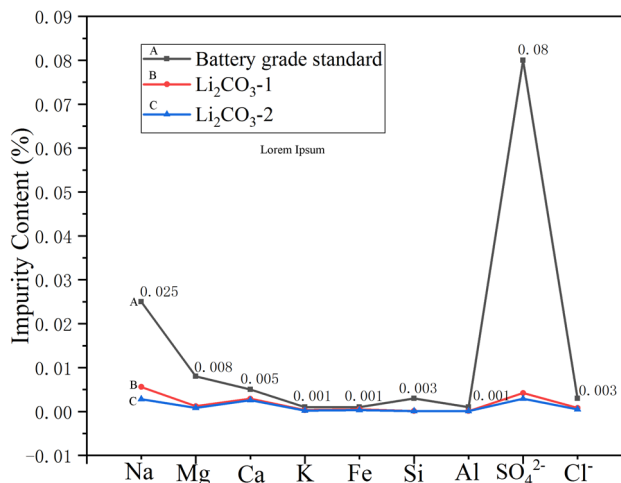


Figure 6: Comparison of the Li_2CO_3 battery-grade standard and product impurity content for several common ionic impurities.

20–25% of the total lithium. The extraction of lithium from the mother liquid was performed using a mixed extraction agent ($\text{P507:TBP:NF} = 10:2:1$). Several mother solutions (numbers 1–6) with different lithium content were selected [41], and the residual lithium after extraction was measured with ICP, as presented in Table 2. The remaining lithium content was less than $50 \mu\text{g/mL}$ in the mother liquor, yielding a 98% recovery [42]. Combining these results with a leaching efficiency near 99% for lithium from LiFePO_4 , the total yields for lithium recycling averaged near 97% [43].

4 Conclusion

A refined method has been tested for lithium extraction from the peridotite structure of LiFePO_4 by controlling the addition of NaClO and HCl . Mole ratios of NaClO and HCl

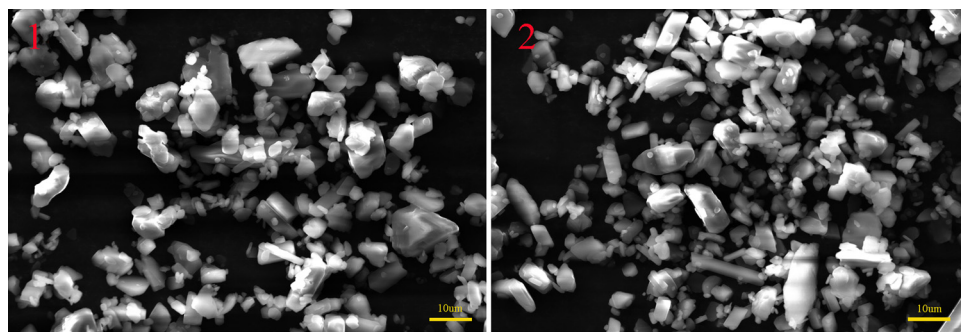


Figure 5: Micrographs of battery-grade Li_2CO_3 -1 and Li_2CO_3 -2 products.

Table 1: Purity, particle size, and water content of the Li_2CO_3 standards and the products

Name	Li_2CO_3 content (%) (no less than)	H_2O (%)	Size distribution (μm)		
			D10	D50	D90
Battery level standard	99.5	0.25	≥ 0.5	3.0–8.0	9.0–15.0
Li_2CO_3 -1	99.89	0.01	2	5.8	12
Li_2CO_3 -2	99.93	0.015	1.5	6.2	11.8

Table 2: The yield of lithium during extraction and lithium total yield

Extraction	Number	Lithium content	Li content in raffinate ($\mu\text{g mL}^{-1}$)	Extraction yield of Li (%)	Total recovery (%)
P507 + TBP + NF	1	1.24	35.5	99.093	98.102
	2	1.31	41.3	98.945	97.956
	3	1.58	49.1	98.619	97.633
	4	1.67	44	98.876	97.887
	5	1.86	24.5	99.374	98.380
	6	1.95	28.7	99.267	98.274

to moles of lithium at 1 and 0.8 produced the best leaching with minimized impurities. Compared with the traditional total solution process, the total acid and alkali needed are reduced to improve the economic and environmental factors in the recycling process. A pH of 1.5–2 gave an optimal dissolution recovery rate (99%) and limited the dissolution of Fe^{3+} to below 2%.

High-quality battery-grade Li_2CO_3 was prepared by extraction using a mixed extraction agent (P507:TBP:NF = 10:2:1) with a recovery that gave a total yield of lithium above 97%. The economic benefits such as the efficiency of energy savings and consumption reduction also increased. This modified method showed an obvious advantage compared to traditional lithium extraction processes. It was helpful to establish recycled lithium as a viable pipeline for future resource and energy demands.

Acknowledgments: The authors would like to thank all the reviewers who participated in the review and MJ Editor for its linguistic assistance during the preparation of this manuscript. This work was supported by the National Natural Science Foundation of China (51874151), the Scientific Research Foundation for universities from the Education Bureau of Jiangxi Province (GJJ170510), the Natural Science Foundation of Jiangxi Province (20151BBE50106), and the Jiangxi University of Science and Technology (NSFJ2014-G13, Jxxjbs12005).

Conflict of interest: The authors declare no conflict of interest regarding the publication of this paper.

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