Environmental friendly approach for lithium extraction from discarded EV batteries.

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Abstract

The demand for EVs has led to an increased urge for LFP battery recycling technique. This study introduces an oxalic acid-hydrogen peroxide system for selective leaching of lithium from scrap EV batteries. By optimizing factors like acid concentration, H_2O_2 volume, pulp density, reaction time, and temperature, a remarkable 99.4% lithium leaching efficiency is achieved. The optimal conditions for include 10% (w/v) oxalic acid, 5% (v/v) H_2O_2, 100 g/L pulp density, at speed of 300 rpm, and 60°C temperature for 4 hours. Advanced characterization techniques such as XRD and SEM confirm the purity and morphology of the recovered material. This pioneering approach enables closed-loop recycling of LFP batteries, supporting a circular economy, environmental friendly and reducing waste management challenges, ultimately having significant implications for the electric vehicle industries.

Keywords: Blackmass, Hydrometallurgy, LFP battery, Selective leaching, Oxalic acid.

1. INTRODUCTION

The widespread adoption of lithium iron phosphate (LFP) batteries in EVs is due to their notable characteristics, including cost-effectiveness, high power output, and extended lifespan (Li et al., 2024). The growing demand for LFP batteries has led to a significant increase in their production, with the global market valued at $17.54 billion in 2023 and projected to reach $48.95 billion by 2031 (Lu et al., 2024). The extensive use of LFP batteries poses substantial economic and environmental concerns and resource conservation. The disposal of scrap LFP batteries can lead to environmental degradation, contaminating groundwater and soil (Alipanah et al., 2021). To mitigate these impacts, recycling has emerged as a vital strategy, enabling the recovery of valuable metals and reducing the economic and environmental costs associated with primary resource extraction. Recovery process includes mechanical processing to recover the black mass. Then black mass is undergone for leaching using various methods, like pyrometallurgy, hydrometallurgy, and bio-hydrometallurgy (Mrozik et al., 2021). Hydrometallurgy is notable for its advantages, including high metal recovery rates, reduced energy consumption, and lower emissions. In hydrometallurgy, strong inorganic acids like HCl, HNO_3, and H_2SO_4 have been used to extract metals from spent LFP batteries (Yadav et al., 2020). However, the adverse environmental impacts of conventional acid-based extraction methods have sparked interest in organic acids as a viable alternative. Organic acids like oxalic acid, tartaric acid, ascorbic acid, citric acid, and malic acid offer a more environmentally friendly solution (Mahandra & Ghahreman, 2021). In this research, a novel approach using an oxalic acid-hydrogen peroxide system has been developed to selectively leach lithium metals. This technique provides an efficient, selective, and eco-friendly solution for recovering valuable metals, characterized by cost-effectiveness and operational simplicity. By developing pioneering recycling technologies and refining existing methodologies, the industry can ensure sustainability and reduce waste management challenges.

2. MATERIALS & METHODOLOGY

2.1. Materials and Reagents

The study utilized 10 kg of spent prismatic-type LFP batteries was collected from local vendor of Pune, India. Commercial-grade oxalic acid ($C_2H_2O_4 \cdot 2H_2O$) and hydrogen peroxide (H_2O_2) were used for leaching studies. Ultrapure de-ionized water was used for sample preparation, dilution and washing.

2.2. Disassembling and Pre-processing of LFP Battery

Spent LFP batteries were safely discharged in a 15% sodium chloride (NaCl) solution to prevent potential hazards (Mishra et al., 2025). The black mass recovered from the electrode materials by thermal treatment and a hot water rinse to remove impurities.

2.3. Leaching of Black Mass Powder

Oxalic acid was chosen for leaching due to its environmentally friendly profile and targeted extraction capabilities. A measured amount of black mass was introduced into a solution of oxalic acid, water, and hydrogen peroxide, maintaining a consistent temperature. The resulting suspension was filtered using Whatman No. 42 filter paper, and the retained residue was washed with de-ionized water and then dried in a oven at 130°C for 24 hours.

The efficiency of metal leaching was determined using the following formula (1):

$$\% \, Leaching\ efficiency = \frac{C_E * V}{C_o * m} * 100 \qquad (1)$$

In the formula, C_E denotes metal concentration in the leachate solution (g/L), V represents the total volume of the leachate (L), C_o signifies initial metal content in the black mass (weight %), and m corresponds to the mass of black mass subjected to leaching (g). The leachate solutions were analyzed using ICP-OES and AAS, while the residues were characterized through comprehensive chemical analysis, SEM and XRD techniques. To ensure accuracy, all experiments were performed in replicate, and the average results were utilized to determine the final metal recovery percentages.

3. RESULT AND DISCUSSION

3.1. Characterization of black mass powder

The black mass powder underwent characterization to determine its elemental composition and structural properties. ICP-OES (Thermo Scientific iCAP 7600 pair instrument) and AAS (Shimadzu AA-7000F spectroscopy) analysis revealed significant amounts of lithium (3.13%), iron (13.7%), phosphorus (7.3%). XRD analysis identified distinct phases, including lithium iron phosphate ($LiFePO_4$) and iron phosphate ($FePO_4$) (figure 2). Scanning electron microscopy showed particles with irregular shapes and sizes, indicating a heterogeneous distribution (figure 1). These findings highlight the complex composition and morphology of the material before leaching. The results demonstrate the material's intricate nature, comprising various elements and phases, which is crucial for optimizing leaching processes and further analysis.

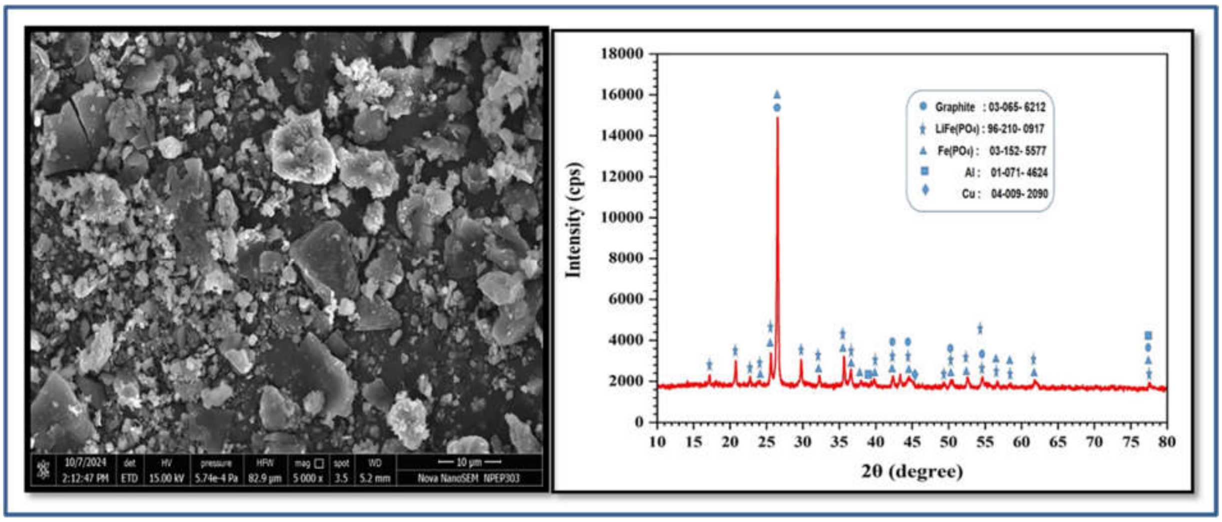

Figure 1: SEM & XRD of spent LFP black mass powder.

3.2. Effect of different leaching parameters

The environmentally friendly and cost-effective properties of oxalic acid made it an ideal choice for selective leaching of lithium. A parametric study was conducted to optimize lithium extraction from scrap LFP black mass, examining the effects of oxalic acid concentration, H_2O_2, pulp density, reaction time, and temperature as shown in figure 2.

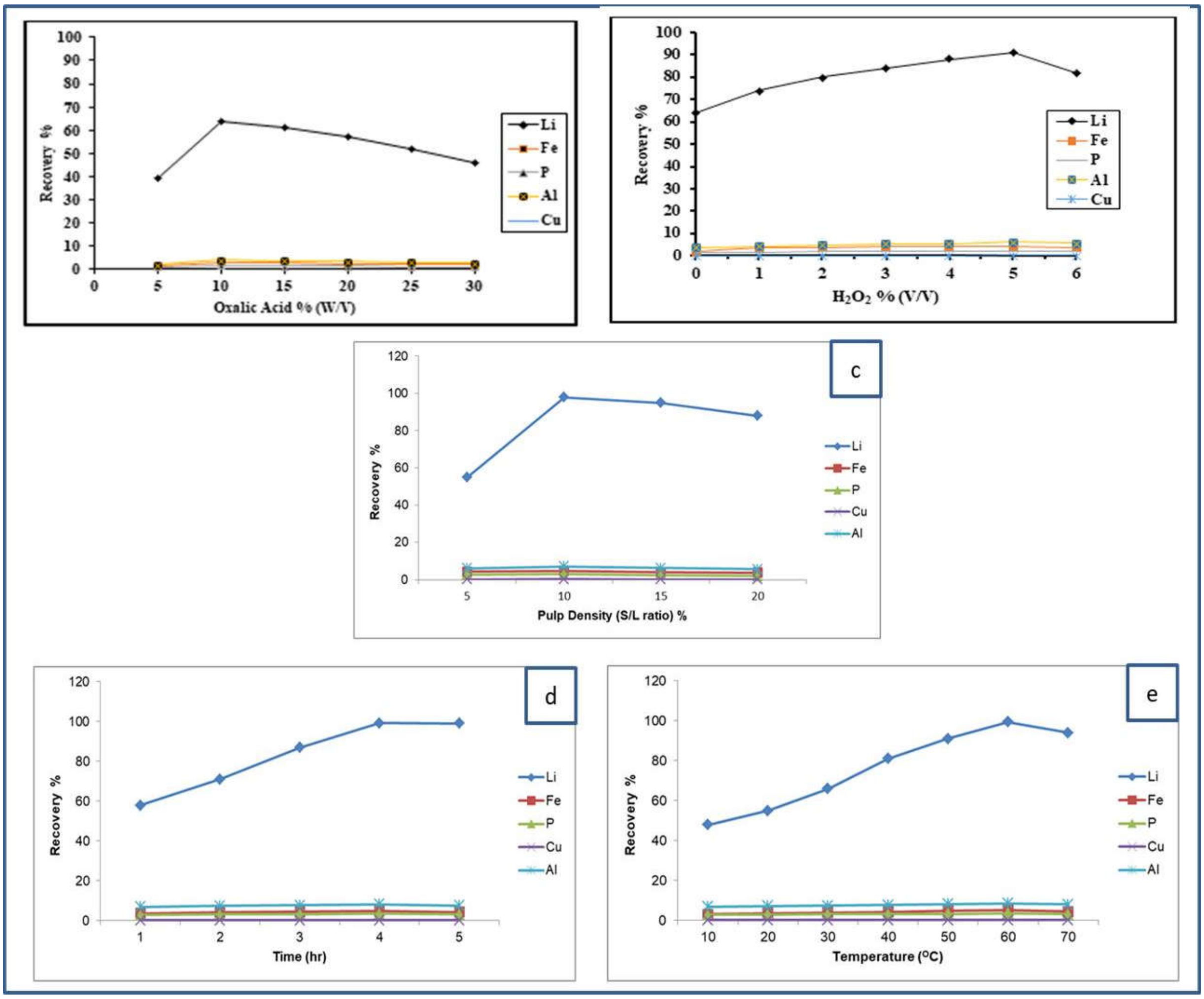

Figure 2: Influence of oxalic acid concentrations(a), H_2O_2 volume(b), pulp density(c), reaction time(d) and temperature(e) on leaching efficiency.

3.2.1. Effect of oxalic acid concentration

The influence of oxalic acid concentration on the leaching efficiencies of various metals was investigated. From Figure 2(a), it is evident that increasing the oxalic acid concentration from 5% to 10% (w/v) improved leaching efficiencies of lithium (from 39% to 66%), iron (from 2.5% to 4.2%) and phosphorus (from 1.5% to 2.5%). The optimal concentration of 10% (w/v) oxalic acid is likely due to the sufficient availability of protons and acid radicals necessary for metal leaching. Further increases beyond 15-30% (w/v) resulted in marginal improvements, possibly due to increased ionic strength hindering further leaching (Li et al., 2018).

3.2.2. Effect of H_2O_2

This section includes the effect of H_2O_2 on leaching efficiency, as shown in Figure 2(b), increasing H_2O_2 concentrations significantly improved leaching efficiencies. The optimal concentration of 5% (v/v) H_2O_2 is likely due to the effective breakdown of organic materials, conversion of Fe^{2+} to Fe^{3+}, and disruption of mineral structures, enhancing metal recovery. Beyond 5% (v/v), excessive oxidation may lead to the formation of insoluble compounds, reducing metal recovery (Mahandra & Ghahreman, 2021).

3.2.3. Effect of pulp density (S/L ratio)

The levels of oxalic acid in solutions tend to be greater when pulp density is lower. As evident from Figure 2(c), a pulp density of 10% emerged as the optimal level, likely due to the optimal balance between oxalic acid availability and metal ion concentration. Higher pulp densities may result in decreased leaching efficiencies due to increased pH and reduced metal solubility (Zheng et al., 2016).

3.2.4. Effect of reaction time

The effect of reaction time on the leaching efficiencies of various metals was investigated. As shown in Figure 2(d), longer reaction times significantly boosted leaching efficiency, with 4 hours being the optimal duration. This is likely due to the sufficient time allowed for reactants to diffuse into the black mass and products to emerge from the solid matrix (Luo et al., 2021).

3.2.5. Effect of temperature

The impact of temperature on the leaching depicted in Figure 2(e), the study revealed that 60°C is the optimal temperature, likely due to the enhanced molecular energy and diffusion rates promoting metal recovery. Temperatures above 60°C may lead to decreased efficiency due to reductant instability and accelerated H_2O_2 decomposition (Mishra et al., 2025). The optimal conditions for maximizing leaching efficiency were determined to be 10% (w/v) oxalic acid, 5% (v/v) H_2O_2, 10% pulp density, 4 hours reaction time, and 60°C temperature.

3.3.Characterization of residue after leaching.

After leaching, the slurry was filtered, and the residue was rinsed and dried. Analysis showed the leach liquor was rich in lithium, while the residue mainly consisted of iron (13.02%) and phosphate (7.03%). SEM analysis revealed significant morphological transformations, with particles forming aggregates with prismatic structures (figure 4). XRD analysis (figure 3) identified phases of $FePO_4$ and $FeC_2O_4{\cdot}2H_2O$, indicating significant dissolution of $LiFePO_4$. This analysis shows the leaching process effectively separated lithium from the black mass powder, resulting in a residue with distinct compositional characteristics. The residue's primary constituents were $FePO_4$ and $FeC_2O_4{\cdot}2H_2O$.

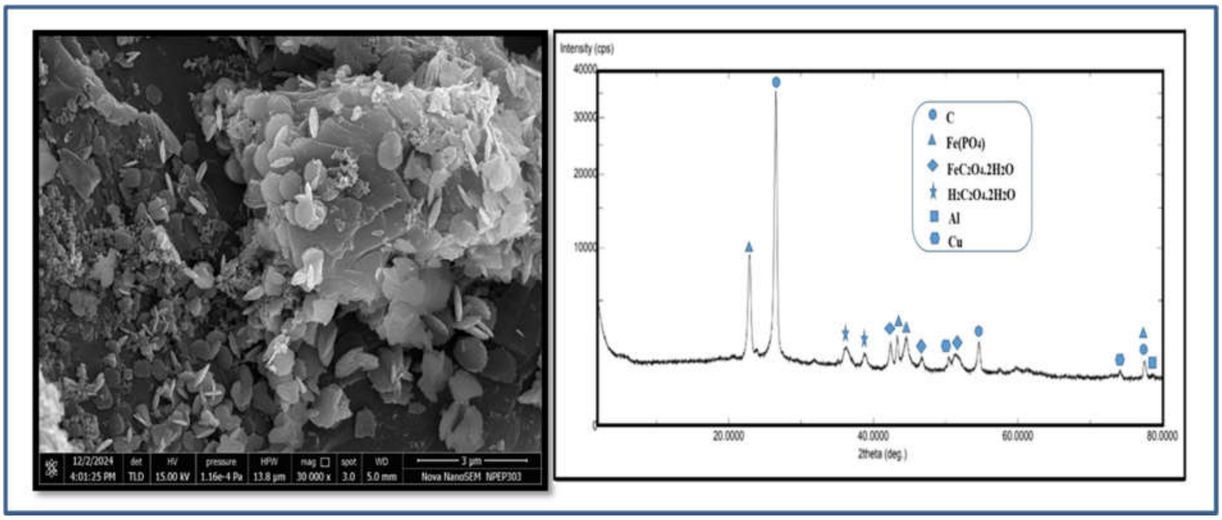

Figure 3: SEM & XRD of black mass powder after leaching.

CONCLUSION

This study presents a pioneering eco-friendly approach to extract lithium from spent lithium-iron phosphate (LiFePO4) batteries. Optimal leaching conditions were established, including 10% (w/v) oxalic acid combined with 5% (v/v) H_2O_2, a pulp density of 100 g/L, at speed of 300 rpm, and temperature of 60°C for 4 hours. Under these conditions, the oxalic acid-hydrogen peroxide leaching system achieved a remarkable lithium leaching efficiency of 99.4%, with minimal co-leaching of other metals. The leaching residues primarily consisted of $FeC_2O_4{\cdot}2H_2O$, a valuable precursor for $LiFePO_4$ synthesis. Characterization results using ICP-OES and XRD confirmed the high purity and morphology of the recovered lithium. ICP-OES analysis detected the presence of various elements in the black mass, while XRD analysis revealed the composition of the leaching residues. This process promotes a circular economy, reduces environmental impacts, and supports sustainable energy storage, alleviating waste management challenges and paving way for eco-friendly future.

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