Facile Deposition of the LiFePO₄ Cathode by the Electrophoresis Method

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ABSTRACT: Lithium iron phosphate (LiFePO₄, LFP) is one of the most advanced commercial cathode materials for Li-ion batteries and is widely applied as battery cells for electric vehicles. In this work, a thin and uniform LFP cathode film on a conductive carbon-coated aluminum foil was besieged by the electrophoretic deposition (EPD) technique. Along with the LFP deposition conditions, the impact of two types of binders, poly(vinylidene fluoride) (PVdF) and poly(vinylpyrrolidone) (PVP), on the film quality and electrochemical results has been studied. The results revealed that the LFP_PVP composite cathode had a highly stable electrochemical performance compared with the LFP_PVdF counterpart due to the negligible influence of the PVP on the pore volume and size and retaining high surface area of LFP. The LFP_PVP composite cathode film unveiled a high discharge capacity of 145 mAh g⁻¹ at 0.1C and performed over 100 cycles with capacity retention and Coulombic efficiency of 95 and 99%, respectively. The C-rate capability test also revealed a more stable performance of LFP_PVP compared to LFP_PVdF.
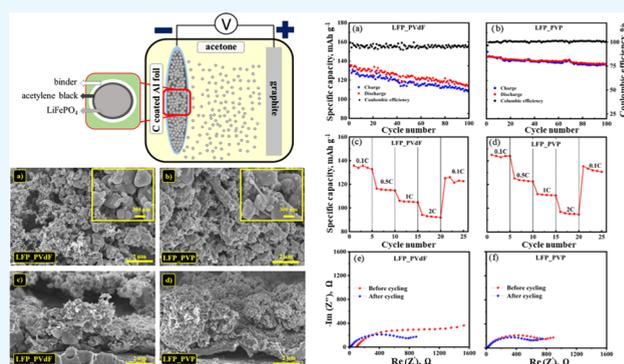
1. INTRODUCTION

Among various types of energy storage systems, the rechargeable batteries are considered one of the most efficient due to their high energy deposition and release rate, high energy and power density, cycling capability, longer life span, and cost-effectiveness.[1] Particularly Li-ion batteries (LIBs) have gained a significant amount of attention because of their versatility to power advanced consumer electronics as well as plug-in hybrid and electric vehicles. Therefore, LIBs are also expected to be one of the crucial components for the transition from fossil fuel-based electricity generation to renewable energy sources, leading to a cleaner and more sustainable environment.[2] Nowadays, the development of LIBs has attracted even more attention due to an urgent demand for advanced rechargeable batteries to power new application devices. This encourages the development of novel electrode materials for the next-generation Li-based rechargeable batteries. Despite the advances in the fabrication of new active materials, there is another challenge to further adapt and improve these electrode materials for use in different dimensions, forms, and types of batteries.

Presently, various types of positive electrode materials are commercially available, with the most common being LiCoO₂ (LCO), LiFePO₄ (LFP), LiMn₂O₄ (LMO), Li-Ni₁/₃Mn₁/₃Co₁/₃O₂ (NMC), and LiNi₀.₈Co₀.₁₅Al₀.₀₅O₂ (NCA).[3−7] It should also be noted that these active materials

have their own advantages and trade-offs concerning energy density, power capabilities, cost, toxicity, safety, and stability.[8] Among these cathodes, LiFePO₄ is one of the most promising materials with a long cyclability, moderate theoretical capacity (170 mAh g⁻¹), and flat discharge plateau at 3.4 V vs Li⁺/Li.[9] In addition, it has significantly higher thermal and chemical stabilities along with the most efficient cost and environmental benefits. It is also compatible with a wide range of organic solvent-based liquid electrolytes.[10] Low-toxicity LFP is an environmentally benign cathode that brings remarkable benefits upon utilization and recycling of batteries afterward. Along with this, the chemical and electrochemical stabilities, durability, and high performance of the LFP cathode allow its deposition on a current collector by nontraditional casting methods.[11] This is especially beneficial in the case of thin-film materials that enhance active material utilization by facile deposition methods without compromising performance. There are several electrode deposition techniques to prepare thin-film cathodes on a variety of substrates such as chemical

vapor deposition, pulsed laser deposition, atomic layer deposition, and magnetron sputtering.[12−15] For instance, Sugiawati et al. presented a radio frequency sputter deposited thin film of amorphous $LiCuPO_4$ for Li-ion microbatteries.[16] Liu et al. exploited the atomic layer deposition technique to coat LFP.[17] Tiurin et al. applied the same technique to prepare a $LiMn_{1.5}Ni_{0.5}O_4$ cathode.[18] There is a review summarizing research on the physical vapor deposition of cathode materials on different types of substrates.[19] Notwithstanding, the availability of deposition methods to develop cathodes on various substrates require multistep preparation procedures and conditions of an inert atmosphere, high temperatures, and low pressure.[19] Occasionally, the outcome of such harsh conditions could be severe phase transitions of the compounds, resulting in undesired aftermath and performance deterioration.[20,21] Thus, the electrophoretic deposition method could be considered a very promising technique to obtain single-phase thin-film electrodes. The deposition technique is based on the movements of charged particles in a steady colloidal suspension onto a conductive substrate driven by the electric field. This concept has a low cost and is highly efficient because of a facile suspension preparation and basic equipment. The technique does not require specific conditions and sophisticated machinery. Moreover, there are many other advantages of this technique such as simplicity, scalability, and controllability for a high deposition rate. Progress of flexible, one-dimensional (1D), or three-dimensional (3D) batteries require the technique with a facile approach. In addition to its great efficiency and cost-effectiveness, the ability to manipulate micro/nanoparticles in a colloidal solution enables EPD to be the most efficient deposition technique for cathode film growth on planar and nonplanar substrates. The suspension preparation and process parameters can be adjusted to control the particle size, film thickness, and mutual structure of films. The electrophoretic deposition also aids in achieving homogeneous deposits on conductive materials in a single step and may be utilized to coat composite materials as well. High-performance electrodes have been produced via electrophoretic deposition, and their performance was identical to those formed using conventional techniques such as blade casting and slot die.[22−24]

To date, few works have been reported on the electrophoretic deposition method utilizing LFP as a cathode. Michaud et al.[22] used several complex compounds including poly[1-[4-(3-carboxy-4-hydroxyphenylazo)-benzenesulfonamido]-1,2-ethanediyl, sodium salt] (PAZO-Na), and carboxymethyl cellulose sodium salt (CMC-Na) as dispersing, charging, and film-forming agents along with carbon black, respectively.[22] The deposition process occurred at a very high potential of up to 50 V. Moyer et al. (2019) used EPD to deposit LFP on the 3D structures and exploited additional components such as Super C45 powder and a xanthan gum binder.[23] Yet, the resulting electrodes demonstrated sudden capacity fading. Sanchez et al.[24] presented an electrophoretic coating of LFP/graphene oxide on carbon fibers.[24] Dimethylformamide (DMF) and poly-(diallyldimethylammonium chloride) (PDDA; 20 wt % in H_2O) were used for charging particles and fixation agent. A relatively low discharge capacity of 130 mAh g^{-1} was collected with high polarization from the obtained cathode. Despite designing several deposition approaches through electrophoresis, there is still a demand for a facile procedure that

does not require the addition of extra solvents, charging agents, gum binders, and surfactants.

Herein, we report on a facile $LiFePO_4$ (LFP) deposition by the electrophoresis method with only the cathode composition, consisting of active material, conductive carbon, and binders. In addition to the deposition process, the effect of the polymer binders such as PVdF and PVP on the capacity, cyclability, and rate capability of LFP film was also investigated and compared. The deposited thin-film cathode was characterized using various physical−chemical characterization methods.

2. EXPERIMENTAL PROCEDURE

2.1. Materials. $LiFePO_4$ (LFP, MTI Corporation), acetylene black (AB, MTI Corporation), poly-(vinylpyrrolidone) (PVP, Merck), or poly(vinylidene fluoride) (PVdF, Merck) dispersion in acetone in a weight ratio of 0.9:0.05:0.05 was used to prepare a composite cathode via EPD. A carbon-coated aluminum foil was used as a current collector. A Celgard 2400 microporous polypropylene membrane (TMAX Battery) and Li metal were utilized as a separator and an anode, respectively. Commercial 1 M $LiPF_6$ (LPF, Sigma Aldrich) solution in a mixture of ethylene carbonate/ethyl-methyl carbonate/dimethyl carbonate (EC/EMC/DC, 1:1:1 vol %) was used as an electrolyte.

2.2. Electrophoretic Deposition of LFP. The composite cathode dispersion was prepared as follows. First, LFP and AB powders were ground and added to acetone. Then, the suspension was stirred for about 24 h and then ultrasonicated to disperse nanoparticles and avoid particle agglomeration. Further, the binders were added to the suspensions and stirred for about 12 h. The suspension with well-dispersed particles was separated from precipitations for the EPD process. A carbon-coated aluminum foil and graphite kernel were used as cathode and anode substrates, respectively. The distance between the two electrodes was hard-fixed to 2 cm to prevent the short-circuit while also allowing the particles in the suspension to travel a short distance. Prior to the deposition, the substrates were cleaned with diluted nitric acid (HNO_3), then rinsed with distilled water, and dried. The deposition process was conducted at a constant voltage of 30 V for 10 min at room temperature (RT). The final LFP composite film on a carbon-coated Al foil was dried in a vacuum oven for 12 h at 80 °C. The schematic illustration of the EPD process is demonstrated in Figure 1.

2.4. Material Characterization. The crystalline structure of LFP in the composite cathodes was confirmed by X-ray diffraction (XRD, Rigaku SmartLab XRD system). The morphology, structure, and composition of the LFP
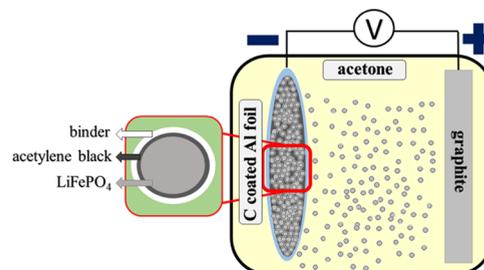
Figure 1. Electrophoretic deposition process illustration of the $LiFePO_4$ composite on a substrate.

composites with different binders were investigated using scanning electron microscopy (SEM, Crossbeam 540).

2.5. Electrochemical Characterization. Cyclic voltammetry (CV), galvanostatic charge and discharge, cyclability, C-rate capability, and electrochemical impedance spectroscopy (EIS) measurements were conducted on a VMP3 potentiostat/galvanostat (Bio-Logic Science Instr. Co.) and a multichannel battery tester (Neware Co.). CV measurements were performed in a potential range from 2.5 to 4.2 V vs Li/Li$^+$ with a scanning rate of 0.1 mV s^{-1}. Cyclability tests were conducted at a lower current density of 0.1C over 100 cycles, while the C-rate analysis was carried out at 0.1, 0.5, 1, and 2C current densities. EIS was conducted to measure the overall resistance of the cells in a frequency range of 1 Hz to 1 MHz. The electrochemical measurements have been conducted with Li metal anode as reference and counter electrodes.

3. RESULTS AND DISCUSSION

The electrophoretic deposition method allowed to uniformly deposit LiFePO$_4$ (LFP) composite cathode on a carbon-coated aluminum foil from acetone-based suspensions using two types of binders, poly(vinylpyrrolidone) (PVP) or poly(vinylidene difluoride) (PVdF). No surfactants or additives were applied to avoid any contamination with inactive components and to develop a facile electrode preparation process. It should also be noted that criteria such as ζ potential, solvent, particle concentration, and conductivity of the suspension were taken into consideration from various sources.[23−25] Despite being an inactive component in a cathode composition, a binder plays a crucial role in obtaining uniform cathode deposition with a desired morphology.[25] PVdF is a widely used binder in the traditional electrode preparation for Li-ion batteries because it is a nonreactive thermoplastic fluoropolymer that is resistive to solvents, acids, and hydrocarbons. In the case of PVP, it is a polymer material mainly used for preparing medical tablets. However, recently, PVP has also acquired compelling consideration as the binder for electrode slurry preparation. Because of its high adhesive property, PVP can significantly influence the uniformity of LFP and AB particles' deposition on current collectors. The process of LFP accumulation is cathodic, meaning that the composite particles in the suspension are positively charged. The thickness of the cathode layers is simply controlled by the deposition time. During the 10 min deposition period, the film thickness and mass of samples were 3 and 5 μm and 2 and 1 mg (1 and 0.5 mg cm^{-2}), respectively, for LFP with PVP and PVdF binders. The XRD patterns of the LFP powder and the as-deposited film are plotted in Figure 2.

The XRD patterns (Figure 2) show that for both PVP and PVdF binder composites, a pure olivine LFP was successfully coated on the carbon-coated Al foil substrate by the EPD method without any degradation. All XRD peaks can be indexed as those of pure LFP, and the additional XRD diffraction peaks are related to carbon and aluminum from the current collector.

The SEM images provided a closer view of the morphology of electrodeposited LFP with different binders (Figure 3). The SEM images clearly show the morphological difference between the LFP_PVdF and LFP_PVP samples, exhibiting a strong effect of the binders on the formation of the cathode deposit during the EPD process. Even though PVdF is the most utilized polymer binder in composite electrodes, its application in EPD unfavorably affected the quality of the
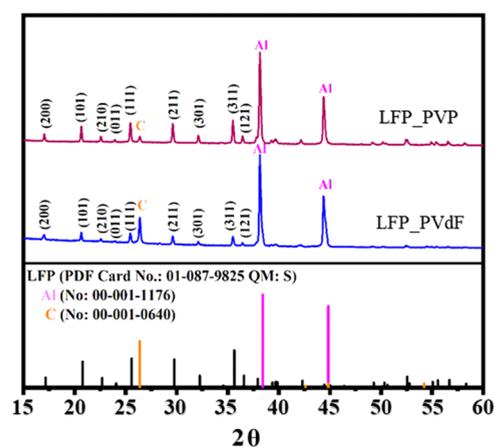
Figure 2. X-ray diffraction patterns of LiFePO$_4$ films with PVdF and PVP binders on a carbon-coated Al foil and reference data.

deposited cathode film. The SEM results clearly indicate that the LFP_PVdF sample did not have a uniform layer on the substrate, leaving larger amounts of voids (uncoated areas) forming a less compact film than that of LFP_PVP (Figure 3a,c). The results of element mapping are presented in Figures S1 and S2 in the Supporting Information.

In contrast, PVP positively influenced the quality of the cathode film, which was formed in a more consistent and compact deposition without any interruptions (Figure 3b,d). Additionally, the contact between the current collector and particles is well established, proving that the PVP binder has higher adhesion, which was also reported previously.[25]

Electrochemical tests of these cathode compositions have delivered quite interesting results. First, the reversibility of redox reactions and electrochemical behavior of LFP by EPD were studied by cyclic voltammetry (CV) at a scanning rate of 0.1 mV s^{-1}. The CV electrochemical response of the samples is depicted in Figure 4a. The CV measurements revealed that LFP by EPD with both PVP and PVdF binders have typical reversible redox reactions related to the Li-ion intercalation and deintercalation processes (Fe^{2+}/Fe^{3+}). The electrodeposited electrodes demonstrated distinct peaks at 3.3 and 3.6 V for oxidation and reduction, respectively. The LFP_PVP composite showed sharp and symmetrical peaks with a small potential difference, proving to have lower polarization. A possible explanation for an enhanced performance could be the morphology of the electrophoretically deposited cathode composite. In contrast, LFP_PVdF has exhibited less intense peaks during the oxidation and reduction processes due to its nonuniform coating with deficient adhesion and compactness.

To evaluate and compare the practical capacity of the electrodeposited LFP electrodes with two different binders, galvanostatic charge and discharge tests were conducted. The initial discharge capacity values of two LFP cathodes at 0.1C have a distinctive difference. The sample with the PVP binder depicted a capacity of 145 mAh g^{-1}, while the one with LFP_PVdF demonstrated a value of 137 mAh g^{-1} (Figure 4b,c). The obtained electrochemical result of LFP_PVP by EPD was identical to that obtained for the traditionally casted LFP in terms of initial discharge capacity value (Figure S3). Both cells performed over 100 cycles at a lower current rate of 0.1C (Figure 5a,b). Yet, a notable difference was detected after 50 galvanostatic charge and discharge cycles. It can be seen that while LFP_PVP showed no significant decline in capacity,
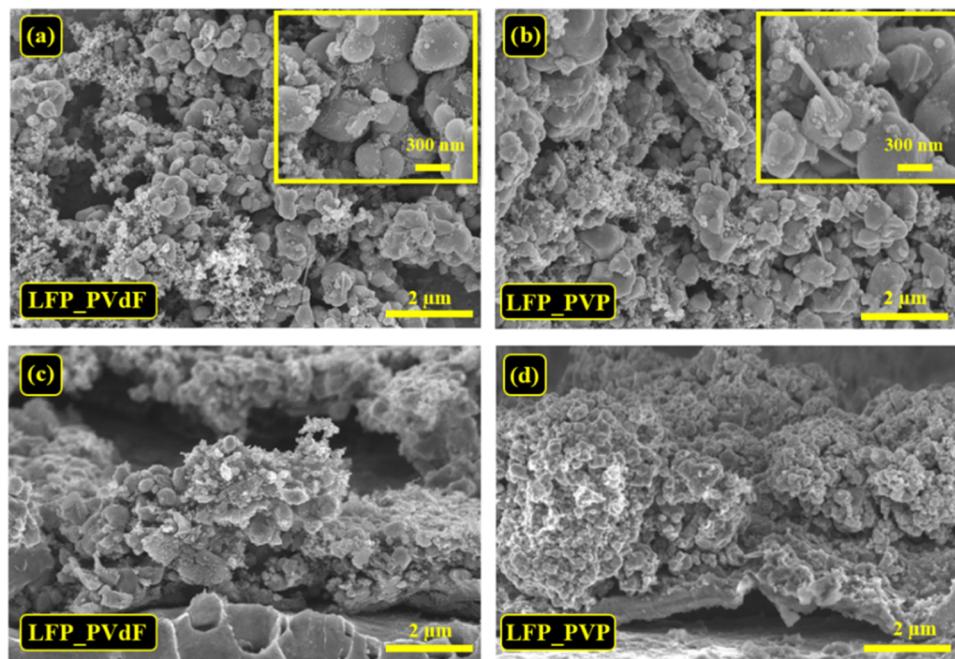
Figure 3. Top (a, b) and cross-sectional (c, d) views of LiFePO$_4$ films with PVdF (a, c) and PVP (b, d) binders.
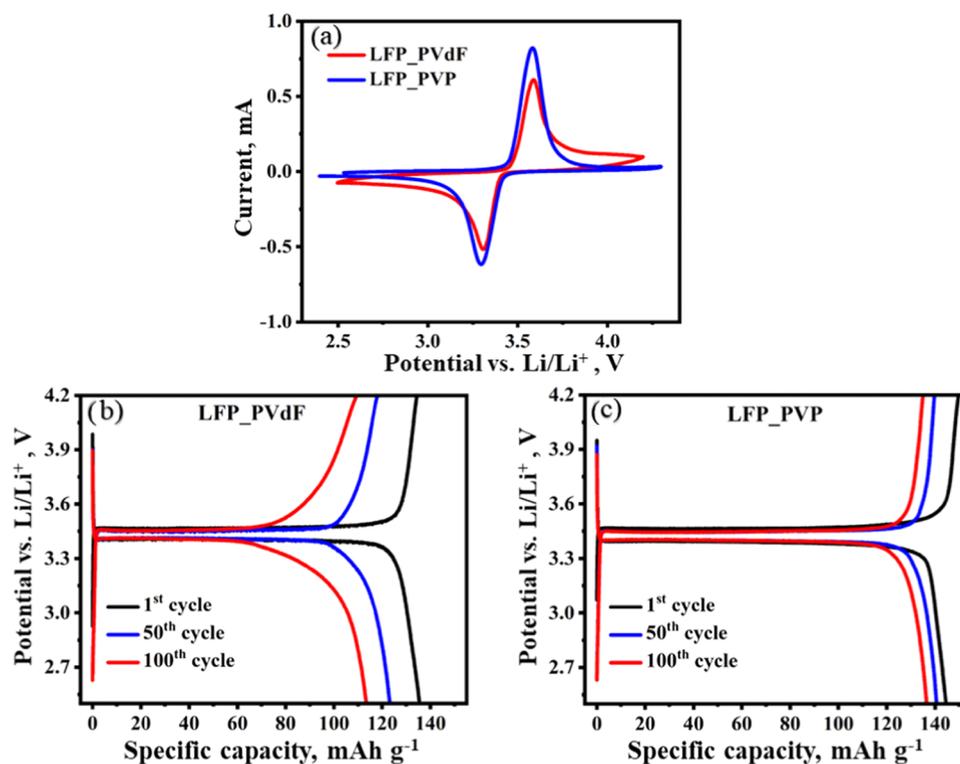
Figure 4. CV profile (a) and galvanostatic charge and discharge profiles (b, c) of LFP_PVdF and LFP_PVP in the half-cell configuration.

LFP_PVdF dramatically lost its capacity after each cycle. The LFP_PVP composite maintained a discharge capacity of around 137 mAh g^{-1} with a capacity retention of ~95%, while LFP_PVdF exhibited performance values of 120 mAh g^{-1} while maintaining only ~87% of its capacity after the 100th cycle. The Coulombic efficiency of these samples was around 99 and 95% for LFP_PVP and LFP_PVdF, respectively. Consequently, it can be disclosed that the reversibility of the redox reaction in LFP_PVP was higher than that of

LFP_PVdF. The performance of the electrophoretically deposited LFP with PVdF and PVP was further tested at different current densities to examine their kinetics, and the results are presented in Figure 5c,d. The rate capability tests also demonstrated greater performance of the electrodeposited LFP_PVP over LFP_PVdF. LFP_PVdF delivered a discharge capacity of 138, 118, 105, and 93 mAh g^{-1} at 0.1, 0.5, 1, and 2C, respectively. When the cell was returned to 0.1C, it maintained only 91% of the initial capacity, showing 125 mAh
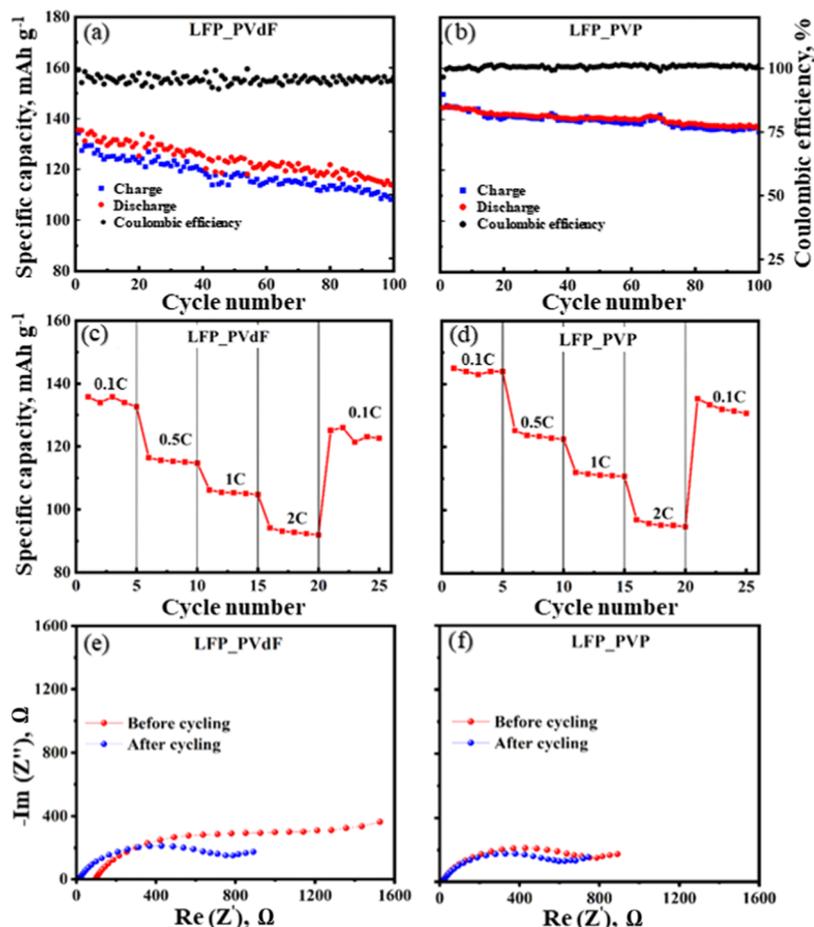
Figure 5. Cyclability (a, b) at 0.1C, performance examination at different C rates (c, d), and electrochemical impedance spectra (e, f) of electrodeposited LFP with PVdF and PVP binders.

g^{-1} with instability in further cycles. In the case of the electrodeposited LFP_PVP sample, the discharge capacities at 0.1, 0.5, 1, and 2C were 145, 125, 110, and 98 mAh g^{-1}, respectively. After the high current tests, the cell showed a steady discharge capacity of 135 mAh g^{-1} at 0.1C, losing only 6% of the initial value.

Additionally, the difference in the electrochemical performance of these two samples was investigated through electrochemical impedance spectroscopy to understand the resistivity trends in the cells and the kinetics of lithium-ion mobility. The examination demonstrated that the cell with the PVdF binder revealed the highest resistivity ($\sim$1200 Ω) before cycling, and it declined after the first cycle ($\sim$800 Ω) (Figure 5e,f). At the same time, the sample of LFP_PVP with identical geometry showed much lower initial resistivity ($\sim$800 Ω), which decreased in the following cycles ($\sim$600 Ω). The superior electrochemical performance of LFP_PVP over LFP_PVdF can be attributed to lower intrinsic resistivity, which enabled a higher ionic conductivity due to large pore volume and specific surface area that was enhanced by PVP, while PVdF blocked the access to pore network.[25] This blocking could cause a decline in the electrical and ionic conductivities of the electrode composition. In addition, the adhesion of the electrode composite was improved in the LFP_PVP sample, resulting in a uniform and smooth film that further provided enhanced contact between the particles and substrate, which could be well seen from the SEM images. PVP has a greater

rate of gluing effect on particles that did not compromise the surface area of LFP and obstruct porous carbon. Because of this, enhanced particle-to-particle contact was observed for better electron and ionic conductivity between particles.

It also should be taken into consideration that the facile deposition technique of electrophoresis can be applied to other cathode composite materials such as NCM by carefully optimizing the casting conditions (Figure S4). Overall, the work presented a very simple approach to depositing cathode composites with controlled thickness by carefully designing the procedure.

4. CONCLUSIONS

The work demonstrated a facile electrophoretic deposition of an olivine-structured $LiFePO_4$ cathode layer onto the current collector without using any additives, complicated equipment, and specific operating conditions. The effect of two different binders such as PVdF and PVP on the quality of the electrophoretically deposited cathode composition has been examined. Subsequently, the electrochemical behavior of these samples was different. LFP with the PVP binder demonstrated remarkable electrochemical performance compared to the LFP with the PVdF counterpart. A uniform film and an improved electrochemical performance were achieved due to the morphology and compactness of the PVP composite, which ameliorated the transport of ions and electrons between

particles and current collector without compromising the surface area of LFP.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c07937.

SEM EDX images of LFP with PVdF and PVP binders; galvanostatic charge and discharge profile and cyclability of traditional casted LFP with Li metal anode; and cyclic voltammetry of NCM 333 vs Li\Li$^+$ (PDF)

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Notes

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