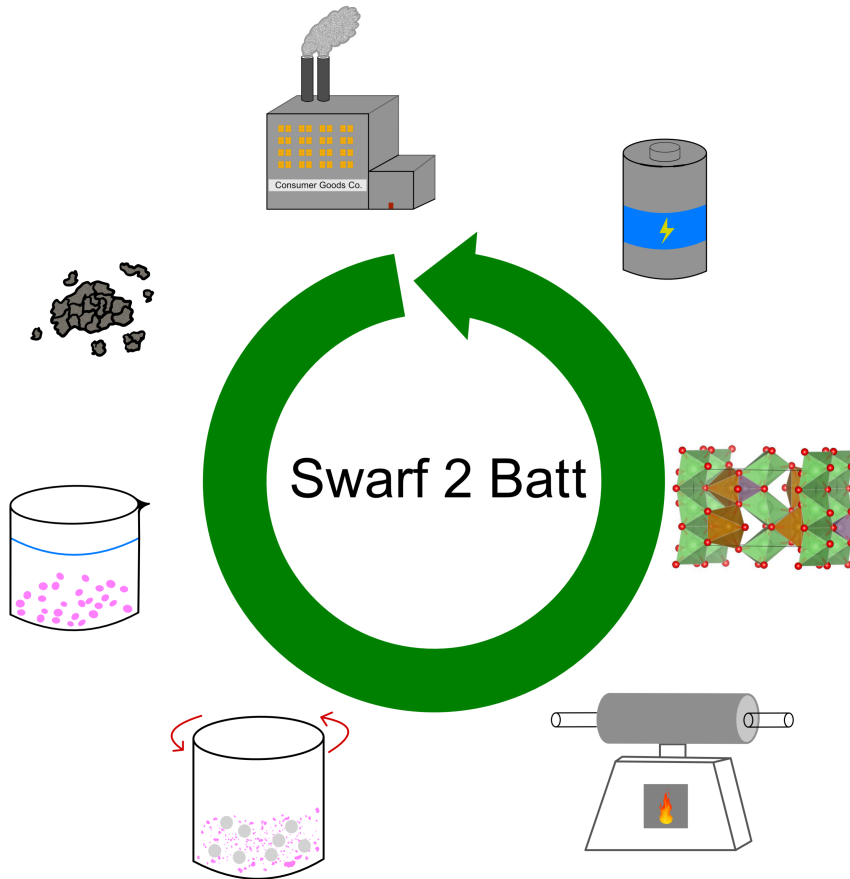




Development of Sustainable Battery Materials from Steel Waste

Particle size and morphology control of the swarf-derived iron phosphate precursors

Master's thesis in Materials Chemistry

ERIK CHARLES SMITH

Chemistry and Chemical Engineering

CHALMERS UNIVERSITY OF TECHNOLOGY

Gothenburg, Sweden 2026

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MASTER'S THESIS 2026

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Erik Smith



Department of Chemistry and Chemical Engineering
Division of Energy and Materials

Weber Lab

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Cover: Circular economy cartoon of swarf recycling into battery cathode material.

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Abstract

The global transition away from fossil fuels has driven surging demand for battery production and the associated materials such as lithium iron phosphate (LFP). Yet the LFP supply chain remains heavily concentrated in China, posing significant supply chain vulnerabilities through geopolitical tensions. In order to onshore battery manufacturing in the European and North American markets new feedstocks must be developed. This thesis investigates the feasibility of producing LFP from steel machining swarf through the lens of particle size optimization utilizing high energy ball milling. Surfactant assisted ball milling with 5 wt% myristic acid addition was identified as the optimal condition for sufficient particle size reduction of the iron phosphate precursor, reducing the median particle size from approximately 3 μm to 0.5 μm . The resulting LFP cathode achieved a specific capacity of 102 mAhg^{-1} at 0.5C with approximately 85% capacity retention after 90 cycles, demonstration the viability of converting steel waste into LFP cathode material while necessitating further optimization to become competitive with commercial LFP.

Keywords: Lithium iron phosphate, lithium ion batteries, high energy ball milling, swarf, circular economy

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Finally, a thank you to my parents who sparked my fascination with chemistry.

Erik Smith, Gothenburg, June 2026

List of Acronyms

Below is the list of acronyms that have been used throughout this thesis listed in alphabetical order:

BPR	Ball-to-powder ratio
DOE	Design of Experiments
FTIR	Fourier Transform Infrared
LFP	Lithium Iron Phosphate
SAHEBM	Surfactant Assisted High Energy Ball Mill
SEM	Scanning Electron Microscopy
XRD	X-Ray Diffraction

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1

Introduction

The pressing matter of climate change, caused by excessive anthropogenic carbon dioxide production, has prompted a multinational effort to transition away from fossil fuel energy production. This effort is embodied by the Paris climate agreement, adopted by 195 parties in 2015, attempting to limit global heating to below 2°C above pre-industrial levels [1]. Meetings on improving our impact on the climate continue with the "Conference of the Parties" (COP) that occurs annually to discuss progress and new measures to be taken [2]. Such commitments to transition from fossil fuels have led to a surge in demand for battery production, as energy storage plays a critical role in ensuring a reliable and abundant supply of electricity for consumers and businesses [3]. This has led to predictions for the Li-ion battery market to increase by six-fold by 2030 from 2022 [4].

Global Li-ion battery cell demand, GWh, Base case

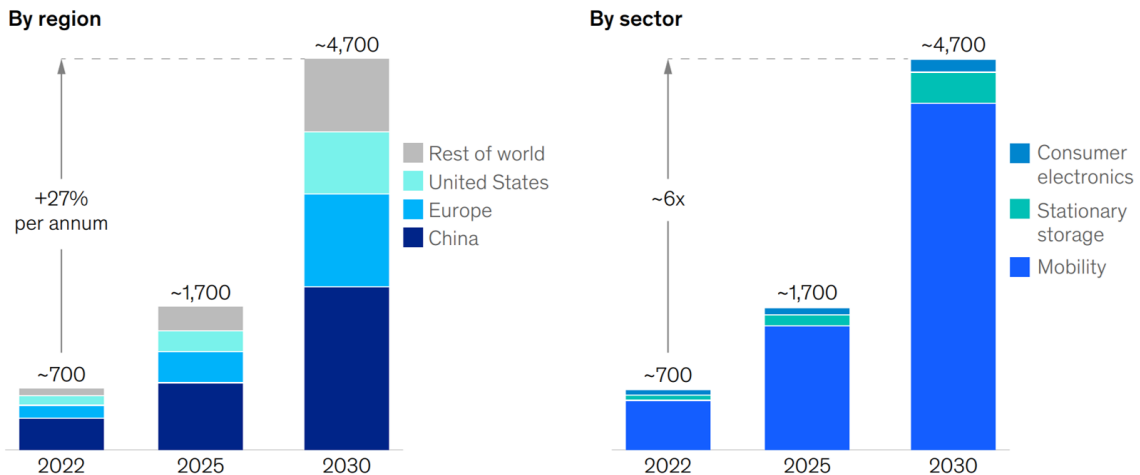


Figure 1.1: Predictions for increased Li-ion battery demand with roughly 27% annual growth [4].

Subsequently, this has led to increased demand for the materials needed for battery production including iron [5].

Similarly to battery demand and production, consumer goods manufacturing has increased over the past century [6]. A significant driver of this growth is the steel industry, which underpins most major manufacturing sectors, from automotive and construction to electronics and energy infrastructure. Total crude steel production reached 1,882.6 million tonnes in 2024 [7]. A significant by-product of this production is machining swarf: fine particles that are unusable in industry and largely end

up in landfills [8]. The swarf produced from machining of steel goods is iron-rich, providing a potential secondary source of battery-grade iron.

The iron precursor is a foundational input in LFP cathode production, typically from ferrous sulfate (FeSO_4) or iron phosphate (FePO_4). These materials are often available in Western countries through the collection of by-products from acid surface cleaning of steel, though the industrial infrastructure to convert the intermediates into battery-grade precursors is heavily skewed towards China, creating potential geopolitical risk [9]. The concentration of iron precursor production capacity sits within the broader pattern of Chinese dominance across the LFP supply chain. Over 98% of LFP cathodes material and LFP battery cells are currently produced in China [10]. This market concentration is only predicted to grow as demand and manufacturing of Li-ion batteries increase [11].

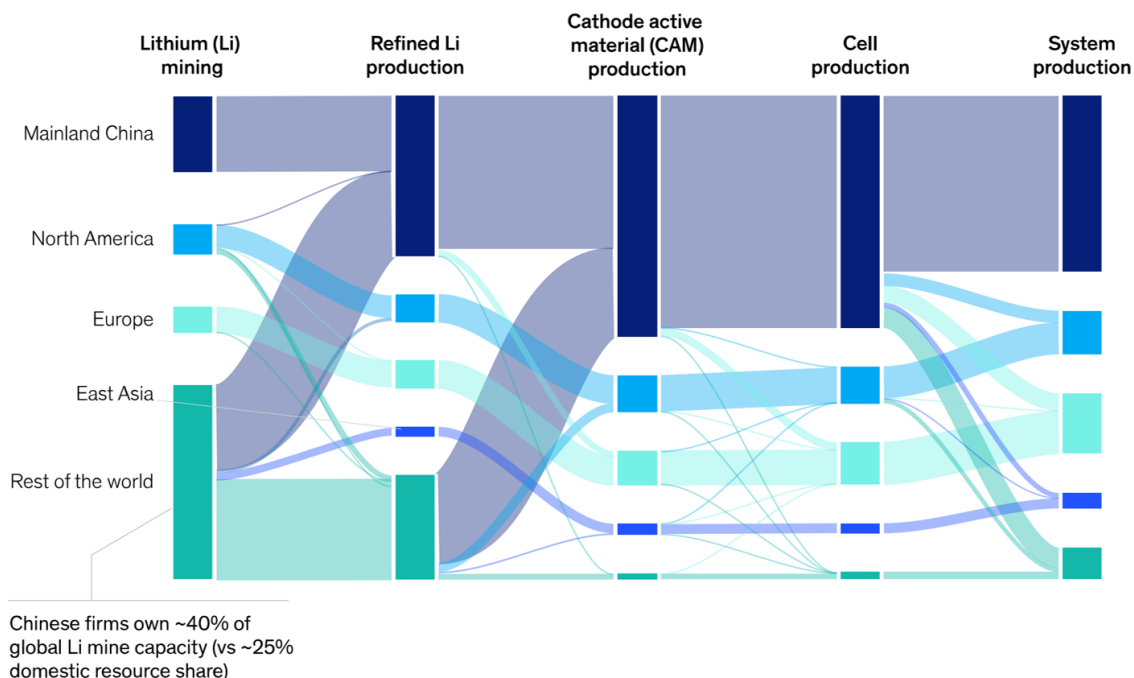


Figure 1.2: A prediction of material flows for 2035 lithium-ion battery manufacturing [11].

The overwhelming majority of batteries used for power grids and electric vehicles rely on components or batteries sourced from China [5]. China has begun to actively leverage this position: in January 2025, China’s Ministry of Commerce proposed export restrictions requiring Chinese firms to obtain licenses before exporting technologies used to produce the most advanced versions of LFP [12]. Based on current trends, Europe and North America combined are estimated to hold only five to ten percent of global production capacity for key battery components by 2030 [9]. Societal and industrial reliance on battery production along with geopolitical risks posed by Chinese export restrictions highlight the pressing need to develop alternative, regionally sourced, iron precursor feedstocks capable of supporting LFP production outside of China.

1.1 LFP Synthesis

Various synthetic methods have been developed for lithium iron phosphate since Padhi, Nanjundaswamy, and Goodenough first identified its electrochemical potential as a cathode material in 1997 [13]. The methods can largely be divided into two categories, solid-state and solution-based.

Solution-based methods such as hydrothermal, sol-gel, and co-precipitation have attracted considerable interest for their precise control over particle morphology and size, both strongly influencing the electrochemical performance of LFP [14]. In hydrothermal synthesis lithium, iron, and phosphate precursors are dissolved in an aqueous solution of the desired pH, sealed in an autoclave and reacted under high pressure and elevated temperatures, between 120 and 200°C, to produce LFP [15]. The sol-gel method involves the formation of a homogeneous gel from dissolved precursors through mixing and drying, which is subsequently calcined to form LFP. The homogeneous mixing of the precursors at the molecular level has made sol-gel an attractive synthetic method [14]. The co-precipitation method involves dissolving precursors, adjusting to the desired pH, then drying, milling, and heat treatment. This method offers good morphological control but is sensitive to impurities in the precursor materials [14, 16]. These solution-based synthetic methods offer great control over the resulting morphology and particle-size; however, they are difficult to scale up to mass production due to complex procedures resulting in high costs and environmental risks [14].

Due to the mass production issues associated with solution based synthesis methods, solid-state methods typically dominate industrial LFP production primarily due to simplicity and ease of scaling. In this approach, dry precursor powders including a lithium source, an iron source such as ferrous oxalate or ferrous sulfate, and a phosphate source are thoroughly mixed to ensure uniform distribution before calcination between 600-800°C under an inert or reducing atmosphere [14]. A carbon source such as glucose or sucrose is typically introduced during a milling or mixing step to form a carbon layer during calcination. This is to improve the poor inherent electronic conductivity of LFP [17]. Although there is an increased energy consumption associated with solid-state methods, compared to solution-based, the scalability of this method has made it ideal for LFP cathode production.

1.2 Batteries & Energy Storage

Batteries have become so ubiquitous in the modern world that they are almost a given in consumer goods as well as industry. The possibility of storing energy for later use is far from a novel concept with the term 'electrical battery' dating back to the mid-1700s [18]. While early investigations into energy storage and electricity itself were quite limited, one of the first battery-like apparatuses was developed in 1800 by Alessandro Volta. Known as the 'Voltaic Pile', it consisted of multiple discs of tin or zinc stacked with discs of copper, brass, or silver. Water-soaked intermediates made from paper, fiber board, or leather separated the discs while wires connected

the top and bottom discs pairs. This early battery device allowed researchers to access a consistent electric current [18]. Volta's early battery functioned through the same oxidation and reduction mechanism that modern batteries do, though the Voltaic Pile produced an electric current not through the reduction of the cathode material, the silver or copper discs in this case, but rather the hydrogen in the surrounding water [18]. This conclusion is drawn from that fact that there were no ions of the cathode material readily present, which are needed for the reduction of the cathode species, as well as by the fact that Volta and other researchers noted polarization issues, reductions in the voltage output, arising from hydrogen bubbles adhering to the electrode surfaces blocking electrochemical reactions [18].

Later advances in battery materials brought about the use of lead grids and lead oxide electrodes in an acidic electrolyte solution, commonly known as 'lead-acid' batteries. The early iterations of lead-acid batteries were capable of being recharged, although the processes needed to create the necessary current for efficient charging would not be developed until about 30 years after the development of lead-acid energy storage [18]. Lead-acid batteries were and continue to be quite popular and serviceable, accounting for over 35 billion USD in 2015 and roughly 60% market share in 2021 [19, 20]. This is largely due to the low cost associated with the production of lead-acid batteries, even though the energy density is significantly lower than the theoretical limit, as well as concerns of hydrogen gas production during overcharging or at metal impurity sites [21]. Following lead-acid batteries came nickel based batteries, namely nickel cadmium (NiCd) or nicad batteries. This battery material offered a far lighter overall battery compared to lead-acid due to the lower density of nickel and cadmium (7.8 and 8.0 g/cm³ respectively, compared to 10.66 g/cm³ for lead) [18]. While preferable to lead-acid batteries for more mobile applications, NiCd batteries suffered from several drawbacks, namely memory effects that required the complete discharge prior to recharging in order to avoid charge capacity loss as well as disposal concerns related to the toxic cadmium cathode. Though there have been improvements to nickel batteries, such as the replacement of the cadmium cathode with a nickel-metal alloy, lithium-ion batteries have far overtaken global interest as the predominant battery type [18].

Early uses of lithium in batteries were fraught with several hurdles. Lithium metal, being an alkali metal, reacts with water meaning aqueous electrolyte solutions were not feasible. Other lithium-based batteries required high operating temperatures, exceeding 600 °C [18]. Secondary lithium-based batteries suffered from uneven redeposition during recharging, this rendered batteries susceptible to shorting through the formation of dendrites. Finally in the 1970's the intercalation of lithium-ions into a host framework was described, which allowed for the phasing out of pure lithium metal electrodes [18]. Several framework materials have been investigated and come into circulation such as cobalt oxide (CoO₂), titanium(IV) sulfide (TiS₂), vanadium pentoxide (V₄O₁₀) and graphite [18]. The major Li-ion battery materials currently are nickel-manganese-cobalt (NMC) and lithium iron phosphate (LFP), occupying 52% and 23% respectively of cathode materials for Li-ion batteries in 2020 [20]. Predictions continue to place NMC and LFP cathodes as the dominant materials for lithium-ion batteries through 2030 [20].

The cost of producing these Li-ion batteries has been steadily decreasing over the years as cell manufacturing, separator, and electrolyte components become more streamlined and accessible. The cost associated with the cathode portion, however, has not seen a marked decrease [20]. This is largely due to the cost of raw material procurement [22, 23]. As there is increasing demand for battery production, ethical and environmental concerns have risen surrounding the sourcing of virgin battery materials [23].

1.3 Swarf

Swarf is a by-product of the manufacturing sector. As metal goods are machined, ground, or drilled, small metallic particles get produced. In 2006 it was estimated that between 2.3 and 5.8 million tons of metal swarf was produced [24]. As consumer goods production increases with each passing year, recent estimations put the production of metal swarf between 10-12 million tons in 2024 [25]. Grinding swarf is comprised of 50-80 weight percent iron with the remaining percentages spread amongst the material of the grinding apparatus that break off and adsorbed cutting oils [26, 27].

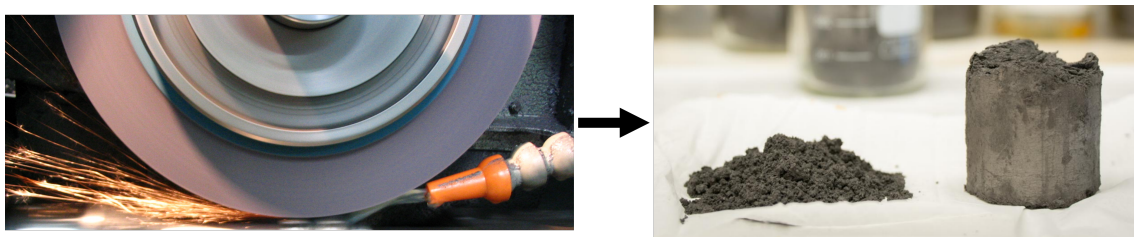


Figure 1.3: Example grinding process (left) [28] that produces swarf (right) [29].

The disposal of swarf in landfills poses grave environmental concerns as the metals and adsorbed, non-biodegradable oils can leach into local soil and groundwater [30]. In an effort to reduce the environmental impact of swarf waste, manufacturers have attempted various cleaning and recycling methods. However, these methods often require high energy consumption or result in further environmental concerns. For example, Fu et. al. explored two methods of cleaning swarf, aqueous surfactant washing and supercritical carbon dioxide (SC-CO₂) extraction [27]. In aqueous surfactant washing, a water-soluble phosphorus additive is used to remove cutting oils, lubricants, and other organic compounds from the swarf. This method was able to remove 86% of the absorbed oil after several washing with only one of the tested surfactants [27]. This process also resulted in a large quantity of aqueous surfactant emulsion the required further treatment before disposal [27]. The supercritical CO₂ method involves passing the carbon dioxide mobile phase through a bed of swarf to dissolve the absorbed contaminants. This process requires high pressure and elevated temperature to ensure efficient extraction by the SC-CO₂ [27]. In short, high energy consumption. Neither of these processes were economically feasible when compared to the low financial cost of simply disposing of the swarf in a landfill [27]. Though these pathways of cleaning swarf are possible, manufacturers are less inclined

to process the swarf themselves, as the oil can cause fires or explosions [24]. A more novel approach uses hydrometallurgy to sequester the iron in swarf into ferric chloride (FeCl_3) through leaching [31]. This approach requires maintaining specific pH levels and dilute hydrochloric acid to separate the iron from the cutting fluids and other contaminants [31]. The iron chloride could then be used to synthesize iron phosphate as was done for this research.

To reduce the amount of swarf waste new processes need to be developed to cycle metal swarf into usable material. As metal swarf consists of mostly iron it can be reclaimed for use in other consumer goods. This thesis will explore the recycling of swarf waste into usable battery cathode material.

2

Theory

2.1 Energy Storage

Numerous consumer electronics use stored energy through batteries, such as electric vehicles (EVs), mobile phones, or toothbrushes. Simplistically, batteries are chemical circuits that store electrical energy as chemical energy that can be readily accessed through an oxidation-reduction (redox) reaction. A typical battery consists of four parts, a positive electrode or anode, a negative electrode or cathode, an electrolyte, and a separator. The anode of a battery undergoes reduction during discharge, while the cathode undergoes oxidation. The electrolyte is responsible for selective conduction of ions involved in the redox reaction but should be insulating to free electrons. The separator physically separates the electrodes but must be permeable to the ions necessary in the redox reaction [32].

There are two main types of batteries: primary (nonrechargeable) and secondary (rechargeable). For the purposes of this report, secondary or rechargeable batteries will be the focus.

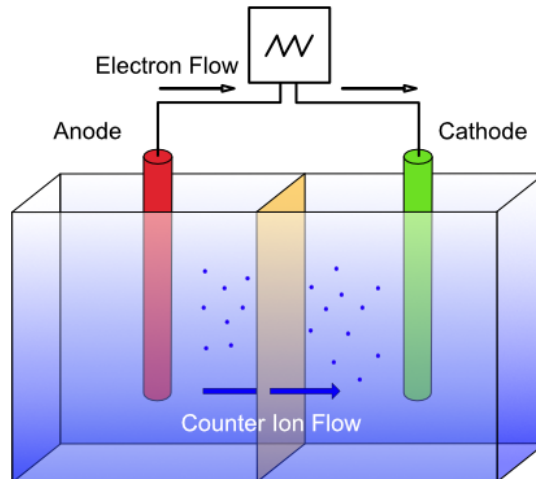
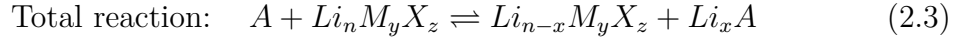
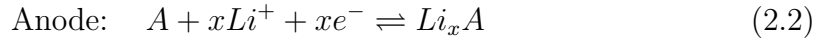
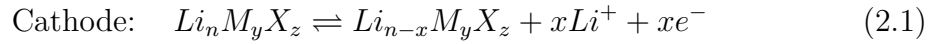


Figure 2.1: Battery cartoon depicting discharge operation. The electrons flow from the anode (red electrode) to the cathode (green electrode) as counter-ions flow through the electrolyte (blue gradient) and the separator (yellow sheet).

The individual material of the cathode and anode can vary greatly with each affording various benefits and drawbacks. The efficiency of the battery in terms of capacitance and longevity depends largely on the material utilized in the battery. Lithium-ion (Li-ion) batteries have become one of the main contenders for optimal battery material in rechargeable batteries due to its combination of thermal safety, long cycle life, low cost, and reliance on abundant raw materials, making it particularly attractive for grid storage and electric vehicle applications [14].

2.1.1 Lithium-Ion Batteries

Lithium-ion batteries operate on the principle of intercalation and deintercalation of lithium ions that accompany redox behavior. As the Li-ion battery discharges, lithium ions deintercalate from the anode, which is oxidized as electrons travel toward the cathode. The lithium ions then pass through the electrolyte and separator to intercalate into the cathode. For the charging process, the reverse happens. This process of inter- and deintercalation of lithium ions yields stable cycling performance and safety as the electrode structures are far less reactive than the lithium metal [33]. The electrochemical reactions are as follows:



Li-ion batteries boast exceptional longevity, being able to undergo thousands of discharge cycles [34]. Li-ion batteries themselves can come in several forms, including lithium-oxide batteries, lithium-ion polymer batteries, or lithium-iron phosphate batteries. LFP batteries specifically, have become one of the best in terms of performance and production. With long life cycle, and optimal operating temperatures, LFP batteries have found their way into numerous consumer electronics [17].

2.2 Lithium Iron Phosphate

LFP crystallizes into an olivine-type structure in the Pnma space group. The lithium ions in Figure 2.2 occupy one-dimensional channels encased by the iron phosphate tetrahedra. During the discharging of an LFP battery the lithium ions travel along these channels with low probability of jumping to different channels. This one-dimensional motion of lithium ions means that channels can be susceptible to blockage by defects or impurities [35]. Anti-site defects, where an iron atom and lithium atom swap places, are fairly common and pose a problem when they occur in high number or in the same channel. This prevents lithium diffusion in that channel and can drastically reduce the specific capacity, the amount of charge a battery electrode can store per unit mass, and efficiency of an LFP battery if a large number of channels are blocked.

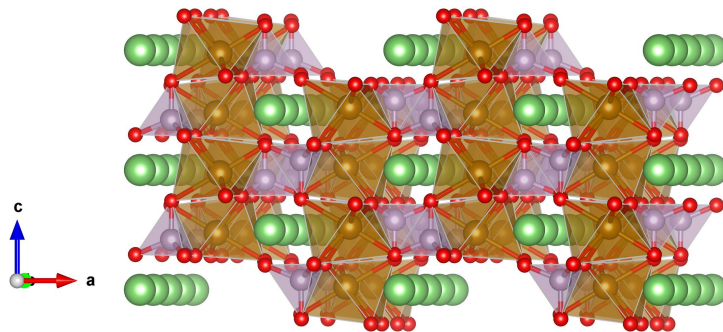


Figure 2.2: Olivine crystal structure of LiFePO_4 with FeO_6 octahedra (brown), PO_4 tetrahedra (purple), lithium ions (green), and oxygen (red).

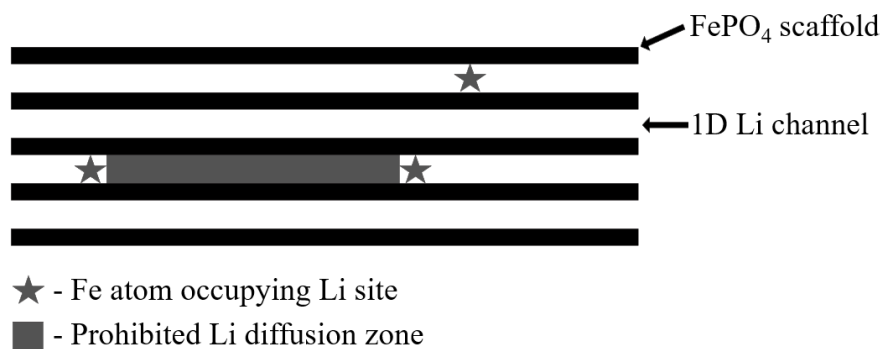


Figure 2.3: Cartoon representation of anti-site defects blocking 1-dimensional travel of lithium ions.

2.3 Ball Milling

2.3.1 General Overview

In order to reduce the particle size or alter the morphology of a powder sample, energy needs to be supplied to break some of the bonds holding the structure together. Ball mills supply this energy through the movement of grinding media that grind and crush powders either on other grinding balls or on the walls of a container. The energy supplied powder sample can be heavily regulated by tailoring the speed or time the ball mill is operating. The conditions inside the milling container can also be tailored to provide efficient particle size reduction. The grinding material, such as zirconium oxide (ZrO_2) or stainless steel, can provide different hardness and mass of the grinding balls affecting the energy imparted per collision. Though, considerations must be taken to prevent the grinding media itself from being eroded in cases where the material of the powder sample is sufficiently hard. The number of grinding balls in relation to the powder is also a consideration, often referred to as the ball-to-powder ratio (BPR). This parameter is sometimes the only one reported in scientific literature though Kuziora, et. al., points out in their investigation of

BPR on ball milling efficiency, BPR alone is insufficient to describe the effectiveness of ball milling conditions [36]. The many important parameters to consider when ball milling can be found in Table 2.1.

Ball Milling Parameters
Sample hardness
Ball size
Total ball mass
Container volume
Powder volume
BPR
Milling speed
Milling time
Milling atmosphere
Additives

Table 2.1: Important parameters for consideration to achieve efficient particle size reduction from ball milling [36, 37].

When all considerations have been made, ball milling can be a versatile method of controlling the particle size of various compounds. Though most commonly used for minerals or other inorganic powders, ball milling has found use in the preparation of nanocellulose derivatives or of polymer nanocomposites [38, 39, 40]. To accompany the diverse applications of ball milling there are a plethora of milling instruments.

2.3.2 Ball Mill Types

Attrition mills, also known as the attritor or stirred ball mill, use a fixed cup with grinding balls that are then stirred by horizontal arms attached to a rotating central shaft. Horizontal mills are comprised of a large cylinder that rotates around a horizontal central axis. The centrifugal force of this rotation combined with gravity causes the grinding balls to rise and fall within the powder material. Planetary mills

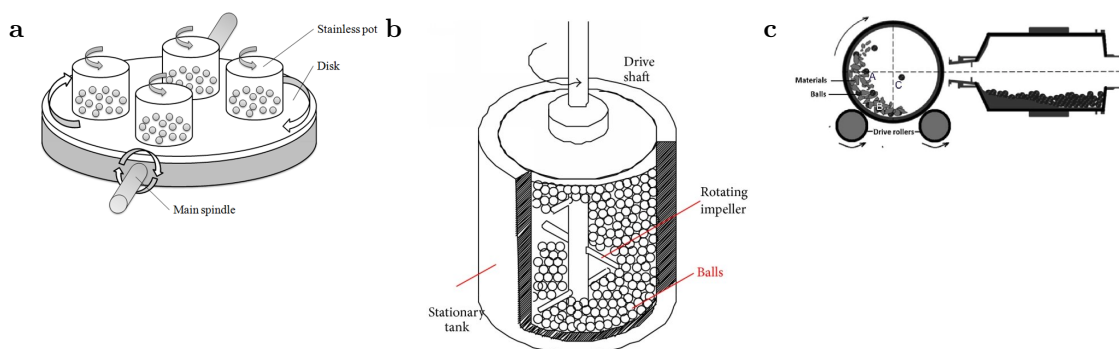


Figure 2.4: Types of ball mills a. planetary [41] b. attritor [42] c. tumbler [43].

utilize fixed containers atop a rotating plate. Each container can rotate individually, though coupled to the rotation of the spinning plate. This form of ball milling produces high-energy impact and shear forces by rotating grinding jars in the opposite

direction of the central disk, causing the grinding balls to follow a curved trajectory rolling over one another as seen in Figure 2.5 [44, 37].

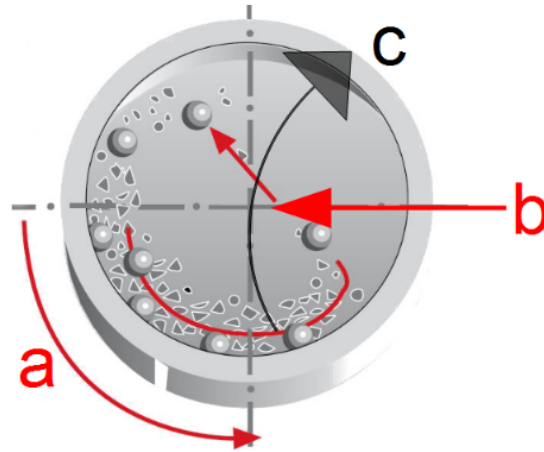


Figure 2.5: Motion of grinding media in a planetary ball mill jar [45].

2.3.3 Surfactant Assisted Ball Milling

During ball milling there is continuous process of fracturing and coalescence [46]. Meaning that as primary and secondary particle structures are broken down new secondary particles are created through agglomeration. In some cases, this can be porous secondary particles where the primary particles can be easily identified and potentially separated through processes such as sonication. In other cases, excessive agglomeration can occur where primary particles are difficult to separate from the secondary structure. This agglomeration occurs as the newly formed nanoparticles attempt to reduce their surface energy [47]. In order to prevent agglomeration of the primary particles into large secondary particles the use of surface-active agents, or surfactants, can aid in maintaining separation between the milled particles [48]. The function of a surfactant in ball milling is to bind loosely to the surface of newly milled nanoparticles and prevent agglomeration or cold welding. The adsorbed surfactant does this by reducing the surface energy of the newly milled particles as well as, depending on the surfactant composition, provide a steric barrier between particles [46]. Thus, the choice of surfactant must be predicated on the powder material being milled as well as the milling media. Sufficient care must be taken to ensure the surfactant will not react with the powder sample, creating impurities or altering the phase of the sample. In one example Dapeng et. al., prepared $\text{Sm}_2\text{Co}_{17}$ nano-flakes through high energy ball milling with oleic acid as a surfactant [49]. They concluded that the addition of the surfactant played a key role in preventing the re-agglomeration of particles [49].

2.4 X-Ray Diffraction

X-ray diffraction (XRD) is a critical analytical technique in determining the atomic structure of crystalline materials. A non-destructive technique, XRD works by measuring the diffraction of X-rays by a sample. The resulting interference spectra allows researchers to determine parameters including unit cell dimensions, symmetry, strain, and defects [50]. XRD relies on measuring the intensity of diffracted X-rays while varying the diffraction angle (2θ). Varying 2θ provides insight into the inter-planar spacing (d-spacing) of the crystal lattice used in the determination of the crystal structure [50]. Special considerations must be taken when choosing an X-ray source depending on the sample composition. For example, a common X-ray source in XRD is the copper (Cu) emission tubes, which produces a characteristic wavelength that is adequate for most samples. However, for samples containing iron (Fe), the copper source X-rays cause fluorescence in the sample leading to high backgrounds counts and making structure determination far more arduous [51]. This is a consequence of how the X-rays interact with the electrons of the specific atoms. In the case of copper source X-rays and iron atoms, the X-ray interacts with an inner shell electron via inelastic scattering. The X-ray is diffracted, but some of the energy of the photon has been transferred to the electron, raising it out of the inner shell. This decreases the intensity of the diffracted X-ray while producing fluorescence in the iron atom. In cases such as this, other X-ray sources such as cobalt (Co) tubes should be used. Co X-ray sources work without causing fluorescence as the $K\alpha$ wavelength is greater than the K absorption edge of the iron atoms which stands at $1.743 \text{ \AA}$ [51]. Another good alternative to a Cu source is a molybdenum (Mo) source, however, as the $K\alpha$ of Mo is than the K absorption edge of iron, a sensitivity window must be added to the detector. This sensitivity window allows the collection of diffraction data without interference from fluorescence for Mo but not Cu sources [51].

X-ray Source	$K_{\alpha 1}$	$K_{\alpha 2}$	K_{β}
Cu	1.54056	1.54439	1.39222
Co	1.78897	1.79285	1.62079
Mo	0.7093	0.71359	0.63229

Table 2.2: X-ray sources and their characteristic radiation wavelengths in Ångström.

3

Methods

3.1 Iron Phosphate Precipitation

Swarf derived iron phosphate was obtained from two feedstock pools: unmodified swarf and washed swarf. The washed swarf had undergone additional cleaning steps prior to processing to reduce the content of adsorbed organic compounds and metal contaminants relative to the unmodified material. With the exception of the pre-treatment, the synthetic pathway was identical for both feedstocks.

The iron contained in the swarf was leached into solution using hydrochloric acid to form iron chloride salts. These salts were subsequently precipitated using phosphoric acid, yielding the iron phosphate precursor used in this work. The iron phosphate derived from unmodified swarf and washed swarf are referred to as recycled iron phosphate and washed recycled iron phosphate, respectively. It should be noted that the precipitation of iron phosphate was carried out by Thomas Ottink at a collaborating lab.

3.2 Ball Milling

The primary goal of ball milling the iron phosphate precursors was to obtain a particle size and morphology comparable to that of the commercial iron phosphate used as a reference material for initial testing. A design of experiments (DOE) was established to determine the optimal combination of milling time, grinding media diameter, and rotation speed for particle size reduction. The parameters tested are presented in Table 3.1. All experiments for the DOE were conducted with a ball-to-powder ratio (BPR) of 10:1 using zirconium oxide (ZrO_2) milling balls and cups on a TMAXCN planetary ball mill. Isopropanol (IPA) was added to each sample to form a slurry, as such all experiments in Table 3.1 are classified as wet milling.

The effectiveness of surfactant addition on particle size reduction was also investigated. Two surfactants, glycerol and myristic acid, were tested at two weight percentages, as detailed in Table 3.2. The high weight percent addition for glycerol in Table 3.2 was intended to replicate wet milling conditions by creating a slurry. Myristic acid, being a solid at room temperature, could not be incorporated in the same manner and was therefore tested at a lower quantity. All surfactant-assisted milling experiments were conducted without IPA or any additional liquid medium. A grinding media diameter of 2mm and a BPR of 10:1 was held constant across all surfactant assisted experiments.

Time (hr)	Ball Diameter	Speed (rpm)
0.5	10	200
0.5	2	200
0.5	2	800
0.5	5	800
4	10	200
4	2	200
4	10	800
4	2	800
6	10	200
6	2	200
8	2	500

Table 3.1: DOE tests for determining optimal wet ball milling conditions.

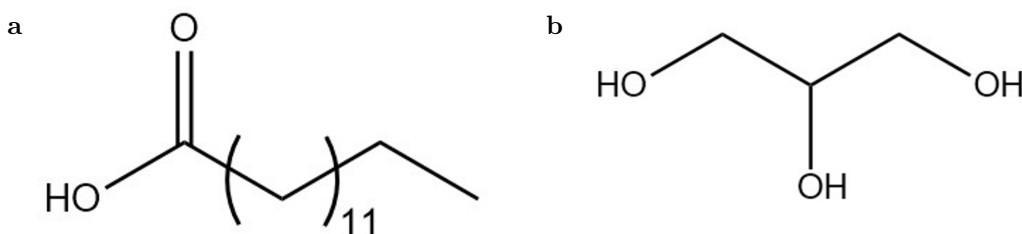


Figure 3.1: Lewis structures of a. myristic acid and b. glycerol

surfactant	Weight percent (wt%)
Glycerol	5.0
	200
Myristic acid	5.0
	10

Table 3.2: surfactant and weight percentages tested.

Following surfactant-assisted milling, the iron phosphate particles retained a surface coating of the respective surfactant, which required removal prior to calcination to prevent unintended carbon deposition or side reaction during the subsequent heating. Glycerol was removed by washing with deionized water, while myristic acid was removed using either ethanol or methanol. In each case, the milled powder was transferred to an Erlenmeyer flask with approximately 100 mL of the chosen solvent and stirred slowly for one hour, after which the suspension was vacuum filtered and washed with a further 50 mL of fresh solvent. Fourier-transform infrared spectroscopy (FTIR) was performed on the washed iron phosphate precursors to confirm the absence of detectable surfactant residue prior to further processing.

3.3 Particle Size Determination

Particle size distributions were obtained through machine learning-assisted analysis of SEM images using the Cellpose algorithm implemented in MATLAB. Cellpose is a generalized segmentation model originally developed for biological applications, where it is used to segment, size, and count cells [52]. However, its underlying segmentation framework is applicable to non-biological imaging contexts, making it a practical tool for estimating particle size distributions when access to dedicated particle size analysis instruments is limited.

The Cellpose model requires four inputs: a grayscale SEM image, a segmentation model selected from the Cellpose MATLAB library or trained independently, an estimated cell diameter, and a cell threshold. The "cell" diameter parameter defines the approximate diameter of the features to be detected, directing the algorithm toward particles within a specific size range. The "cell" threshold parameter controls the stringency of particle detection, a higher threshold produces more conservative detections with fewer errors but risks missing particles that are in close proximity or overlapping, while a lower threshold increases detection sensitivity at the cost of potentially merging adjacent particles into a single detection or over-counting particles within a given diameter range.

The algorithm returns the projected areas of detected particles in units of pixels, which are converted to physical units, either micrometers or nanometers, using the scale bar provided in the SEM image. The equivalent circular diameter of each particle is then calculated from its measured area using the following equation:

$$d = 2 * \sqrt{\frac{A}{\pi}} \quad (3.1)$$

This process relies on the assumption that the particles are roughly spherical. If the particle morphology deviates significantly from this assumption, a new model must be trained to detect the non-spherical shape. Example code and representative segmentation images are provided in Appendices A.1 and A.1.1.

3.4 Calcination

Once the ball-milled recycled iron phosphate was obtained and washed, it was then calcined to synthesize LFP. A 1:1 molar ratio of iron phosphate dihydrate ($\text{FePO}_4 \cdot 2\text{H}_2\text{O}$) and battery grade lithium hydroxide monohydrate ($\text{LiOH} \cdot \text{H}_2\text{O}$), obtained from Sigma-Aldrich, were mixed in a Retsch mixer mill for 90min at a speed of 30Hz. IPA and 5mm ZrO_2 balls were added to the mixing cup. This ensured intimate mixing of the reactants to prevent regions of off stoichiometry. The resulting slurry was then briefly dried in an 80°C furnace to remove the IPA. The mixed powder was then loaded into a Nabertherm tube furnace and heated with heating and cooling ramp rate of 300K/hr to a maximum temperature of 700°C, held for two hours. During heating, a reducing atmosphere of 5% H_2/Ar was flowed through the furnace at a rate of 5ml/min controlled by a Brooks Instrument mass

flow controller. The reducing atmosphere was necessary to reduce the Fe^{+3} species to Fe^{+2} during synthesis.

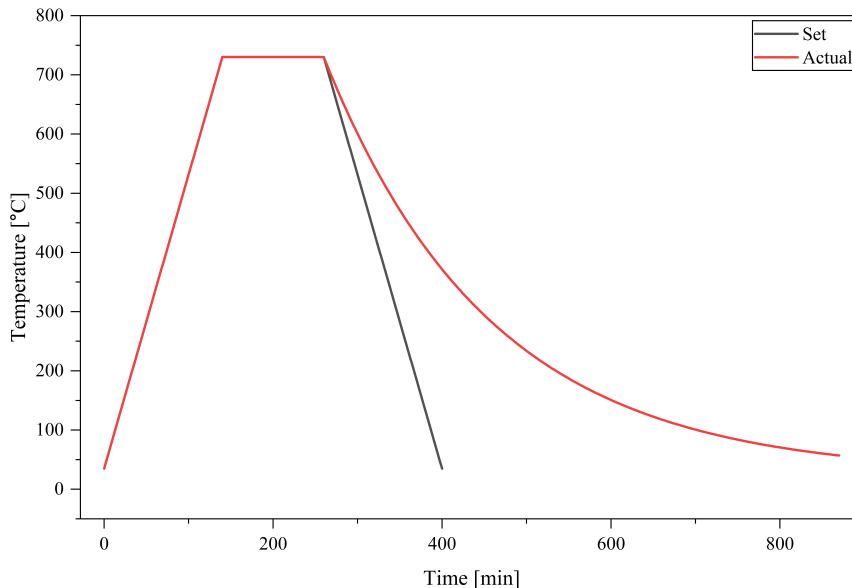


Figure 3.2: Temperature profile set on tube furnace for calcination. Gray line shows the set profile with equal heating and cooling rates. Red line displays the actual cooling rate.

The set heating regime had a maximum temperature of 730 due to a difference of -30°C within the tube measured using an external thermocouple. It should also be noted that the actual cooling rate was slower than the 300K/hr set, as we did not have access to cooling instruments for the furnace. The variation between the set and observed cooling rates can be seen in Figure 3.2. The heat simply radiated away according to Newton’s law of cooling:

$$T(t) = T_{amb} + (T_i - T_{amb})\exp(-kt) \quad (3.2)$$

Where $T(t)$ is the temperature at a given time, T_{amb} is the ambient temperature of the surrounding atmosphere, T_i is the initial temperature, k is the cooling constant that can be calculated from the furnace, and t is time. The longer cooling rate may have aided in calcination as it gave the sample ample time to crystallize. If the cooling rate is too fast, the product would end up in an amorphous phase.

3.5 Carbon-coating

To improve the inherently poor electronic conductivity of LFP, an ex-situ carbon coating was applied to the synthesized LFP powder. A combined carbon source consisting of cellulose acetate and sucrose was dissolved in acetone to form a homogeneous solution, prepared at 10 wt% respective to the mass of the LFP product.

This solution was combined with the LFP powder and mixed in a mixer mill at 30 Hz for 90 minutes using 5 mm zirconium balls to ensure an intimate mixing of the LFP powder and carbon precursors. The resulting mixture was dried at 80 °C to remove the acetone before undergoing a second calcination under identical conditions mentioned in section 3.4. During this calcination, the organic carbon precursors undergo pyrolysis, depositing a conductive layer of carbon to the surface of the LFP particles.

3.6 Analysis

3.6.1 X-Ray Diffraction

X-ray diffraction (XRD) was performed on the carbon coated LFP samples to assess phase purity and confirm the successful formation of the olivine LiFePO_4 crystal structure. As the samples contained iron, a Mo X-ray source with an appropriate filter was employed to suppress the detected iron fluorescence signal. Pawley and Rietveld refinements were performed on the collected diffraction patterns using TOPAS software to quantitatively determine phase composition and identify any secondary phases present in the product.

3.6.2 Battery Cycling

Once the phase purity had been determined, the carbon coated LFP (C-LFP) powder was sent to collaborating lab for electrochemical characterization by PhD candidate Haohang Xu. Two electrochemical test protocols were employed: rate capability testing and long cycling tests. Rate capability tests were conducted by cycling the cells at progressively increasing current densities to evaluate the specific discharge capacity of the cathode material. Long cycling tests were performed at a fixed C-rate over 100 cycles to assess capacity retention and coulombic efficiency.

4

Results

4.1 Ball Milling

The ball milling of swarf derived iron phosphate served two primary objectives: to reduce the particle size to enhance lithium diffusion within the material, and to obtain a morphology comparable to that of commercially available iron phosphate used as a reference (commercial $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$). The reduction in particle size allows for improved lithium diffusion as the pathways for intercalation and de-intercalation are shortened. Additionally, the increased surface area of the iron phosphate particles increases the reactivity of the precursor, potentially resulting in higher phase purity and limiting site defects within the crystal structure.

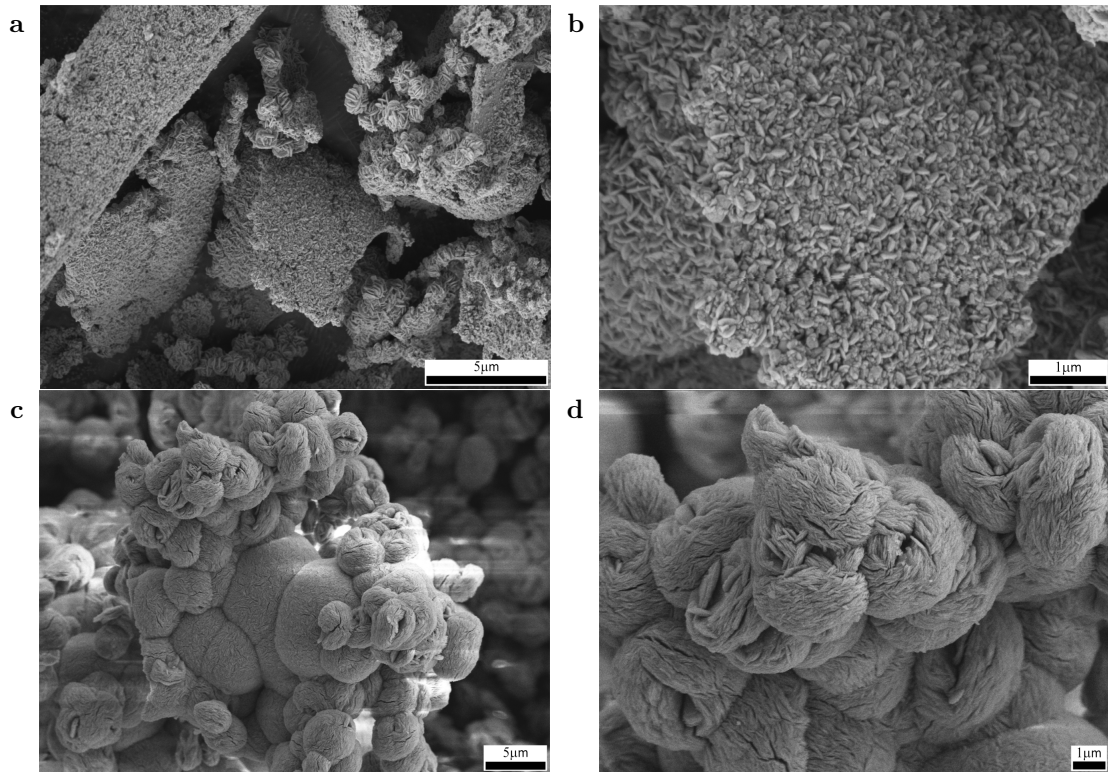


Figure 4.1: SEM images of initial commercial (a,b) and recycled (c,d) iron phosphate.

Representative SEM images of commercial and recycled iron phosphate are presented in figure 4.1. The commercial iron phosphate exhibits a porous secondary

particle morphology on the micrometer scale, composed of aggregated nanometer sized primary particles. The primary particles are relatively uniform in shape and size while the secondary particles vary greatly in size and overall morphology.

In contrast, secondary iron phosphate displays dense, spherical secondary particles composed of sheet-like primary particles. The dense secondary particle morphology could prevent effective lithium diffusion into the bulk of the spherical particle, potentially leading to lower specific capacities of the resulting LFP or residual phase impurities arising from incomplete lithiation during calcination.

4.1.1 High Energy Ball Milling

The wet milling conditions evaluated across the design of experiments yielded poor particle size reduction from the secondary $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$. At sufficient milling energies, that being a high rotational speed and extended time, the original spherical morphology of the particles would break down, however, large and dense agglomerations would form. The tendency toward agglomeration appeared to intensify with increased milling time, while showing no clear dependence on rotational speed across the conditions tested. At shorter milling durations, some of the dense morphology remained along with the newly formed agglomerations as can be seen in Figure 4.2. This behavior was consistent across all wet milling conditions tested without the presence of a surfactant.

The observed agglomeration is attributed to the increase in surface energy of the milled particles. As the large particles get milled more surface area gets exposed, or in other terms, as more atoms rest on the surface of the particles in relation to those in the bulk. While there are several factors that could result in the agglomeration of our iron phosphate particles, increased surface energy is the most likely candidate as changes in binding energy and particle surface strain are often negligible for particles on our size scale of $\sim 100\text{nm} - 500\text{nm}$ [53, 54, 55]. It should also be noted that agglomeration behavior may not be a major issue with every milled powder as it can be largely material or bond energy dependent. However, iron phosphate nanoparticles in our case agglomerate to reduce this increased surface energy [53].

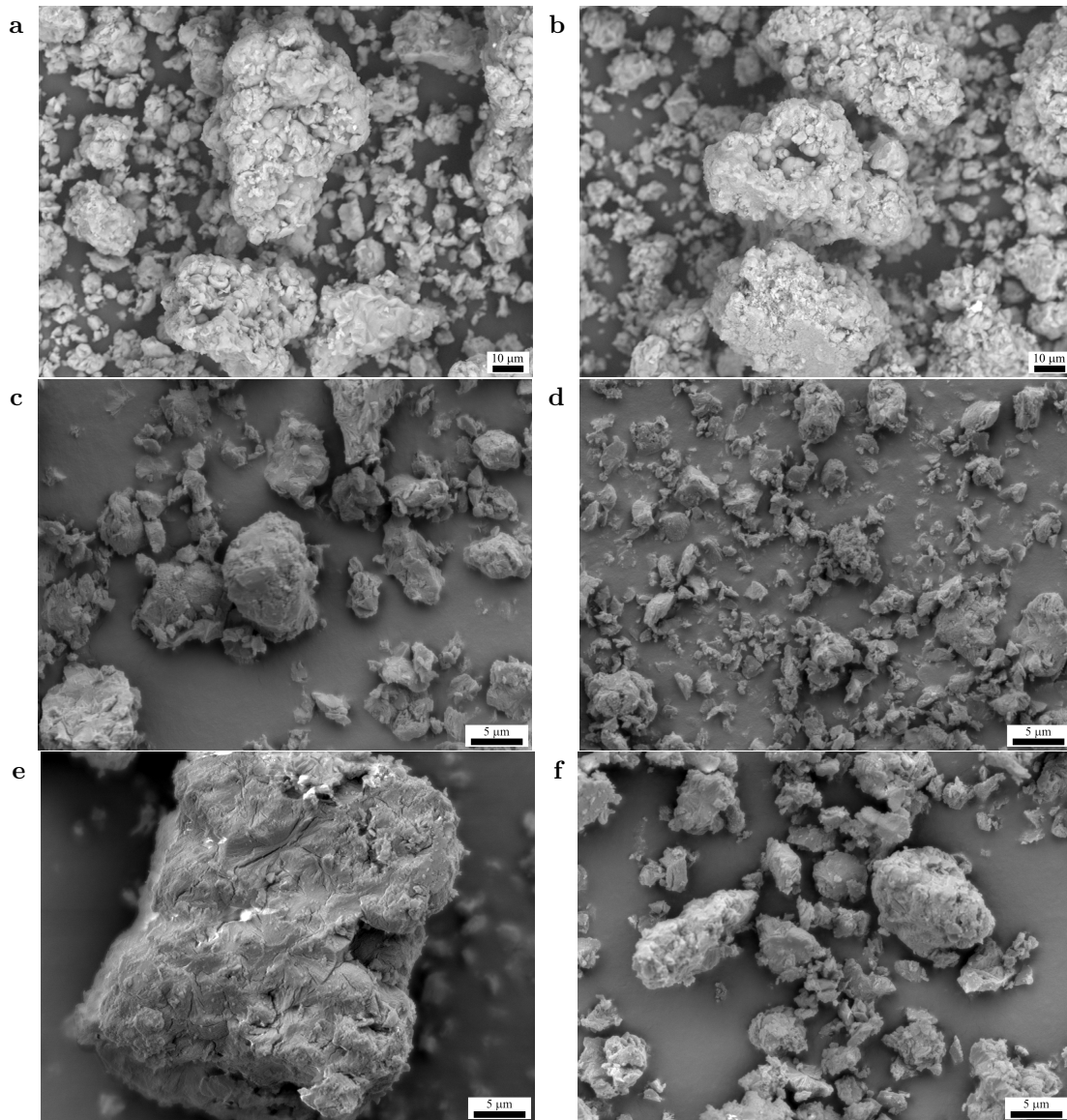


Figure 4.2: SEM images of wet ball milled secondary iron phosphate at 200rpm for (a,b) 1hr, (c,d) 2hr, (e,f) 3hr.

4.1.2 Surfactant Assisted Ball Milling

As established in the above section, achieving sufficient breakdown of the original dense secondary particle morphology requires extended milling times, however, prolonged milling times promoted agglomeration driven by the increased surface energy of the newly formed nanoparticles. The addition of a surfactant addresses this by adsorbing onto freshly milled particle surfaces, reducing their surface energy and providing particle dispersion at submicron sizes. The two surfactants tested were myristic acid and glycerol as mentioned in section 3.2. High rpm values with moderate times were used to ensure sufficient breakdown of the original particle morphology and size. The glycerol additions produced limited improvements over the wet milling conditions. The low weight percent glycerol addition yielded marginally better particle size reduction compared to the high weight percent addition, though

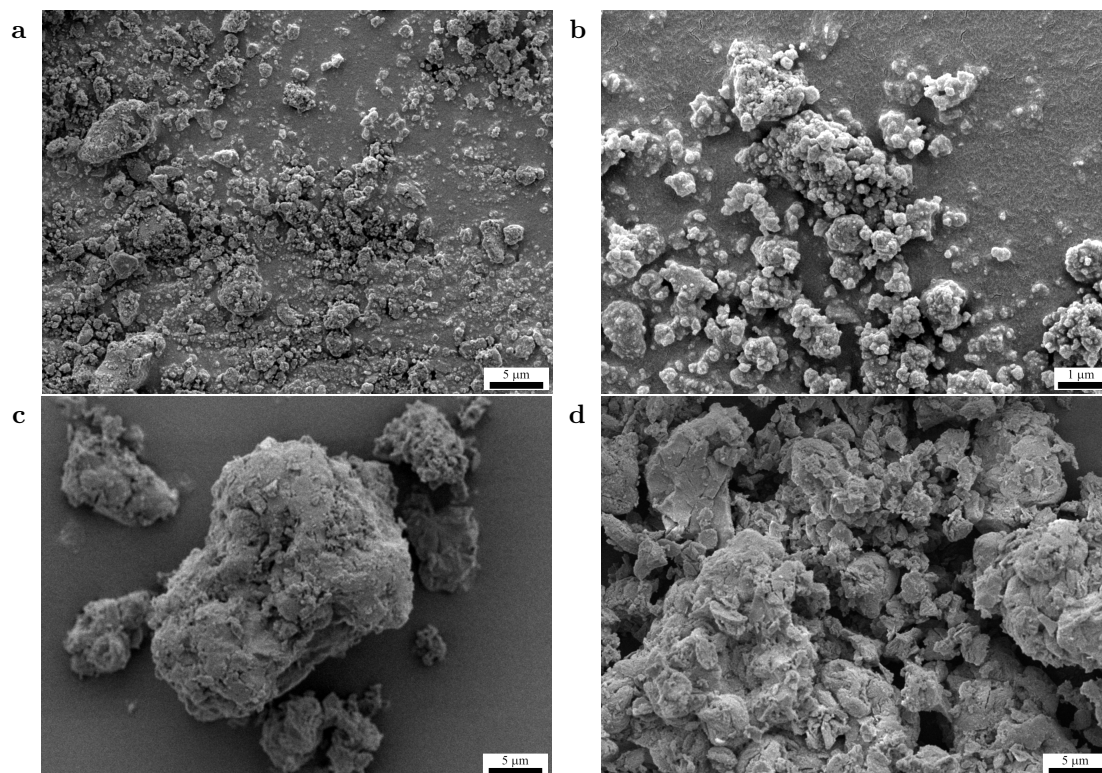


Figure 4.3: SEM images of surfactant assisted ball milled secondary iron phosphate (a,b) 5 wt% and (c,d) 200 wt% glycerol.

significant agglomerations remained in both cases. The high weight percent glycerol addition produces the most severe agglomeration of all four surfactant conditions tested, with some agglomerated structures exceeding 10 μm in size.

The myristic acid additions produced markedly superior results, achieving far greater particle size reduction while substantially suppressing agglomeration compared to both glycerol additions. The 5 wt% myristic acid addition yielded fewer and smaller agglomerates than the 10 wt% addition. At higher weight percentages, the myristic acid molecules may preferentially interact with each other rather than adsorb onto the iron phosphate particles, resulting in lower surface coverage of newly formed nanoparticles. Of the conditions evaluated, the 5 wt% myristic acid addition produced the greatest degree of particle size reduction while most effectively suppressing agglomeration, and was therefore selected as the optimal milling condition for subsequent processing.

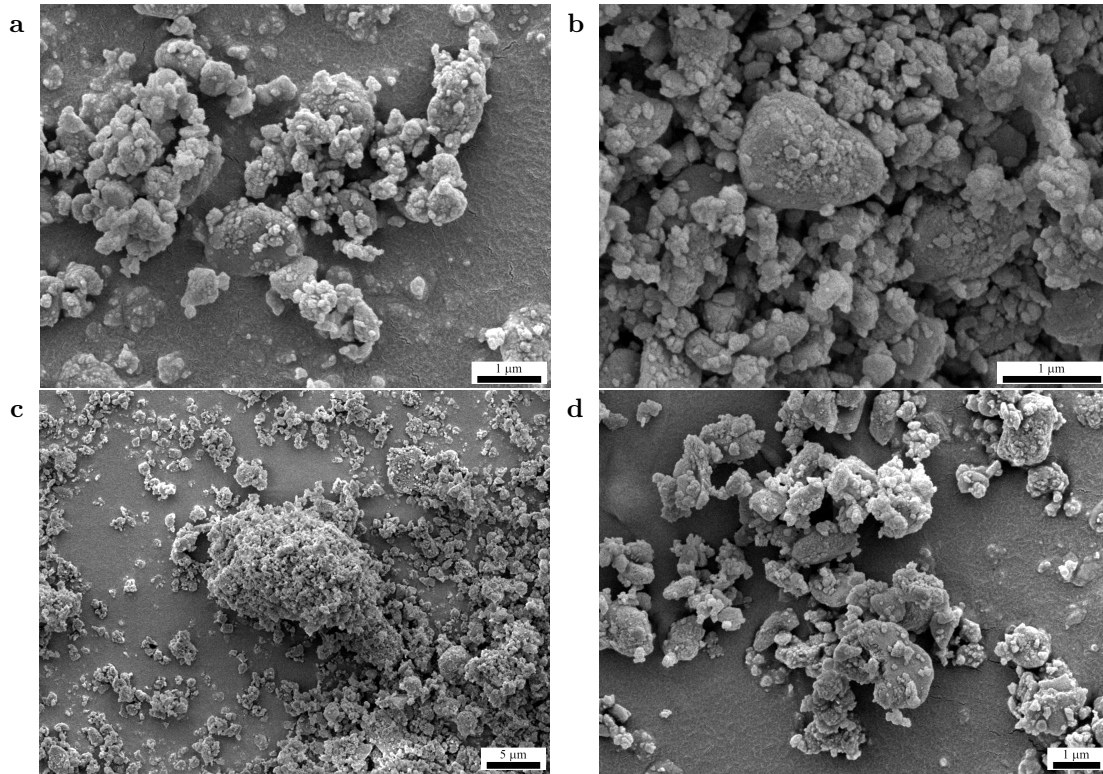


Figure 4.4: SEM images of surfactant assisted ball milled secondary iron phosphate (a,b) 5 wt% and (c,d) 10 wt% myristic acid.

4.1.3 Particle Size Distribution

The particle size distributions of secondary iron phosphate before and after surfactant assisted ball milling (SABM) with 5 wt% myristic acid were determined using the Cellpose machine learning segmentation method described in Section 3.3 and presented in Figure 4.5 and Table 4.1.

Sample	Counts	D ₁₀ [μm]	D ₅₀ [μm]	D ₉₀ [μm]
Pre-BM	343	1.78 ± 8.4%	3.02 ± 4.3%	4.13 ± 3.4%
Post-BM	3341	0.121 ± 2.9%	0.484 ± 5.4%	1.11 ± 2.7%

Table 4.1: Particle size distribution characteristics for pre- and post-ball milled samples, including D₁₀, D₅₀ (median), and D₉₀ percentiles, together with total particle counts.

The pre-milling material exhibited a D₁₀, D₅₀, and D₉₀ of 1.78, 3.02, and 4.13 μm respectively, while the post-SABM material yielded values of 0.121, 0.484, and 1.11 μm, representing a sixfold reduction in median particle size. The narrowing of both the D₁₀ and D₉₀ values further confirms that myristic acid assisted milling conditions were effective in achieving a more homogeneous, sub-micrometer particle size distribution.

It should be noted that the particle size data obtained through machine learning assisted segmentation carries inherent limitations that warrant cautious interpretation.

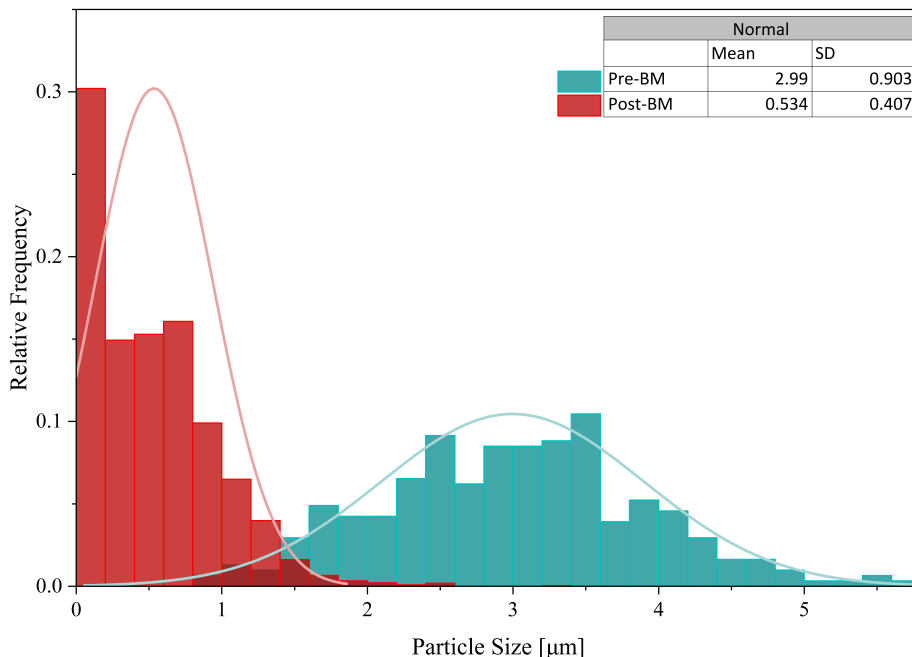


Figure 4.5: Particle size distribution histogram.

The Cellpose algorithm occasionally experienced difficulty in resolving boundaries between adjacent or overlapping particles, particularly in the post-milling samples, where the finer particles tend to cluster and stack upon one another. In such cases, the algorithm may segment a cluster of particles as a single large feature, resulting in an overestimation of particle size. The reported distribution values should therefore be regarded as indications of relative trends rather than absolute values. Nevertheless, the data provides quantitative evidence of the effectiveness of surfactant assisted ball milling with low weight percentage myristic acid, supporting the qualitative observations made from the SEM images.

4.2 Fourier-Transform Infrared Analysis

Fourier-transform infrared (FTIR) spectroscopy was used to confirm the successful removal of myristic acid from the ball milled iron phosphate following the washing procedure described in Section 3.2. As the addition of 5 wt% myristic acid was identified as the optimal milling parameters in the preceding section, FTIR analysis was done exclusively on this sample.

The FTIR spectrum of the post-wash $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$ showed no detectable absorption bands attributed to the stretching of sp^3 C-H bonds, which are clearly present in both the neat myristic acid reference spectrum and the prewash ball milled iron phosphate spectrum, confirming that the washing procedure was effective in removing the surfactant below the instrument detection limit.

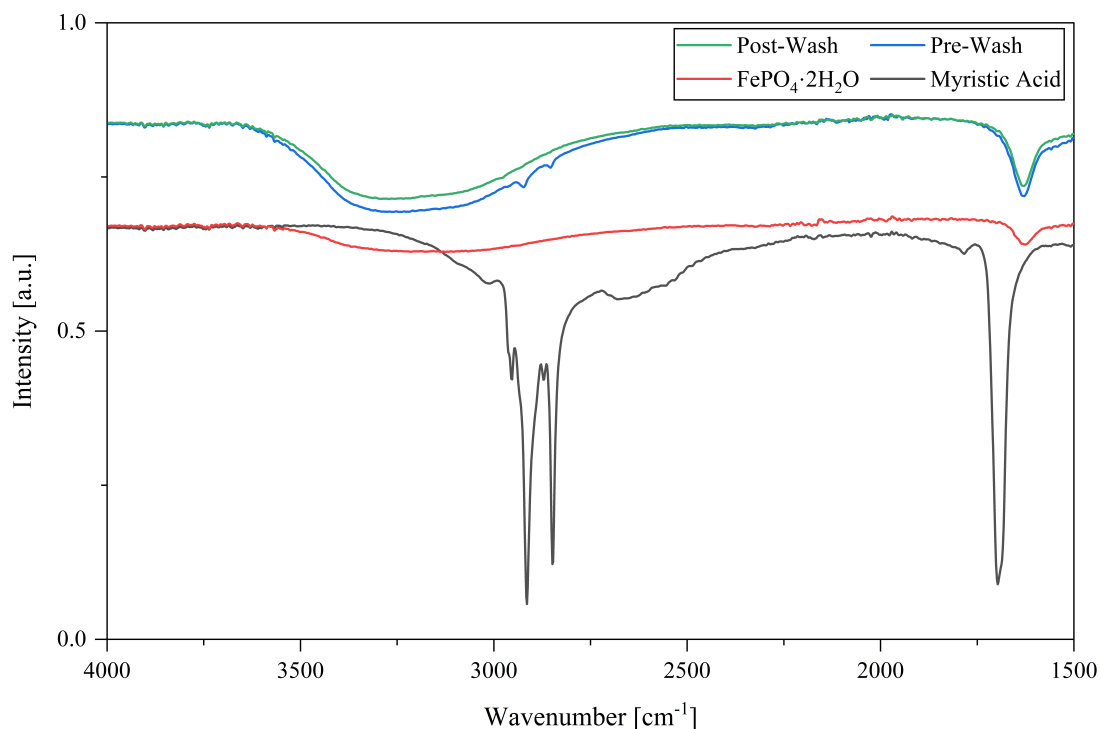


Figure 4.6: FTIR spectra of myristic acid (gray), iron phosphate dihydrate (red), and SAHEBM iron phosphate pre (blue) and post (green) washing with ethanol. The post washing sample showed no detectable signal for the sp^3 C-H bonds within the myristic acid FTIR signal between 3000 and 2800 cm^{-1} .

4.3 X-Ray Diffraction

The XRD patterns of the LFP samples derived from swarf, both from the unmodified and washed swarf, were compared with the calculated reference pattern for phase-pure LiFePO_4 , with the collected patterns presented in Figure 4.7. The primary diffraction peaks of both samples align well with the calculated pattern, confirming the successful formation of LiFePO_4 .

The recycled LFP pattern exhibits additional impurity peaks at $2\theta = 10.7^\circ$ and potentially 9° , corresponding to Li_3PO_4 . This is a common impurity, though one that should be avoided as it can hinder the specific capacity of the material. To eliminate the Li_3PO_4 impurity from the recycled LFP sample, subsequent synthesis attempts were made with adjusted stoichiometric ratios. As the presence of Li_3PO_4 indicates a potential excess of Li, the ratio of $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$ increased to 1.05 and 1.15 respectively. However, this failed to eliminate the phase impurity. Additionally, the presence of Li_3PO_4 without the presence of iron-based impurities could be the result of increased phosphorous ratio within the $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$ precursor. An additional synthesis attempt was made adding $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ to the reaction mixture in the ratio of $\text{FePO}_4 \cdot 2\text{H}_2\text{O}:\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}:\text{LiOH} \cdot \text{H}_2\text{O}$ 1:0.05:1. Adding iron nitrate to the reaction mixture resulted in an amorphous XRD pattern that was difficult to

refine.

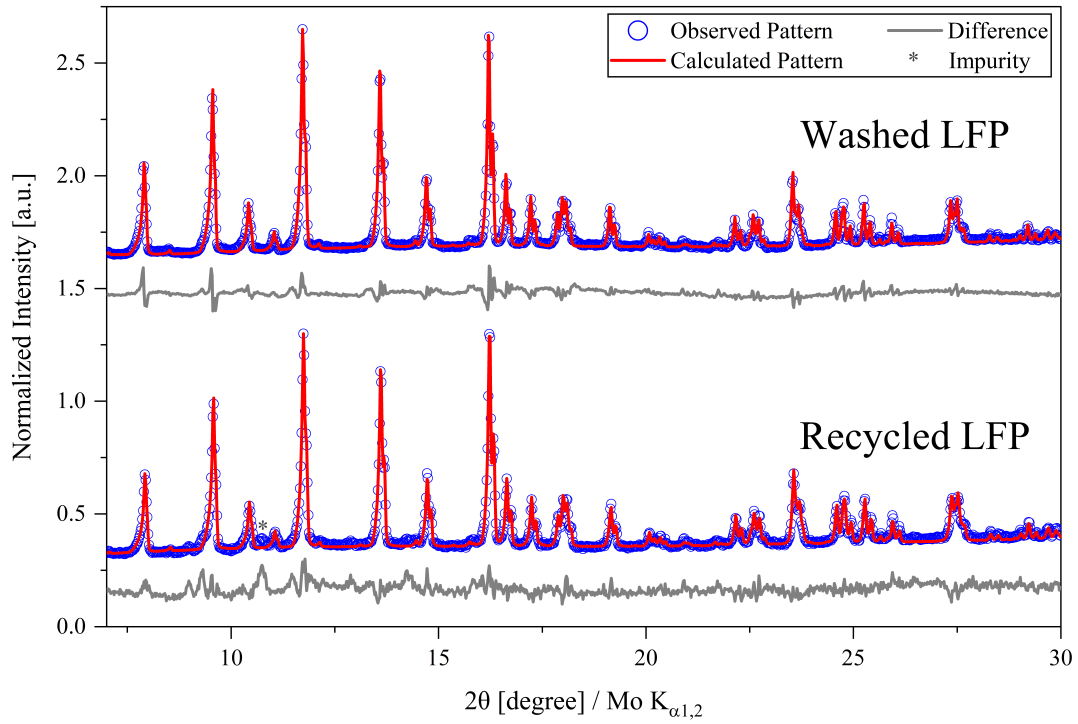


Figure 4.7: XRD spectra of recycled LFP samples derived from washed (upper) and unwashed (lower) swarf. Blue data points represent observed pattern from the diffractometer; the red lines are the calculated spectrum for LFP with impurity peaks labeled with grey stars and respective difference curves represented with the grey line.

4.4 Battery Cycling

The electrochemical performance of the synthesized LFP cathodes was evaluated through rate capability testing at varying C_{rates} and long-term cycling to assess both specific capacity and capacity retention over extended cycling. The theoretical capacity of LFP cathodes is 170 mAhg^{-1} , which in practice is difficult to achieve due to low inherent electronic conductivity and restricted lithium movement within the crystal structure [17]. Instead, typical values for LFP cathodes range from 120-160 mAhg^{-1} depending on various factors ranging from carbon coating content and type, to particle size [17].

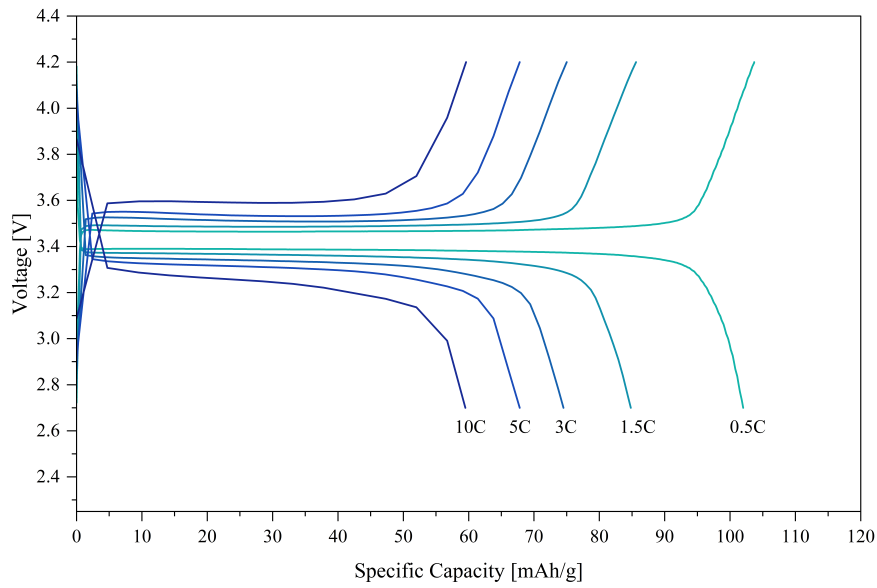


Figure 4.8: Rate Capability curves for washed swarf derived LFP.

C_{rate}	Spec. Cap. [mAh/g]
0.5C	102
1.5C	84.8
3C	74.5
5C	67.8
10C	59.5

Table 4.2: Specific capacity values for varying C-rates of washed swarf derived LFP.

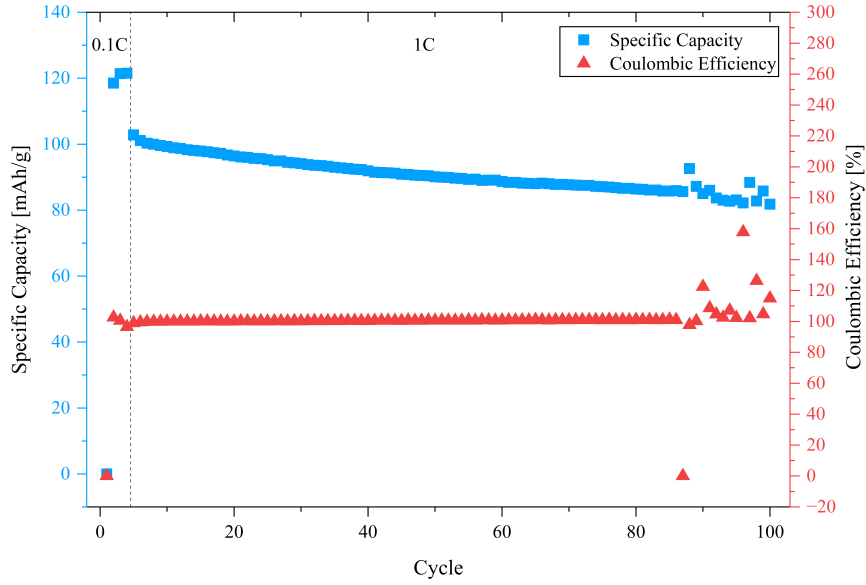


Figure 4.9: Long cycling stability plot for washed swarf derived LFP.

The washed secondary LFP cathode yielded a specific capacity of 102 mAhg^{-1} at a rate of 0.5C , which falls below the typical performance of commercially available LFP. Despite the reduced specific capacity, the long-term cycling stability of the washed secondary LFP cathode was encouraging. A capacity retention of 85 mAhg^{-1} from an initial 100 mAhg^{-1} was observed after approximately 90 cycles as seen in Figure 4.9. However, this also falls short of the commercial benchmarks as commercial LFP cathodes are typically known for lasting hundreds of cycles with little capacity loss [17].

The poor specific of our cathode is likely due to an inhomogeneous carbon coating, since the coating procedure was originally developed for the un-ball-milled iron phosphate morphology. Ball milling reduced the particle size and increased the material's total surface area, which may have led to uneven or insufficient carbon coverage on the milled particles. This limited coverage would hinder electronic conductivity and reduce the specific capacity.

5

Conclusion

This thesis investigated the feasibility of synthesizing lithium iron phosphate cathode material from swarf-derived iron phosphate, primarily through the lens of particle size and morphological control of the precursor material through ball-milling. With the aim of contributing to the development of regionally sourced LFP precursor feedstock less susceptible to geopolitical risks.

The synthesis route was successfully demonstrated as a proof of concept. The dense spherical morphology of the received secondary iron phosphate was effectively broken down through surfactant assisted high energy ball milling, with 5 wt% myristic acid identified as the optimal milling condition, achieving a reduction in median particle size from approximately 3 μm to approximately 0.5 μm . Subsequent removal of the myristic acid through alcohol washings was confirmed by FTIR. Successful synthesis of LiFePO_4 was confirmed by Rietveld refinement of XRD data though Li_3PO_4 impurities were present in the unwashed swarf sample.

The washed secondary LFP cathode, yielded a specific capacity of 102 mAhg^{-1} at 0.5C, falling below the typical commercial range of 120-160 mAhg^{-1} . The long-term stability of the washed secondary LFP was encouraging, with a capacity retention of approximately 85% after roughly 90 cycles.

Overall, this work demonstrates the feasibility of turning swarf waste into usable cathode materials and provides manufacturers with a potential feedstock. The performance gap compared to commercial LFP indicates that further optimization of this pathway is needed before secondary LFP can be considered competitive. Alternatively, other size and morphology control pathways should be investigated, such as pH-controlled precipitation to produce nanometer scale particles or spray drying to control the size and morphology of the secondary particles.

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A

Appendix 1

A.1 Example MATLAB Code for Particle Size Determination

```
1 %Determination of appropriate cell threshold and input
   cell diameter
2 clc
3 clear all
4 close all
5 img=imread('image.tif') %Reads in image
6
7 cp=cellpose(Model='cyto2') %Sets the cellpose model
8
9 %Varying cell thresholds, range of -6 to 6 could be used
   but would require a double for loop or several more
   individual for loops which causes the program to run
   inexorably slow (at least on my computer)
10 ct0=0;
11 ct1=1;
12 ct2=2;
13
14 %Tested cell diameter range from 10 to 100 in increments
   of 10
15 d=10:10:100
16
17 figure(1);
18 %Starts a tiled layout for easy identification
19 tiledlayout("TileSpacing","compact","Padding","tight");
20 for i=1:numel(d)
21     label = segmentCells2D(cp,img,"CellThreshold",ct0,"
       ImageCellDiameter",d(i));
22     loverlay = labeloverlay(img,label);
23     nexttile
24     imshow(loverlay)
25 end
26 figure(2);
27 tiledlayout("TileSpacing","compact","Padding","tight");
```

```
28 for i=1:numel(d)
29     label = segmentCells2D(cp,img,"CellThreshold",ct1,"
        ImageCellDiameter",d(i));
30     loverlay = labeloverlay(img,label);
31     nexttile
32     imshow(loverlay)
33 end
34 figure(3);
35 tiledlayout("TileSpacing","compact","Padding","tight");
36 for i=1:numel(d)
37     label = segmentCells2D(cp,img,"CellThreshold",ct2,"
        ImageCellDiameter",d(i));
38     loverlay = labeloverlay(img,label);
39     nexttile
40     imshow(loverlay)
41 end
42
43 %This portion calculates the areas of the particles
    according to the determined optimal cell thresholds
    and diameters.
44
45 %Image 1
46 %Optimal cell threshold determined above
47 ct1=0
48
49 %Image scale determined by measuring the amount of pixels
    across the SEM scale bar (micrometer/pixel or
    nanometer/pixel)
50 img1sc=10/267
51
52 %Optimal cell diameters determined above. Two-three
    values are used so both large and small particles get
    counted.
53 img1CD40=40
54 img1CD90=90
55
56 %This sections has the program label the identified
    particles with a mask
57 labels1_40=segmentCells2D(cp,img,"CellThreshold",ct1,"
    ImageCellDiameter",img1CD40);
58 labels1_90=segmentCells2D(cp,img,"CellThreshold",ct1,"
    ImageCellDiameter",img1CD90);
59
60 %This sections returns the areas of the generated masks
    in pixels
61 img1CD40CellAreas=regionprops(labels1_40,"Area");
```

```
62 img1CD90CellAreas=regionprops(labels1_90,"Area");
63
64 %This section converts the pixel areas to micrometer
   using the scale
65 img1CD40AreaMicro=[img1CD40CellAreas.Area]*img1sc^2
66 img1CD90AreaMicro=[img1CD90CellAreas.Area]*img1sc^2
67
68 %Lines 46-67 are repeated for all viable SEM images to
   get sufficient data points. Examples are not included
   for the sake of brevity but follow the same process
   with different data identifiers.
69
70 %Micrometer areas are combined into one array for easy
   modification.
71 TotAreaMicro=[img1CD40AreaMicro,img1CD90AreaMicro]'
72
73 %Particle areas are converted to diameters as it is the
   common metric given to describe particle size
74 TotDiamMicro=2*sqrt(TotAreaMicro/pi)
75
76 %A more adept coder with a more powerful computer could
   certainly condense this code. This format was needed
   for my instrument to run the program within the hour.
```

A.1.1 Images Used to Determine Optimal Cell Thresholds and Input Diameters

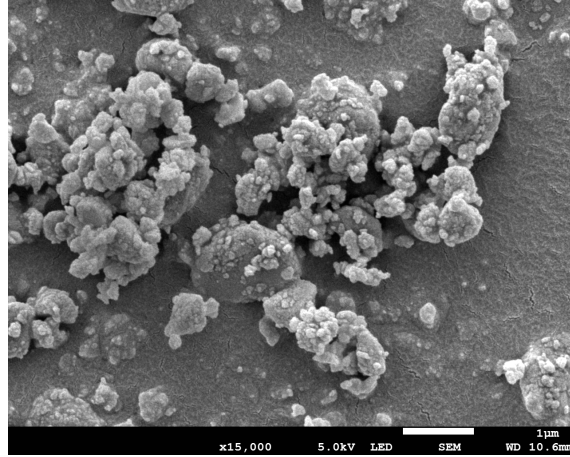


Figure A.1: Example SEM used for particle size determination.

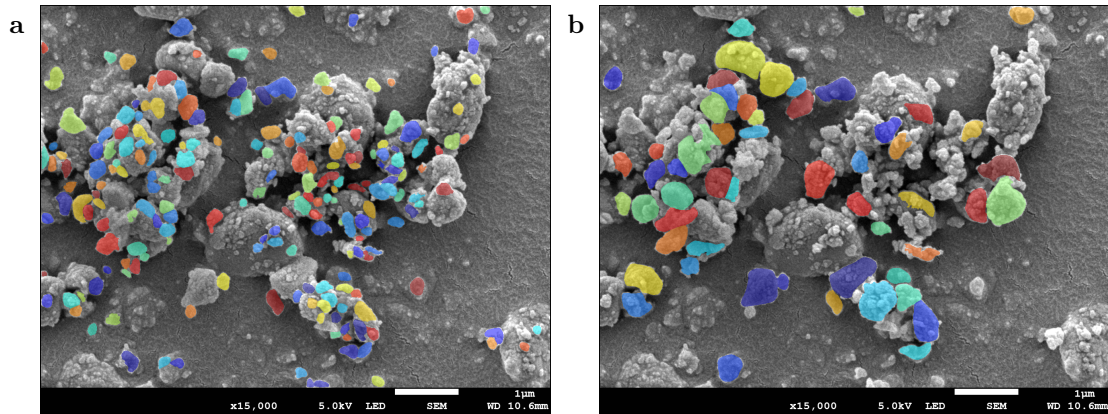


Figure A.2: Example mask images used to determine optimal cell threshold and input cell diameter for particle size determination. Cell threshold: (a,b) 2. Cell Diameter: (a) 40, (b) 90.

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