## Thermal treatment of lithium concentrated fraction from end-of-life LiFePO<sub>4</sub> automotive batteries as a prior process for lithium recovery

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## Abstract

As the demand for electric vehicles grows, effective solutions for recycling end-of-life lithium-ion batteries become crucial. Given that lithium iron phosphate (LFP) batteries represent a significant portion of the automotive battery market, this research presents an innovative method to produce concentrated lithium solutions combining a thermal treatment with a microwave-assisted hydrometallurgical process. Initial steps involve safe collection and disassembly of discarded batteries to preserve components and minimize contamination. Cathode coils were separated and ground to a particle size smaller than 0.25 mm, concentrating 96% of lithium compounds. Afterwards, thermal treatment, with 1h plateau temperature from  $300^{\circ}$ C to  $900^{\circ}$ C were performed to induce the phase transformation of LiFePO<sub>4</sub> to Li<sub>2</sub>Fe<sub>3</sub>(PO<sub>4</sub>)<sub>3</sub>, and it was observed that above 600°C this phenomenon occurs entirely. Different sulfuric acid concentrations (1,5; 1,0; 0,5; 0,25 and 0,1 mol/L) were performed in microwave assisted leaching process for all thermal treated and the non-calcined cathodic powders. Considering leaching with aqua regia as a reference for complete leaching of metals, the best results obtained in terms of lithium selectivity were found with samples calcined at 600°C degrees and leached with 0.25mol/L sulfuric acid. Under these conditions, 60% of all lithium, 0.23% of all iron, 17% of all phosphorus, 2.2% of all copper and 1.33% of all aluminum were detected in solution and if considering only the metal extraction under these conditions, 95.3% lithium and just 0.37% copper, 1.47% aluminum, and 2.93% iron were obtained, showing high selectivity in the developed process.

Keywords: hydrometallurgy, thermal treatment, microwave leaching, selectivity, lithium extraction.

#### Introduction

In recent years, the demand for lithium-ion batteries (LIBs) has experienced remarkable growth, driven primarily by the increasing adoption of electric vehicles (EVs), renewable energy storage systems, and portable electronic devices [1-3]. Currently, lithium iron phosphate (LFP) and lithium nickel cobalt manganese oxide (NCM) batteries stand as the prevailing choices among power LIBs for electric vehicles. Life cycle assessment results have shown favorable performance for NCM batteries, but with a short service life. As the performance of LFP batteries, mainly in terms of charging and discharging efficiency, improves, these batteries will emerge as an environmentally interesting option. Added to this fact, if the recovery processes of the strategic materials that make up such batteries are widely developed and optimized, there will be a more promising scenario for the consolidation of this type of energy solution for global mobility [4-5]. This surge in demand has prompted significant advancements in lithium battery technologies, with particular focus on lithium iron phosphate (LFP) and lithium nickel manganese cobalt oxide (NMC) chemistries [6]. Despite the dominance of NMC batteries in current production figures, there is a growing recognition of the advantages offered by LFP batteries, particularly in certain applications. LFP batteries stand out due to their superior thermal stability, enhanced safety profile, and longer lifespan compared to NMC counterparts. These attributes make LFP batteries highly suitable for applications demanding high safety standards, such as energy storage systems for grid stabilization and residential installations, where thermal runaway events pose significant risks [7-8].

On the other hand, as the demand for high-performance rechargeable batteries increases, so does the amount of electronic waste generated, since batteries, despite being rechargeable, have an estimated lifespan. Considering that resources or raw materials are finite, increasingly scarce, and have a greater environmental impact in their extraction, it becomes necessary to adopt an economic model that opposes the traditional linear model of extract, manufacture, use, and dispose. In this sense, the circular economy emerges, as a modern economic model that aims to keep resources in use for as long as possible, extracting maximum value during use, and subsequently recovering and regenerating products and materials at the end of each life cycle This model emphasizes the reduction, reuse, recycling, and manufacturing of materials, aiming to minimize waste, decrease the consumption of new resources, and reduce the ecological footprint [9].

The circular economy applied to automotive battery recycling emerges as an essential and strategic pillar for sustainability in the modern era of low environmental impact transition to electric vehicles. By recycling batteries, not only valuable metals like lithium, cobalt, and nickel are recovered, reducing dependence on conventional mining, and associated environmental degradation, but also the reuse of these materials in new batteries or new manufactured products is promoted, thus closing the product lifecycle flow. This process not only contributes to reducing pollution and resource depletion but also fosters technological innovation and the creation of local industries dedicated to recycling and manufacturing components [10].

In pursuing sustainable solutions for the burgeoning challenge of end-of-life electric vehicle batteries, this research explores a novel approach for the recovery of lithium compounds combining thermal treatment and microwave assisted leaching process for concentration of lithium solutions scrap fraction derived from spent LiFePO<sub>4</sub> automotive batteries. This study starts with the collection of end-of-life LFP automotive batteries which were discarded for safety manipulation. The cathodic material, after being separated, was beneficiated through grinding, and sieving to concentrate powdered fraction rich in lithium. The resulting material underwent several thermal treatments in air atmosphere and was subsequently subjected to a microwave-assisted hydrometallurgical process with sulfuric acid as the leaching agent for selective lithium extraction. The results reveal a strong influence of the calcination temperature of the powders and the concentration of the leaching agent used in the selective extraction of lithium, to the detriment of iron extraction, which is the main competitor in this process of generating lithium-rich solutions.

## Materials and methods

For the development of this research work, unusable batteries containing cylindrical cells, model size 32650, with a capacity of 5500 mAh, nominal voltage of 3.2 V, and composed of lithium iron phosphate (LFP), were used. This material was generously provided by the company Global, headquartered in Porto Alegre - RS. Prior to the beneficiation and concentration process of lithium-based compounds, the cells were disassembled from the packs that make up a battery and completely discharged by stacking a group of 13 interconnected cells in series along with a wire resistor for 24 hours for complete discharge of the cells. Subsequently, the cells were opened using a metal cutting disc attached to a rotary tool with the aim of removing the metal casing without destroying the internal components. This entire process took place within a fume hood to prevent inhalation of volatile compounds present in the electrolyte.

Afterward, the cathode strips or coils were taken for mechanical processing in a knife mill (Retsch, model SM 300), operating at 1500 rpm with sieves opening of 1.00 mm in the first grinding and with the sieve of 0.75 mm in the second pass of the material in the grinding process. Some details such as the appearance of the cathode coils, the knife mill used, and the appearance of the ground material can be seen in detail in Figure 1 below.



Figure 1. Cathodic material (coils) separated by manual processing (a), knife mil process (b) and ground cathodic material (c).

After knife mill process, the ground material was subjected to size separation using a set of sieves with openings of 1.00 mm (#16 mesh), 0.5 mm (#32 mesh), 0.25 mm (#60 mesh), and 0.053 mm (#270 mesh) on a vibrating table with constant oscillation frequency for 30 minutes, and the obtained fractions were weighed. Chemical analysis of the various size fractions was conducted using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) on an Agilent Technologies instrument, model 5110, to identify the lithium concentration in each sample. For this analysis, the samples were leached in aqua regia at a solid/liquid ratio of 30 g/L using the Anton Paar Multiwave 5000 microwave system with a heating rate of 15°C/min and a plateau temperature of 230°C for 15 minutes. After leaching, the samples were filtered and diluted 100 times for ICP-OES analysis. This analysis was performed in duplicate, and the average lithium concentration values are presented in Figure 2, which also illustrates the visual appearance of the size fractions.



Figure 2 – Lithium mass content in the granulometric fractions obtained after grinding and sieving process and their respective mass balance. In detail, below the graph, the visual aspect of the obtained granulometric fractions.

To effectively analyze where the highest concentration or content of lithium is accumulated in the produced size fractions, chemical analysis by ICP-OES serves as the primary analytical tool. Allied with the mass balance obtained, both presented in Figure 2, it was found that the coarser fractions, i.e., > 1.00 mm and > 0.5 mm, show low lithium contents, representing approximately 10% of the total mass of an LFP cell. Concurrently, the finer fractions, namely, the > 0.25 mm, > 0.053 mm, and < 0.053 mm fractions, exhibit a significant lithium mass concentration. Considering that from the 13 cells used in this study, 609.33 g of cathode material was generated, and the overall lithium composition represents 4.57% by mass, we have 27.84 g of lithium in this material, with 26.66 g of lithium in the > 0.25 mm, > 0.053 mm, > 0.053 mm, and < 0.053 mm fractions. This means that these finer size fractions concentrate approximately 96% of all the lithium present. These findings suggest that the knife milling process is efficient and that the sieving process can be optimized by reducing the number of sieves to just one, the 0.5 mm mesh sieve, and that all material passing through this sieve will have a significant concentration of lithium-rich compounds for subsequent recovery. Based on these results, the size fractions smaller than 0.5 mm were mixed, and this material served as the base raw material for the continuation of this work, hereafter referred to as NCC (non-calcined cathode).

The new material produced (NCC) underwent elemental chemical characterization by ICP-OES following the same methodology previously described to determine the concentration of other elements of interest present: Iron, Phosphorus, Aluminum, Copper, and Lithium. Subsequently, this material underwent qualitative analysis of the crystalline phases present by X-ray diffraction performed on the Aeris model diffractometer from Panalytical (with Cu-K $\alpha$  radiation source with  $\lambda = 1.54$ Å) at room temperature. For this analysis, the cathode material was sieved through a 0.045 mm (#325 mesh) mesh, and the powder passing through this mesh was characterized. To assist and guide the sequence of thermal treatments that were carried out, the starting material (NCC) was then characterized by thermogravimetric analysis on the TGA Q50 TA Instruments equipment, with a ramp of 20°C/min from room temperature to 940°C, in an alumina crucible with air atmosphere before performing the proposed thermal treatment. Subsequently, 7 thermal treatments were proposed at temperatures of 300°C (C300), 400°C (C400), 500°C (C500), 600°C (C600), 700°C (C700), 800°C (C800), and 900°C (C900), in a muffle furnace with a heating rate of 10°C/min for 1 hour at the plateau temperature. For all temperature ranges, 15 g of sample were used and calcined in an alumina crucible. The samples were weighed before and after the thermal treatment to evaluate the mass loss as a function of temperature. All calcined samples underwent elemental chemical characterization by ICP-OES following the same methodology previously described to determine the contents of the main elements detectable by this analytical technique, namely, Iron, Phosphorus, Aluminum, Copper, and Lithium.

After the thermal treatment process, the calcined samples and the NCC material were subjected to a hydrometallurgical microwave-assisted process for selective leaching of lithium aiming to evaluate the influence of thermal treatments and leaching agent concentration on the removal of a higher lithium concentration at the expense of other present elements. For this study, sulfuric acid was employed as the leaching agent in a range of concentrations ranging from 0.1 mol/L, 0.25 mol/L, 0.5 mol/L, 1.0 mol/L to 1.5 mol/L, always using a solid/liquid ratio of 150 g/L. This process was conducted in a polytetrafluoroethylene pressure vessel assisted by the Anton Paar Multiwave 5000 microwave model with a heating rate of 5.5°C/min and a plateau temperature of 110°C for 15 minutes. After the leaching process, the samples were filtered, and the filtrate was subjected to elemental chemical analysis by ICP-OES using the same equipment described previously. All samples analyzed by ICP-OES underwent dilutions to comply with the equipment's detection limits and the calibration curves performed.

## **Results and discussion**

The elemental chemical characterization of the beneficiated cathode material sample (NCC), presented in Figure 3, reveals that the lithium content in the starting sample is around 4% by mass, while the iron content is nearly 8 times higher, exceeding 31% by mass, and the mass content of phosphorus is close to 18%. The presence of aluminum, derived from the substrate where the cathode material is

deposited, was observed, and this element is present at around 3% by mass, and a small amount of copper, which is the substrate of the anodic component of the cells, was also detected and is in the range of 0.3% by mass.



Non Calcined Cathode (NCC)

Figure 3. Elemental chemical composition of the metals of interest in the raw material base of this study, NCC. Leaching conducted in aqua regia at a S/L ratio of 30 g/L.

The chemical composition of the crystalline phase of the starting material is presented in Fig. 4 from the X-ray diffractogram, which reveals the predominant presence of LiFePO4 consistent with the crystallographic card ICDD 01-081-1173. Additionally, a discreet presence of graphite (ICDD 03-065-6212) was observed, possibly originating from the anodic component of the cells, resulting from the manual opening and separation process of the cathode material.



Figure 4. X ray diffractogram of non calcined cathode (NCC) powder.

Regarding the thermal degradation of the cathodic material NCC, the thermogravimetric analysis in Fig 5 reveals two main decomposition events of the material. The first event occurs in the range of approximately 100 °C to 350 °C, with a decomposition peak at 127 °C associated with the elimination of the remaining electrolyte in the starting material, as well as possible moisture presence, consistent with literature results [11-12]. The second thermal event occurs between 350 °C and 750 °C, where a stabilization

of mass loss is observed. In this range, the major thermal decomposition events are seen at 469 °C, and 585 °C, attributed to PVDF primary and secondary degradation process, respectively [13-14].



Figure 5. Thermogravimetric analysis of lithium concentrated fraction from end-of-life LiFePO4 cathode material (CNC).

The phase transformation occurring in the cathodic material due to thermal treatments is illustrated in the X-ray diffraction patterns presented in Fig. 6, with detailed visual aspects of the NCC and calcined powders at different proposed temperatures shown in the right side of each diffractogram. It can be observed that the thermal treatment at 300 °C does not alter the phase composition of the starting material (NCC), where the presence of LiFePO<sub>4</sub> and graphite was detected. When the temperature is increased to 400 °C, the beginning of phase transformation from LiFePO<sub>4</sub> (ICDD 01-081-1173) to Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> (ICDD 01-080-1515) can be observed, along with the presence of other crystalline phases such as iron hydroxide (ICDD 00-017-0536) and iron oxide (ICDD 01-089-0596). From 500 °C onwards, the formation of the new lithium iron phosphate phase (Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>) becomes evident, and the presence of the original phase of this compound (LiFePO<sub>4</sub>) is no longer identified, corroborating the findings of [15] who verified the formation of this phase above 500 °C. At this temperature, the presence of the same phase of  $Fe_2O_3$  (ICDD 01-089-0596) and the formation of lithium metaphosphate - LiPO<sub>3</sub> (COD 96-210-7073) are still identified. At 600 °C, the phases present are the same as those at 500 °C; however, the peaks related to the Fe<sub>2</sub>O<sub>3</sub> phase are identified with greater intensity, indicating that the temperature increase favors the formation of this compound. Indeed, at temperatures of 700 °C, 800 °C, and 900 °C, an intensification of the peaks related to the crystallographic planes of  $Fe_2O_3$  and a decrease in the intensity of the peaks referring to the Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> phase are observed, and these are the only two crystalline phases detected in this temperature range. The increase in the reddish coloration of the calcined powders corroborates with the findings of the crystalline phases encountered, indicating that the temperature increase favors the formation of Fe<sub>2</sub>O<sub>3</sub> due to an intense oxidation process. Phase transformation from LiFePO<sub>4</sub> to Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> results in the change of the orthorhombic crystal structure to trigonal, accompanied by atomic rearrangements of Li, Fe, O, and P. The crystal structure of LiFePO<sub>4</sub> features a unit cell with 1 lithium atom, while the crystal structure of Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> has a unit cell with 2 lithium atoms. Additionally, there is a change in one of the oxygen sites in these structures. In LiFePO4, the O2- site is bonded in a 4-coordinate geometry to one Li1+, two equivalent  $Fe^{2+}$ , and one  $P^{5+}$  atom. In the  $Li_3Fe_2(PO_4)_3$  structure, the  $O^{2-}$  site is bonded to two equivalent  $Li^{1+}$ , one  $Fe^{3+}$ , and one P<sup>5+</sup> atom, forming distorted corner-sharing tetrahedra. This structural change, with a higher availability of lithium atoms, added to the formation of Fe<sub>2</sub>O<sub>3</sub>, which is favored with increasing temperature, it may enhance the selective extraction of lithium with low acid concentrations.



1 - LiFePO<sub>4</sub> (ICDD 01-081-1173) ; 2 - Graphite (ICDD 03-065-6212) ; 3 - Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> (ICDD 01-080-1515) ;

Figure 6. X ray diffractograms of non-calcined cathodic material and calcined in different temperatures. In detail, on the right side of each diffractogram, the visual appearance of the powders obtained and analyzed.

The set of column charts presented in Fig 7 reveals the influence of sulfuric acid concentration on the selective extraction of lithium compared to complete metal leaching using aqua regia for all ranges of thermal treatment temperatures conducted. With aqua regia (Fig 7f), it can be observed that the mass values of Li, Fe, P, Al, and Cu remain constant for all calcination temperatures ranges of the cathodic material. On average, it was found that leaching with aqua regia extracts in mass: 4.3% of Li, 33.8% of Fe, 19.3% of P, 3% of Al, and 0.4% of Cu. Thus, it can be assumed that, under these experimental conditions, there is a complete extraction of all the elements present and that the thermal treatment and the consequent phase transformations do not interfere with the full extraction of the chemical elements evaluated.

However, when analyzing the results with sulfuric acid at the proposed different concentrations, a significant influence of the leaching agent concentration ( $H_2SO_4$ ), as well as the proposed thermal treatment for the selective leaching of lithium, is evident. It can be observed that high concentrations of sulfuric acid, such as 1.5 mol/L (Fig 7e), can extract between 3.5% to 4.2% by mass of lithium, however, with considerable values of iron present, in the range of 25% to 8% by mass of iron. Under these conditions, it is observed that thermal treatment favors the reduction in iron extraction, however, the acidic medium is still very aggressive and extracts, or solubilizes, a significant amount of iron, unfavorably affecting lithium selectivity. A similar phenomenon occurs for leaching with 1.0 mol/L sulfuric acid (Fig 7d), however, with the sample calcined at 700°C, a certain selectivity can be observed with the mass extraction of 3.2% lithium and 1.3% iron when compared to the other thermal treatment conditions. On the other hand, the results obtained in leaching with 0.5 mol/L sulfuric acid (Fig 7c) and 0.25 mol/L (Fig 7b) reveal that notable conditions for selective lithium extraction relative to the other elements present were achieved. In general, for both acidic medium conditions, there is a tendency towards the production of selective lithium extraction

for cathodic powders calcined between 500°C and 700°C. However, by using the microwave-assisted leaching process with 0,25 mol/L of sulfuric acid solution and with the material calcined at 600°C was possible to produce a leachate with 2,6% of Li, 0,08% of Fe, 3,35% of P, 0,04% of Al, and 0.01% of Cu, which was considered the best in terms of selectivity and productive performance. For leaching with 0.1 mol/L sulfuric acid solutions, an excellent performance in lithium extraction selectivity relative to the other metals present is perceived, but with low efficiency, with the best selectivity conditions being able to selectively remove 0.6% of lithium without detecting the presence of other metals in solution.



Figure 7. Mass concentration of elements in leachates obtained with different acid medium for calcined and non-calcined cathodic material powders of LFP batteries.

For a better visualization of the effectiveness of the selective leaching process, the graphs in Fig 8 present a ratio of the mass content of lithium divided by the mass content of iron extracted at all tested sulfuric acid concentrations and in comparison, with the standard complete extraction with aqua regia.



Figure 8. Relationship in terms of mass content of lithium to mass content of iron extracted at different concentrations of sulfuric acid and aqua regia in the microwave-assisted leaching process.

In this scenario presented in Fig 8, the higher the value obtained on the ordinate axis, the greater the selectivity in lithium extraction. When the values are greater than one unit, it means that there is a higher mass content of lithium than iron. In this sense, it is evident that leaching with 0.10 mol/L sulfuric acid with the cathodic material calcined at 600°C yields the best results, with lithium extraction being 300 times higher than iron extraction. However, when analyzing the mass contents extracted in this process, respectively, for lithium and iron, were found 0,67% and 0,0021%. Excellent selectivity but with low efficiency. Nevertheless, when observing the mass ratio relationship of %Li : %Fe for all sulfuric acid concentrations, a tendency in the selectivity of lithium extraction becomes evident upon thermal treatments of the cathode material, particularly at temperatures of 500°C, 600°C, and 700°C, compared to values obtained with aqua regia where no significant change in this proposed ratio is observed.

Finally, to elucidate the performance of the proposed leaching processes at different sulfuric acid concentrations, Fig. 9 presents the total lithium values extracted under the proposed conditions compared to leaching with aqua regia, which is assumed to promote complete dissolution of the metals. In this graph, it remains evident that there is a clear trend of selective lithium extraction for all proposed sulfuric acid concentrations, with higher acid concentrations resulting in greater lithium extraction, albeit with an increase in the iron content in solution. For instance, considering materials calcined at 500°C, 600°C, and

700°C and leached with 1,5 mol/L H<sub>2</sub>SO<sub>4</sub>, it is possible to extract, respectively, 95%, 94%, and 90% of all lithium and 37%, 31%, and 25% of all iron present. However, when analyzing the performance of leaching under less aggressive conditions, i.e., with lower sulfuric acid concentrations such as 0,25 mol/L H<sub>2</sub>SO<sub>4</sub>, for cathodic powders calcined at 500°C, 600°C, and 700°C, it is possible to extract, respectively, 37%, 60%, and 31% of all lithium and only 1.6%, 0.2%, and 0.8% of all iron present. These conditions are considered to exhibit the highest selectivity and efficiency in the extraction process developed.



Figure 9. Percentage of total lithium extraction (left) and iron extraction (right) as a function of sulfuric acid concentration, using complete metal extraction with aqua regia leaching as reference.

Based on the presented and analyzed results under different correlations, it is possible to observe that the condition of thermal treatment at 600°C combined with leaching using 0.25 mol/L sulfuric acid yields the best results in terms of lithium extraction selectivity, generating a solution containing 60% of all lithium present and only 0.23% of all iron present. These findings can be correlated with the phase change from LiFePO<sub>4</sub> to Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>, which occurs from 500°C onwards, leading to a higher availability of lithium atoms per unit cell. However, as the thermal treatment progresses, the oxidation of iron to form Fe<sub>2</sub>O<sub>3</sub> becomes more intense, which does not contribute to increased lithium extraction. This indicates that there is no need to promote thermal treatments at high temperatures (700°C, 800°C, and 900°C) since there may be decomposition of lithium compounds and consequent loss of this element of interest. It is inferred that a second leaching cycle of this material under the best proposed conditions could recover the remaining lithium present in the sample selectively, i.e., without extracting the other elements present.

#### Conclusion

This study presents a beneficiation process for cathodic material from LFP batteries, achieving a concentration of 96% of the lithium content in particle size fractions smaller than 0,5 mm. When this material was thermally treated in an oxidizing atmosphere, starting from 500°C, is promoted a phase transformation of the predominant crystalline structure LiFePO<sub>4</sub> to Li<sub>3</sub>Fe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> phase, in which there is a greater quantity of lithium available per unit cell and the consequent formation of iron oxide. With aqua regia leaching serving as a standard for thorough metal extraction, optimal lithium selectivity was observed with samples calcined at 600°C and leached with 0.25 mol/L sulfuric acid. Under these conditions, 60% of all lithium, 0.23% of all iron, 17% of all phosphorus, 2.2% of all copper, and 1.33% of all aluminum were

found in solution. In terms of mass content of the elements, these conditions extracted 2,6% lithium, 0,27% iron, 3,4% phosphorus, 0,01% copper and 0,04% aluminum.

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