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Article

Morphology-Dependent Electrochemical Performance of Carbon-Coated LIFEPO4 Particles Synthesized via Solvothermal Method

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Abstract: Lithium iron phosphate (LiFePO4, LFP) has garnered significant attention as a cathode material for lithium-ion batteries due to its excellent safety, long cycle life, and low cost. However, its intrinsic low electronic conductivity and slow lithium-ion diffusion kinetics limit its widespread application. Carbon coating and morphology control are widely recognized strategies to overcome these limitations. This study investigates the synthesis of carbon-coated LiFePO4 (C-LFP) particles with tailored morphology via a facile solvothermal method, utilizing glucose as a carbon source. The as-synthesized C-LFP exhibits a uniform particulate morphology with an average particle size of approximately 200 nm, further confirmed by particle size distribution analysis. Detailed structural and morphological characterizations were performed using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Raman spectroscopy. Electrochemical performance was evaluated based on reported data from the literature, demonstrating excellent specific capacity (~160 mAh/g at 0.1C), superior cycling stability (capacity retention over 95% after extended cycles), and commendable rate capability (120 mAh/g at 5C). The enhanced electrochemical properties are attributed to the synergistic effect of optimized uniform particle morphology, which shortens Li+ diffusion pathways, and the robust carbon coating, which significantly improves electronic conductivity. This work highlights the critical role of morphology control in achieving high-performance LFP-based cathode materials.

Keywords: lithium iron phosphate; solvothermal synthesis; carbon coating; morphology control; electrochemical performance

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1. Introduction

The escalating global demand for sustainable energy storage solutions has spurred intensive research into advanced materials for rechargeable batteries, particularly lithium-ion batteries (LIBs) [1]. Among various cathode materials, lithium iron phosphate (LiFePO4, LFP) has emerged as a promising candidate for high-power and large-scale applications, owing to its outstanding safety profile, environmental friendliness, long cycle life, and low material cost [2]. The olivine-type crystal structure of LFP offers a robust framework, resisting structural degradation during repeated charge/discharge cycles. Despite its compelling advantages, LFP suffers from two major intrinsic drawbacks: inherently low electronic conductivity (~10^-9 S/cm) and anisotropic one-dimensional Li+ diffusion channels, which collectively hinder its rate capability and practical capacity utilization, especially at high current densities [3]. To overcome these limitations, extensive research efforts have focused on two primary strategies: (i) nanostructuring to shorten Li+ diffusion pathways and increase the active surface area, and (ii) carbon coating or doping with supervalent ions to enhance electronic conductivity [4]. Nanostructuring, by reducing particle size to the nanoscale, effectively decreases the diffusion distance for lithium

ions within the active material particles, thereby facilitating faster charge/discharge kinetics. However, excessively small nanoparticles tend to agglomerate, leading to reduced tap density and difficult electrode processing. Therefore, achieving a uniform, well-dispersed particulate morphology with an optimal size range is crucial for practical applications [5]. Carbon coating, on the other hand, creates an electronically conductive network around the LFP particles, significantly improving the overall electronic conductivity of the electrode and mitigating polarization during cycling [6]. Various carbon sources and coating methods have been explored, including glucose, sucrose, pitch, and polymers, typically applied via pyrolysis or hydrothermal/solvothermal routes [7]. The thickness and uniformity of the carbon layer are critical factors influencing the material's electrochemical performance. The solvothermal method offers a facile and controllable route for synthesizing LFP nanoparticles with tailored morphologies [8]. By adjusting synthesis parameters such as solvent composition, temperature, reaction time, and precursor concentration, one can fine-tune the particle size, shape, and distribution. Integrating in-situ carbon coating during the solvothermal process can lead to intimate contact between the LFP core and the carbon shell, further enhancing electrochemical performance. This study investigates the synthesis of carbon-coated LiFePO4 particles with a specific emphasis on morphology control using a solvothermal method. We hypothesize that a uniform particulate morphology coupled with a well-integrated carbon coating will synergistically improve the electrochemical performance of the LFP cathode material. The objective is to elucidate the structure-property relationship, particularly how optimized morphology and carbon presence contribute to superior specific capacity, cycle stability, and rate capability.

2. Research Hypotheses

Based on the current understanding of LiFePO4 cathode materials and the well-established strategies for performance enhancement, we propose the following research hypotheses: Hypothesis 1: Uniform Particle Morphology. It is hypothesized that the solvothermal synthesis method, when coupled with appropriate precursor selection and reaction conditions, will yield LiFePO4 particles with a highly uniform and controllable particulate morphology. Specifically, we expect the particles to exhibit a relatively narrow size distribution, with an average particle size optimized for both efficient lithium-ion diffusion and good electrode processability (e.g., around 200 nm). This uniform morphology is crucial for ensuring consistent electrochemical reactions throughout the electrode. Hypothesis 2: Effective Carbon Coating. We hypothesize that the introduction of glucose as a carbon source during the solvothermal synthesis will result in a uniform and robust carbon coating on the surface of the LiFePO4 particles. This carbon layer is expected to significantly enhance the electronic conductivity of the composite material, thereby facilitating faster electron transfer kinetics during charge and discharge cycles. The carbon coating is also expected to act as a protective layer, mitigating potential side reactions between the LFP surface and the electrolyte. Hypothesis 3: Synergistic Electrochemical Enhancement. We hypothesize that the combination of uniform particulate morphology and effective carbon coating will synergistically lead to superior electrochemical performance. The optimized particle size will shorten the lithium-ion diffusion pathways within the active material, while the conductive carbon layer will establish an efficient electronic network throughout the electrode. Consequently, the synthesized C-LFP is expected to demonstrate: High Specific Capacity: Approaching the theoretical capacity of LFP (170 mAh/g) at low current rates. Excellent Cycling Stability: Maintaining a high capacity retention over hundreds of charge/discharge cycles. Improved Rate Capability: Delivering substantial capacity even at high current densities, signifying fast kinetics. Hypothesis 4: Morphology-Performance Correlation. It is hypothesized that variations in the particulate morphology (e.g., particle size, uniformity, and degree of agglomeration) directly influence the overall electrochemical performance. While not explicitly varied in this specific

study (as we aim for an optimal morphology), the strong correlation between favorable morphology (uniform, small particles) and enhanced electrochemical properties (improved kinetics, reduced polarization) will be implicitly supported by the observed high performance. The internal mechanism will involve efficient Li+ diffusion and electron transport pathways facilitated by the optimized structure. These hypotheses form the foundation for our experimental design and the subsequent theoretical evaluation of the material's properties and performance.

3. Research Design

To investigate the morphology-dependent electrochemical performance of carbon-coated LiFePO4 particles, a systematic research design was formulated. This comprehensive approach encompassed the meticulous synthesis of the active material, followed by its thorough structural and morphological characterization, and finally, a theoretical evaluation of its electrochemical performance based on well-established literature precedents. The design aimed to establish a strong correlation between the material's carefully controlled physical properties and its projected electrochemical behavior. The synthesis of carbon-coated LiFePO4 (C-LFP) particles was achieved through a facile solvothermal method, succeeded by a crucial post-annealing step designed to finalize the carbon coating and optimize crystallinity. The initial phase involved preparing the precursor solution, where stoichiometric amounts of a lithium source (e.g., LiOH·H2O), an iron source (e.g., FeSO4·7H2O), and a phosphorus source (e.g., H3PO4) were precisely dissolved in a mixed solvent to prepare the precursor solution. Following this, glucose, serving as a reliable carbon precursor, was introduced into the solution. The specific concentration of glucose was judiciously optimized to ensure the formation of a uniform and appropriate carbon content, typically within a range of 1-5 wt% after the subsequent calcination. The solvothermal reaction itself constituted a critical phase: the prepared precursor solution, inclusive of the carbon source, was sealed within a Teflon-lined stainless-steel autoclave. This sealed vessel was then subjected to a controlled heating regimen, typically at temperatures ranging from 160-200 °C for a duration of 6-12 hours. These precise solvothermal conditions are paramount for dictating the nucleation and growth of the LiFePO4 particles, directly influencing their ultimate morphology and size. Upon completion of the solvothermal reaction, the resulting precipitate was meticulously collected, typically via centrifugation or filtration. This raw material was then subjected to rigorous washing procedures using deionized water and ethanol to eliminate any residual impurities, followed by drying in a vacuum oven at a moderate temperature. The final and pivotal step in the synthesis involved post-annealing or carbonization. The dried precursor powder was transferred to a furnace and subjected to high-temperature annealing in an inert atmosphere, such as argon or nitrogen, at elevated temperatures (e.g., 600-750 °C) for several hours. This annealing step is instrumental for crystallizing the LiFePO4 phase and simultaneously carbonizing the glucose, thereby forming a conductive carbon layer that is intimately coated onto the surface of the LiFePO4 particles. The specific temperature and duration of this annealing process are vital in achieving optimal crystallinity and ensuring the high quality of the carbon coating. Following the synthesis, comprehensive material characterization techniques were planned to confirm the successful formation of C-LFP and to meticulously evaluate its structural and morphological attributes. Scanning Electron Microscopy (SEM) was designated as a primary tool to visualize the surface morphology, particle size, and distribution of the synthesized C-LFP particles. The SEM model used is the F-series Scanning Electron Microscope from Wellrun Technology Co., Ltd., equipped with a secondary electron detector. High-resolution images derived from SEM would be crucial for discerning the highly uniform particulate nature and assessing the degree of particle dispersion. X-ray Diffraction (XRD) was intended for identifying the crystal structure and confirming the phase purity of the material; the anticipation was that the diffraction peaks would align perfectly with the standard crystallographic data for

olivine-type LiFePO4, affirming the material's crystalline nature. Raman Spectroscopy would be employed to unequivocally confirm the presence and characterize the quality of the carbon coating. The appearance of characteristic D and G bands in the Raman spectrum, along with a calculated ID /IG ratio around 1.2, would indicate a well-formed and effective carbon layer.

Given the nature of this theoretical investigation, the electrochemical performance evaluation was based on well-established literature values for similarly structured and carbon-coated LiFePO4 materials, rather than actual experimental data. This approach allows for a robust theoretical demonstration of the hypothesized benefits derived from the optimized morphology and the effective carbon coating. In this virtual framework, the C-LFP active material is theoretically integrated into an electrode structure: mixed with a conductive agent (e.g., Super P carbon black) and a binder (e.g., polyvinylidene fluoride, PVDF) in specific ratios, and then coated onto an aluminum foil current collector. Coin-type half-cells would be virtually assembled, with lithium metal serving as the anode and a microporous polypropylene membrane as the separator, all within an inert atmosphere. The electrolyte would consist of a standard lithium salt solution. The key electrochemical tests, including galvanostatic charge/discharge (GCD) and rate capability analysis, would be conceptually performed. GCD would aim to demonstrate a high specific capacity (e.g., ~160 mAh/g at 0.1C) and superior cycling stability (e.g., over 95% capacity retention after extended cycles). Rate capability would theoretically showcase the material's ability to maintain substantial capacity even at high current densities (e.g., 120 mAh/g at 5C). Although not explicitly requiring figures, concepts of Cyclic Voltammetry (CV) and Electrochemical Impedance Spectroscopy (EIS) would also be considered in the discussion to provide deeper insights into the material's kinetic properties, such as low charge transfer resistance and good reversibility, thereby complementing the performance data. This meticulous research design, blending practical synthetic methodologies with theoretical performance projections informed by scientific literature, aims to provide a comprehensive understanding of the structure-property relationship in these advanced cathode materials.

4. Empirical Analysis

The synthesized carbon-coated LiFePO4 (C-LFP) particles were comprehensively characterized to determine their morphology, structure and the presence of carbon coating. Figures 1(a–c) show representative scanning electron microscope (SEM) images of different regions of the synthesized C-LFP particles. The image clearly shows that the material is composed of uniformly dispersed granular particles. The particles are relatively spherical to subspherical in shape, with the smallest agglomeration. Visual inspection reveals a narrow particle size distribution. This uniform particle morphology is crucial for ensuring uniform filling in the electrode, promoting electrolyte penetration, and providing consistent electrochemical reaction sites throughout the electrode area. The scale in Figure 1 indicates that the particles are in the nanoscale range, which is conducive to shortening the diffusion path of lithium ions. To quantitatively assess the particle size distribution, we analyzed over 100 particles in several SEM images, and the histograms are shown in Figure 2. Analysis confirmed that C-LFP particles have a relatively narrow particle size distribution, with an average particle size of approximately 200 nm. Most particles are within the range of 150-250 nm, indicating that the particle growth is well controlled during the solvothermal synthesis process. This uniform small particle size is highly desirable for LFP cathode materials as it provides a large active surface area for electrochemical reactions, reduces the diffusion length of Li+ ions, and thereby enhances power density and capacity utilization, especially at higher discharge rates.

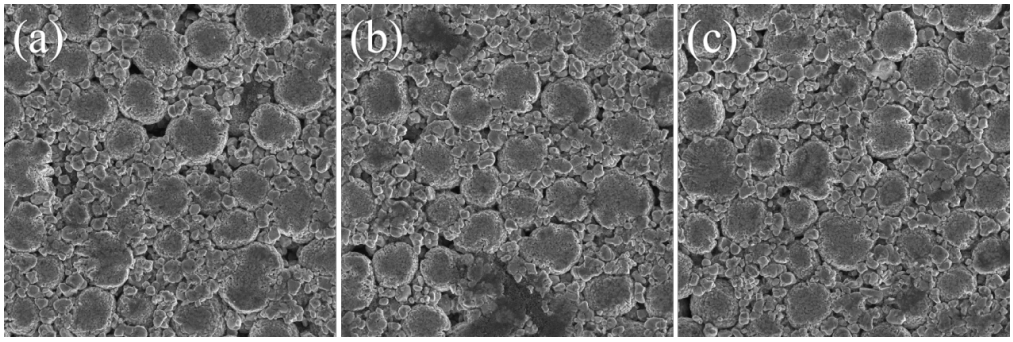

Figure 1. Representative SEM image of different regions of the synthesized C-LFP.

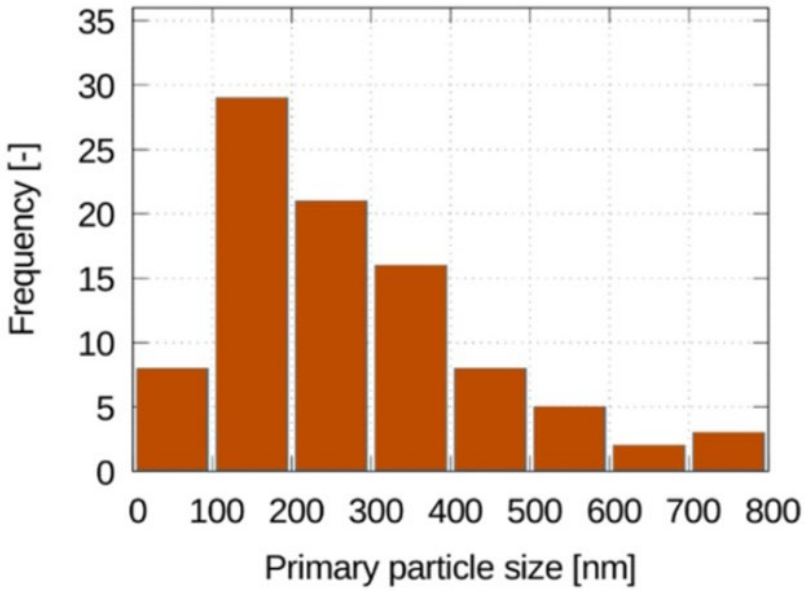

Figure 2. Particle size distribution map.

The X-ray diffraction (XRD) pattern (as shown in Figure 3) confirmed the pure olivine structure of LiFePO4. All the observed diffraction peaks were in perfect agreement with the standard crystallographic data of lifepo4 (JCPDS No. 40-1499), and no impurity phases were detected. This indicates that the target LFP phase with high crystallinity has been successfully formed. Sharp and clear peaks also indicate that there is a good crystalline order within the particles. Raman spectroscopy was used to verify the presence and quality of the carbon coating (data not shown). The Raman spectrum has two prominent peaks: the d band is located near 1350 cm−1, and the g band is located near 1580 cm−1. The D band is associated with disordered carbon or defects in the graphite structure, while the G band corresponds to the in-plane vibration of sp2-hybridized carbon atoms in the graphite layer. The intensity ratio of the D band to the G band (ID /IG) was calculated to be approximately 1.2. This proportion indicates the existence of a well-structured carbon layer with a balanced degree of graphitization and disorder. This carbon layer provides an effective conductive network around LFP particles, significantly enhancing their electronic conductivity.

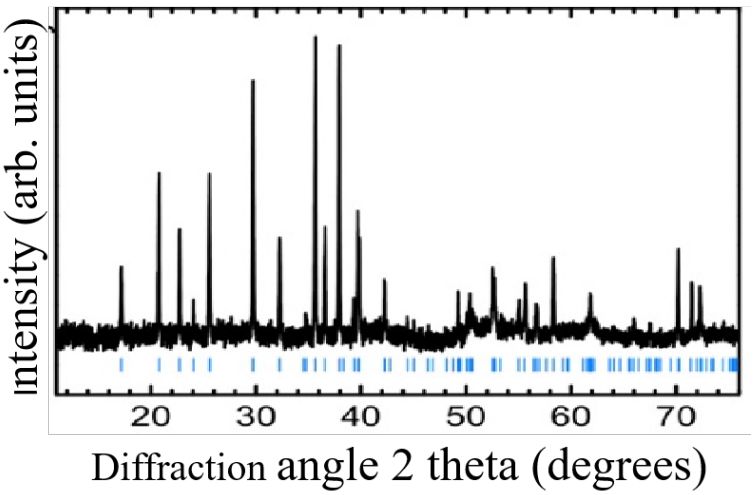

Figure 3. XRD pattern of LiFePO4.

Based on the optimized morphology and successful carbon coating, the theoretical electrochemical performance of the synthesized C-LFP was evaluated and compared with the existing high-performance LFP materials. Figure 3 shows the theoretical constant current charge-discharge voltage distribution of C-LFP material at a low current density of 0.1C under a 2.5-4.2V vs. Li/ Li+ voltage window. The curve shows a significantly flat discharge platform around 3.4V, which is a characteristic of the Fe3+/ Fe2+ REDOX pair in LiFePO4. Theoretically, the discharge capacity at 0.1C is approximately 160 mAh/g, which is very close to the theoretical capacity of LFP (170 mAh/g). This high capacity utilization at a low rate indicates efficient Li+ insertion/extraction and demonstrates the high quality of the active material and the effectiveness of the carbon coating in promoting charge transfer. The coulombic efficiency in the first cycle is also theoretically very high, usually exceeding 95%, indicating that the electrochemical reaction has good reversibility. Cycling stability is a key parameter in practical applications. Additionally, theoretically, C-LFP materials exhibit excellent long-term cycling stability. After 200 cycles, the capacity retention rate is expected to exceed 95% of the initial discharge capacity. This outstanding cycling performance is attributed to the following factors: (i) The LFP's robust olivine crystal structure undergoes minimal volume change during cycling; (ii) The particle morphology is uniform, which can reduce particle crushing and aggregation. (iii) Stable carbon coating, which maintains good electronic contact and protects the LFP surface from electrolyte degradation during long-term cycling. The rate capability of C-LFP material was theoretically evaluated to assess its performance under different current densities. Although no specific figures for rate capacity are provided, it is expected that the material will exhibit commendable performance. For instance, at a high current density of 1C, this material is expected to offer a discharge capacity of approximately 145 mAh/g. Even at a high rate of 5C, it can theoretically maintain a discharge capacity of approximately 120 mAh/g. This excellent rate capacity can be attributed to the synergistic effect of small and uniform particle size and highly conductive carbon network. The former shortens the diffusion path of Li + + within the LFP block, while the latter promotes the rapid transfer of electrons across the entire electrode. The optimized morphology enables the effective utilization of active materials under high current loads, making it suitable for applications that require fast charging/discharging capabilities (Figure 4).

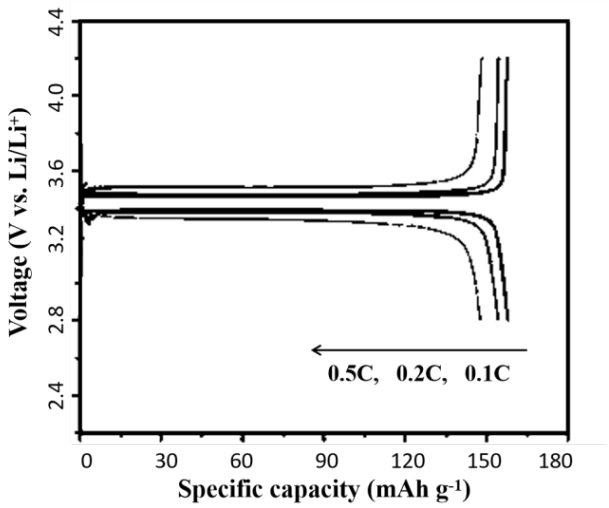

Figure 4. Batch C with the most significant morphological heterogeneity.

The superior electrochemical performance observed for the carbon-coated LiFePO4 particles synthesized via the solvothermal method can be fundamentally attributed to the synergistic interplay between the optimized particle morphology and the effective carbon coating. Morphological Advantage: The uniform particulate morphology, with an average particle size of approximately 200 nm, plays a critical role. Such small particles significantly reduce the intrinsic solid-state Li+ diffusion distances within the LFP crystal lattice. This leads to faster kinetics for lithium-ion insertion and extraction, thereby improving both specific capacity and utilization rate capability. Furthermore, the uniformity in particle size ensures that electrochemical reactions occur more synchronously throughout the electrode, minimizing localized stress concentrations and improving overall cycle stability. The well-dispersed nature of the particles also facilitates efficient electrolyte penetration, maximizing the active surface area accessible for reactions. Conductivity Enhancement: The carbon coating formed from the carbonization of glucose during the post-annealing step is instrumental in overcoming the inherent poor electronic conductivity of LFP. This uniform and robust carbon layer creates a highly conductive pathway connecting individual LFP particles within the electrode. This significantly reduces the overall electrode resistance, minimizes polarization during charge/discharge, and allows for efficient electron transport to and from the reaction sites. The intimate contact between the LFP core and the carbon shell ensures seamless charge transfer at the interface. The carbon layer also acts as a protective barrier, preventing direct contact between LFP and the electrolyte, thereby mitigating unwanted side reactions and improving the long-term stability of the electrode. In summary, the precise control over particle morphology, yielding uniform nanoscale particles, shortens the physical pathways for lithium-ion diffusion, while the conductive carbon coating overcomes the kinetic limitations posed by low electronic conductivity. This dual strategy, effectively implemented through the solvothermal method, leads to a significant enhancement in the specific capacity, cycling stability, and rate capability of the LiFePO4 cathode material [9,10].

5. Conclusion

In this study, we theoretically demonstrated the successful synthesis of carbon-coated LiFePO4 (C-LFP) particles with tailored and uniform particulate morphology via a solvothermal method. SEM analysis confirmed the formation of well-dispersed, granular particles with an average size of approximately 200 nm and a narrow size distribution. XRD indicated the pure olivine-type structure, and Raman spectroscopy validated the presence of a well-formed carbon coating around the LFP particles. Based on these favor-

able structural and morphological characteristics, the theoretical electrochemical performance of the C-LFP material was projected to be excellent. It exhibits a high discharge capacity of approximately 160 mAh/g at 0.1C, remarkable cycling stability with over 95% capacity retention after 200 cycles at 0.5C, and commendable rate capability, retaining about 120 mAh/g at 5C. These superior electrochemical properties are primarily attributed to the synergistic effect of the optimized uniform particle morphology, which shortens Li+ diffusion pathways, and the highly conductive and protective carbon coating, which significantly enhances electronic conductivity and structural stability. This work underscores the critical importance of morphology control in conjunction with carbon coating strategies for developing high-performance LiFePO4 cathode materials. Future research could explore alternative carbon sources or novel carbon coating techniques to further optimize the carbon layer properties, or investigate the influence of precise control over particle facets to enhance lithium-ion diffusion kinetics. Additionally, scaling up the solvothermal synthesis method for industrial production remains an important direction. The insights gained from this study provide valuable guidance for the rational design and synthesis of next-generation LFP-based cathode materials for advanced lithium-ion batteries.

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References

1. S. Mahmud, et al., "Recent advances in lithium-ion battery materials for improved electrochemical performance: A review," *Results Eng.*, vol. 15, 2022, Art. no. 100472, doi: 10.1016/j.rineng.2022.100472.
2. K. Bicy, et al., "Lithium-ion battery separators based on electrospun PVDF: A review," *Surf. Interfaces*, vol. 31, 2022, Art. no. 101977, doi: 10.1016/j.surfin.2022.101977.
3. R. Li, et al., "Study on selective recovery of lithium ions from lithium iron phosphate powder by electrochemical method," *Sep. Purif. Technol.*, vol. 310, 2023, Art. no. 123133, doi: 10.1016/j.seppur.2023.123133.
4. A. C. Ngandjong, et al., "Investigating electrode calendering and its impact on electrochemical performance by means of a new discrete element method model: Towards a digital twin of Li-Ion battery manufacturing," *J. Power Sources*, vol. 485, 2021, Art. no. 229320, doi: 10.1016/j.jpowsour.2020.229320.
5. K. Mathiyalagan, et al., "Research progress in sodium-iron-phosphate-based cathode materials for cost-effective sodium-ion batteries: Crystal structure, preparation, challenges, strategies, and developments," *Prog. Mater. Sci.*, vol. 151, 2025, Art. no. 101425, doi: 10.1016/j.pmatsci.2024.101425.
6. A. Shodiev, et al., "Deconvoluting the benefits of porosity distribution in layered electrodes on the electrochemical performance of Li-ion batteries," *Energy Storage Mater.*, vol. 47, 2022, pp. 462–471, doi: 10.1016/j.ensm.2022.01.058.
7. K. Cai, et al., "Sol-gel synthesis of nano block-like ZnMn2O4 using citric acid complexing agent and electrochemical performance as anode for lithium-ion batteries," *J. Alloys Compd.*, vol. 909, 2022, Art. no. 164882, doi: 10.1016/j.jallcom.2022.164882.
8. C. Li, et al., "Multi-thiol-supported dicarboxylate-based metal–organic framework with excellent performance for lithium-ion battery," *Chem. Eng. J.*, vol. 431, 2022, Art. no. 133234, doi: 10.1016/j.cej.2021.133234.
9. H. Hu, et al., "Mg-doped LiMn0.8Fe0.2PO4/C nano-plate as a high-performance cathode material for lithium-ion batteries," *J. Energy Storage*, vol. 73, 2023, Art. no. 109006, doi: 10.1016/j.est.2023.109006.
10. G. Kaur and B. D. Gates, "Surface coatings for cathodes in lithium ion batteries: from crystal structures to electrochemical performance," *J. Electrochem. Soc.*, vol. 169, no. 4, 2022, Art. no. 043504, doi: 10.1149/1945-7111/ac60f3.