

Research Article

Complexing Agents on Carbon Content and Lithium Storage Capacity of LiFePO_4/C Cathode Synthesized via Sol-Gel Approach

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Olivine-structured LiFePO_4 faces its intrinsic challenges in terms of poor electrical conductivity and lithium-ion diffusion capability for application to lithium-ion batteries. Cost-effective sol-gel approach is advantageous to in situ synthesize carbon-coated LiFePO_4 (LiFePO_4/C) which can not only improve electronic conductivity but also constrain particle size to nanometer scale. In this study, the key parameter is focused on the choice and amount of chelating agents in this synthesis route. It was found that stability of complexing compounds has significant impacts on the carbon contents and electrochemical properties of the products. At the favorable choice of precursors, composition, and synthesis conditions, nanocrystalline LiFePO_4/C materials with appropriate amount of carbon coating were successfully obtained. A reversible capacity of 162 mAh/g was achieved at 0.2C rate, in addition to good discharge rate capability.

1. Introduction

Olivine-structured LiFePO_4 can reversibly extract/intake lithium ions with a theoretical gravimetric capacity of 170 mAh/g. The reaction occurs through the transformation between LiFePO_4 and FePO_4 at the potential of 3.45 V versus Li/Li^+ dictated by the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox couple. The structural similarity of LiFePO_4 and FePO_4 with the volume difference less than 7% results in an excellent discharge/charge cycling performance [1–4]. However, the FeO_6 octahedrons are separated by the PO_4 tetrahedrons, which intrinsically determines the low electronic conductivity and the large bandgap. Meanwhile, the oxygen atoms arrange in the hexagonal close packing order, which restricts lithium-ion diffusion only along the one-dimensional channels [4, 5]. In the past, over two decades, extensive studies have been conducted towards increasing the electronic conductivity and/or Li-ion diffusion rate in LiFePO_4 and hence its electrochemical performances. Strategies include doping with aliovalent metal ions [6–9], coating with conductive layer [10–12], and reducing the particle size [13–16]. Ravet et al. [10] firstly demonstrated that

the surface coating with nanometer-thick carbon effectively increased the capacity rate capability of LiFePO_4 . Gaberscek et al. [13] later submitted that carbon coating not only effectively increased the electrical contact between the particles but also inhibited the growth of LiFePO_4 particle size. Achieving small and uniformly distributed particle size of LiFePO_4 with the minimum content of carbon coating is one of the targets in the efforts of various LiFePO_4 syntheses.

Solution chemical route has indisputable advantages over solid-state route to synthesize carbon-coated LiFePO_4 in terms of producing homogeneous (down to molecular level) and fine particles with narrow size distribution. Sol-gel is more favorable for its low-cost processing, readiness for mass production, flexibility with doping, feasibility for in situ carbon coating, and so forth. Various kinds of carbonaceous organic compounds like glucose [17] or polymers [18] have been added in sol-gel precursors. In addition, carbon containing lithium and iron sources combined with chelating agents have been reported, including ascorbic acid [6], citric acid [19, 20], glycolic acid [21], ethylene glycol [22], lauric acid [23], and polyacrylic acid [24], just to name a few. Studies

and understanding of the key parameters such as precursor species, environment, and amount of chelating agent and their influences on the microstructure and performances of the LiFePO_4 products have significant values for both fundamental research and practical application. However, it is unclear, after reviewing the public results, how the different chelating agents impact the composition, microstructure, and performances of the final products. For instance, the carbon contents in the reported literatures vary from 1.8 wt% up to 18 wt% as consequence of the experimental variables from synthesis conditions to chelating agents. In this study, we are focusing on clarifying the relationship between the carbon content and lithium storage capacity of the final products through fine-tuning the amount of two common chelating agents, that is, citric acid and ethylene glycol, while other precursors and synthesis conditions are fixed.

2. Experimental

In this study, $\text{LiOH}\cdot\text{H}_2\text{O}$ and $\text{FeC}_2\text{O}_4\cdot 2\text{H}_2\text{O}$ were selected as the sources of lithium and iron. H_3PO_4 and/or $\text{NH}_4\text{H}_2\text{PO}_4$ was used as the source of phosphate ions. The pH value of the precursor solution was tuned via the predetermined H_3PO_4 over $\text{NH}_4\text{H}_2\text{PO}_4$ ratio or addition of ammonia solution. Individual complexing agents, citric acid (CA) and ethylene glycol (EG), and the combination of the two (CA+EG) at different ratios were systematically investigated. Most chemicals were purchased from Sigma-Aldrich except H_3PO_4 and $\text{NH}_4\text{H}_2\text{PO}_4$ which were from Alfa Aesar.

The processing to synthesize LiFePO_4/C powders includes sol-gel formation, drying, and high temperature sintering. Firstly, the chelating agent was dissolved in distilled water at room temperature. Subsequently, $\text{LiOH}\cdot\text{H}_2\text{O}$, $\text{FeC}_2\text{O}_4\cdot 2\text{H}_2\text{O}$, and the phosphorous source solution were gradually added to the chelating solution while stirring. After 1h continuous mixing, the solution temperature was raised to 70°C until a yellowish gel gradually formed. The gel was then transferred to a vacuum furnace preheated at 120°C . After being completely dried, the powders were finely grinded and subjected to sinter at the preset temperatures in a tube furnace with a flowing reducing/inert gas, that is, 5 vol.% H_2 + 95 vol.% N_2 . The ramping rate was set at $5^\circ\text{C}/\text{min}$ and sintering time fixed at 10 hours.

The structure, morphology, and composition of as-prepared powders were characterized with the help of X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and thermal gravitational analysis (TGA). The XRD spectral data were collected using a Bruker D8 Discover X-ray diffractometer with $\text{Cu K}\alpha$ radiation. The operation voltage and current were set to 40 kV and 40 mA, respectively. The scans were performed in the 2θ range from 20° to 70° at a scan speed of 0.5 degrees per minute. SEM/EDS results were obtained on the JEOL Bench-top scanning electron microscope. TGA was performed on a 2050 Thermogravimetric Analyzer (TA Instruments) at temperatures ranging from room temperature to 700°C , at a ramping rate of 5°C min^{-1} .

The synthesized LiFePO_4/C active powders were uniformly mixed with the conductive agent (graphite) and

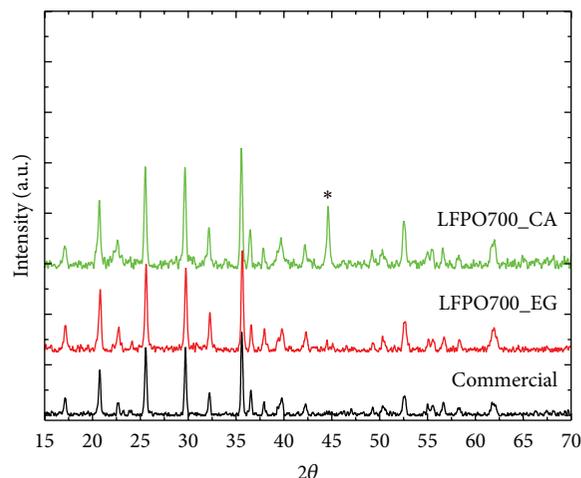


FIGURE 1: XRD profiles of the LiFePO_4 products fabricated using CA or EG chelating agents (chelating to iron precursor molar ratio is 0.5) and sintered at 700°C . The diffraction peak labeled with * indicates the metallic Fe impurity in the corresponding product.

polymeric binder (poly vinyl diene fluoride) at a certain mass ratio with the help of 1-methyl-2-pyrrolidinone (NMP) solvent. Afterwards, the slurry was doctor-blade coated on an aluminum foil and vacuum-dried at 120°C for 12 hrs. Electrochemical testing cells consisting of LiFePO_4 cathode, Li foil anode, and 1M LiPF_6 in EC/DEC electrolyte were assembled in the glove box filled with high purity argon gas. Swagelok cells were used for galvanostatic and cyclic voltammetry studies, while 2032 button cells were assembled for the long-term cycle life testing.

3. Results and Discussion

It was observed from XRD results that specimens start to crystallize at the sintering temperature of 500°C , independent of the choice of chelate type or amount. Upon increasing to 700°C , the amount of olivine structure maximized. Further increasing of the temperature will result in the emergence of impurity phases. For systematic comparison, all the LiFePO_4/C powders in the following studies were synthesized at 700°C while tuning the chelating to iron precursor molar ratios. Figure 1 presents the XRD profiles of the specimens with two different chelate agents synthesized at the 700°C and a commercial LiFePO_4 product for comparison. Apparently, every diffraction peak of the sample synthesized with EG chelate is one-to-one corresponding to the standard XRD pattern, confirming the high purity of the LiFePO_4 product. By contrast, in the product using CA as the chelating agent, the diffraction peak at 45° was constantly observed. The diffraction peak was identified as the metallic Fe. It was further confirmed that the higher the citric acid added in the precursor, the larger the amount of the Fe impurity. It was hypothesized that decomposition of CA during the sintering process resulted in excess reducing environment attributing to the metallic Fe formation, as CA chelate contains three times higher carbon content than EG.

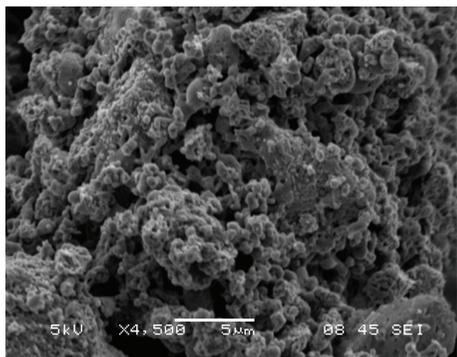
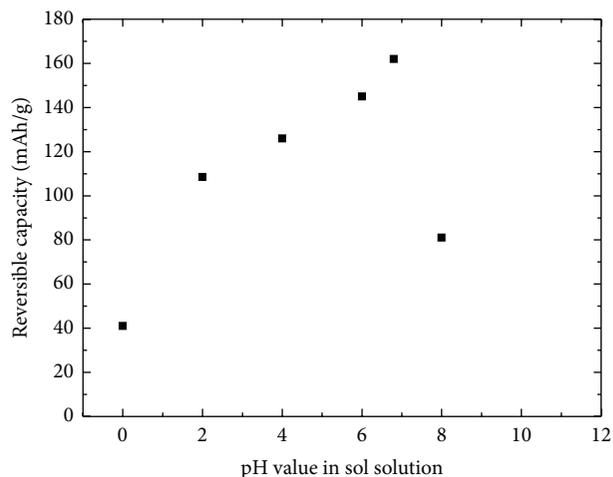


FIGURE 2: A representative SEM image of the as-synthesized LiFePO_4/C powders.

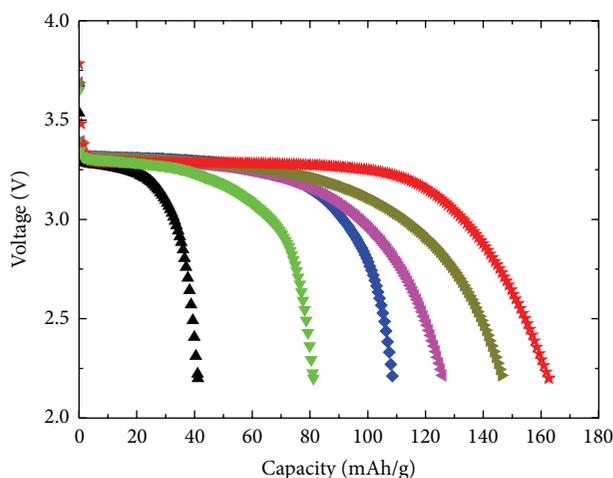
The particle morphologies of the series of LiFePO_4/C powders synthesized at 700°C were characterized with the help of SEM. All morphological images are pretty similar, independent of the precursor composition. Figure 2 exhibits a representative SEM image of LiFePO_4 synthesized with 0.5 molar EG. The product particles are spherical shaped with the size less than 200 nm although they agglomerate in a high magnitude. It is noteworthy that the agglomerate can be easily broken up under ultrasonic dispersion condition during the cathode slurry preparation.

Lithium storage capacity values were found to be sensitive to the pH value in the precursors (see Figure 3). The capacities increased with the increase of the pH value in the acidic case, reached the maximum around $\text{pH} = 6.8$, and then decreased dramatically in the basic environment. The trend was observed for the specimens synthesized with either EG, CA, or CA-EG combination, although the absolute value varied depending on the amount and the type of the chelating agents. The pH value of sol solution affects two primary reactions during the sol-gel process, hydrolysis and polymerization, thereby affecting the complexation between cations and chelating agents. With increasing the pH, the solution viscosity increased significantly and the sol readily transformed into gel. As a result of the rapid gelation, the cation ions and chelating groups are distributed and interact uniformly, leading to homogeneous carbon network and fine LiFePO_4 particles upon sintering. By contrast, as the pH falls into the base region, Fe^{2+} became unstable relative to Fe^{3+} in the presence of oxygen. It is well known that the presence of Fe^{3+} ion has negative impacts resulting in more impurities and low capacities in the final products. It was submitted that pH value in the sol precursor varied the ferrous ions contents during the sol-gel processing, although the starting iron source is ferric oxalate, which eventually affects the properties of the LiFePO_4 .

The carbon content in the products was determined both by EDS and by TGA analyses. TGA profiles showed the mass loss, due to the decomposition of organic matters in the gel, completed around 500°C . The mass of the products was maintained in the range of 44.5% to 50.7% of the initial mass, depending on the type and the amount of chelating complex in the precursors. The weight retention percentage is slightly



(a)



(b)

FIGURE 3: (a) The reversible capacity as a function of pH value in the precursor environment; (b) discharge profile of LiFePO_4 synthesized at the different pH precursor environment.

higher than the computed values based on the assumption that all the extra hydrogen, oxygen, and carbon components were combusted after sintering process. The extra weight was attributed to the carbon content remaining in each product. Table 1 lists the carbon content values obtained from EDS and TGA, as well as the total carbon to iron molar ratio (C/Fe ratio) which is predetermined in the precursors. From Table 1, it is seen that the carbon contents in the products vary in the range of 1.2 wt% to 10.4 wt% and the values derived from TGA data are well corroborated with the EDS analytical results.

It is not surprising to observe that, for single chelating agent in the precursor, the larger the C/Fe ratio, the higher the carbon remnant amount in the products (see series A to C or D to G). When the C/Fe ratio is fixed, the carbon content in the final product also depends on the choice of

TABLE 1: The different molar composition in the precursors results in the variation of carbon content and the reversible discharge/charge capacity of the final products.

Sample	In precursor				C/Fe ratio	In product		Capacity (mAh/g)
	CA C ₆ H ₈ O ₇	EG C ₂ H ₆ O ₂	FeC ₂ O ₄	LiFePO ₄		C (wt%) TGA	C (wt%) EDX	
X			1	1	2:1	N/A	N/A	66
A	0.25		1	1	3.5:1	3.6	3.5	141
B	0.5		1	1	5:1	5.1	4.8	136
C	1		1	1	8:1	10.4	10.0	116
D		0.5	1	1	3:1	1.2	1.0	162
E		1	1	1	4:1	1.8	2.0	152
F		1.5	1	1	5:1	3.6	3.5	145
G		2	1	1	6:1	4.8	5.0	133
H	0.5	0.5	1	1	6:1	6.4	6.6	120
I	0.25	0.25	1	1	5:1	4.2	4.5	141

the chelating agent. For instance, when comparing specimens B, F, and I whose initial C/Fe ratio is 5, the specimen synthesized with the EG chelate (specimen F) has the lowest carbon content, that is, 3.5 wt%. Addition of CA results in increased carbon remnants. For specimen I using EG+CA (1:1 ratio), carbon content increased to 4.0 wt%. Replacing EG with CA, carbon content is much higher, that is, 5.0 wt%. Same trend is also seen by comparing specimen G with specimen H. These results indicate that CA contributes more carbon content remnant than EG. Tuning CA to EG ratio can finely tune the carbon content in the product. This phenomenon is attributed to the different complexing stability between the two chelating agents, which will be discussed more in the following session.

Lithium storage capacity values were found to be a function of carbon content in the products while pH of the precursor was fixed in the range of 6.8 to 7.0. In the present systems, the capacity appears to linearly decrease with increasing carbon content from 1 wt% to 6 wt% and tends to saturate beyond 6 wt% (see Figure 4(a)). Although carbon coating promotes electronic conduction in LiFePO₄, the high carbon content might result in thicker carbon layer [25] on LiFePO₄ which both prohibits lithium effective diffusion into LiFePO₄ and reduces the active material loading. The present experimental results indicated that 1.0 wt% of carbon is sufficient to deliver high reversible lithium storage/removal capacity. Figures 4(b) and 4(c) exhibit typical discharge/charge profiles of the LiFePO₄/C synthesized at the favorite conditions. At first cycle, the discharge capacity reached 150 mAh/g. After 2 cycles, the discharge capacity stabilized at 162 mAh/g (see Figure 4(b)). The initial increase in the capacity of the sample could be associated with the improved wetting of electrolyte, improved interface, and breaking of remaining agglomerate, which synergistically increased the utilization efficiency. Upon increasing the discharge rate, the capacities slightly decreased but there is no obvious reduction of the discharge potential plateaus (see Figure 4(c)). Quantitatively, the discharge capacities just decrease 7 mAh/g as the charge rate increased from 0.1C to 2C. The decreasing amplitude is less than 5%.

TABLE 2: Comparison of reversible capacity, carbon content in LiFePO₄/C products, and the complex stability constants (log β) between chelating agents with iron ions.

Complex agent	Fe state	log β	Capacity (mAh/g)	Carbon content (wt.%)
Hydroxyl	Fe ²⁺	5.7	60	0
	Fe ³⁺	11.9	20	0
Oxalic acid	Fe ²⁺	4.7	66	~0
	Fe ³⁺	9.4	33	~0
Citric acid	Fe ²⁺	15.5	136	5.1
	Fe ³⁺	25.0	95	10.1
Ethylene glycol	Fe ²⁺	9.7	145	3.6
	Fe ³⁺	29.7	75	9.4

Table 2 compares the cumulative equilibrium constant values of complex agents utilized in this study and the two iron ions of different valency [26] and also lists the carbon content and lithium storage capacity of the products with different precursor formulation. Apparently, the iron complexes formed with agent CA have much larger equilibrium constant values than those formed with oxalic acid or EG. The complex equilibrium constant is a direct measure of the stability of the complex which depends mainly on the natures of the central ion and chelating ligand. The larger the complex constant the more stable the complex product. As the complex ability between iron ions and complex agents is stronger, the iron source is more important than the Li source during the sol-gel formation. The stability of complex compound formed with Fe³⁺ is higher than that formed with Fe²⁺ when they use the same complex agent due to the high valency. In the present sol-gel approach, both EG and CA complexing agents act as simple complexing agents which will react with lithium and iron ions. When EG complexes with metal ion, it usually donates the two hydroxyl ligands coordinate with the metal ion in roughly a planar geometry. CA is a ternary hydroxyl carboxylic acid; the structure of the chelate compound is formed by its acid and hydroxyl radicals and the metal ions

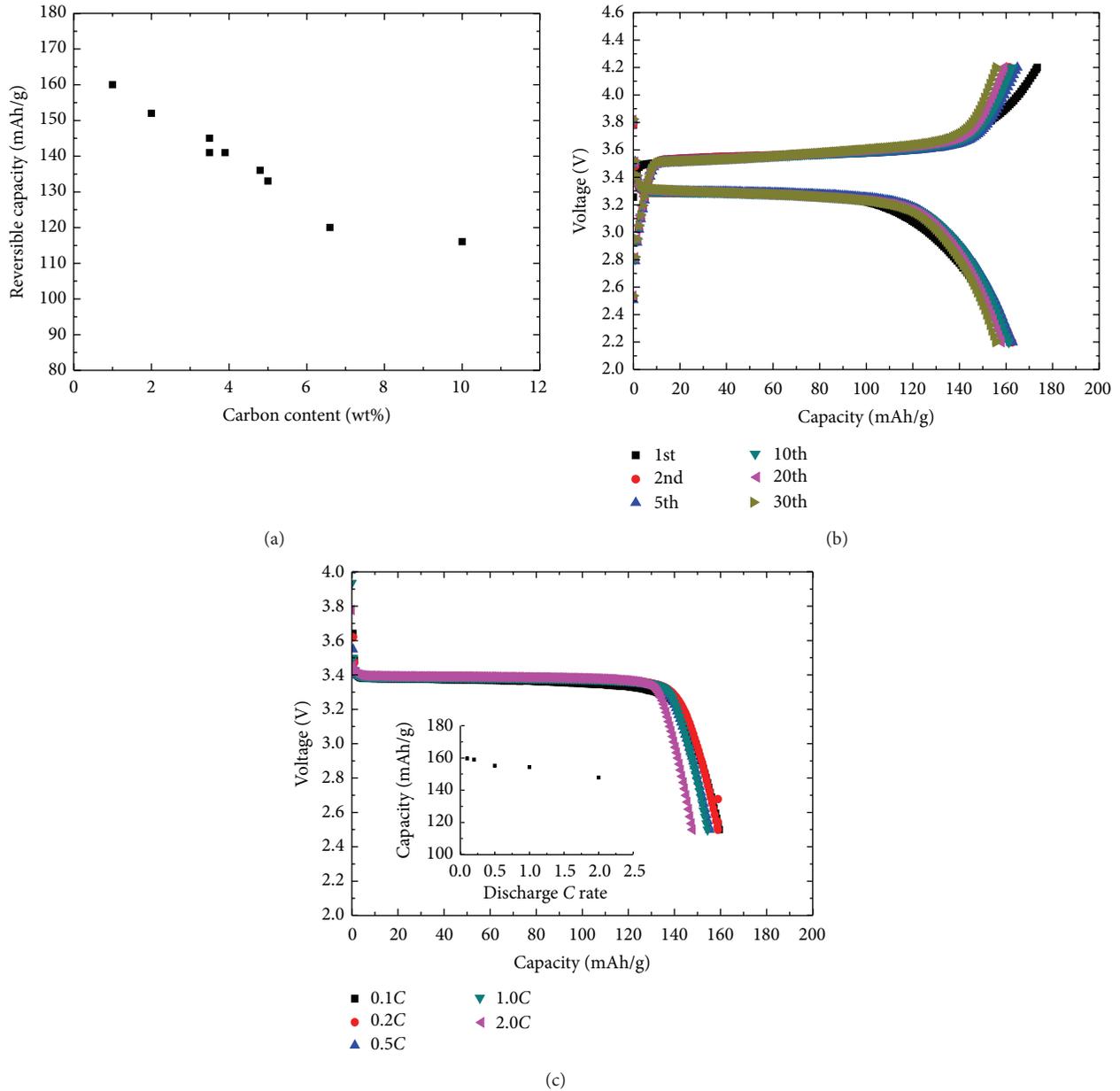


FIGURE 4: (a) The reversible capacity as a function of carbon content; (b) typical discharge/charge profiles of the LiFePO₄/C with 1.0 wt% carbon obtained at 0.2C rate; (c) discharge profiles of the LiFePO₄/C with 1.0 wt% carbon obtained at 0.2C rate obtained at different C rate.

into five or six-membered rings. Stable complex sol results in multiple interwoven ring structure in gelled form which will be relatively difficult to decompose in the sintering process, and, consequently, the proportion of the carbon remaining in the final product is greater. As the complex compound with low stability constant is less stable, a large portion of the carbon frozen in the complex gel is readily released in the form of CO₂ upon combustion at high temperature leaving a small amount coating on LiFePO₄ surface.

The carbon content and the complex stability constants of the complex compounds shown in Table 2 are approximately corroborative. The capacities of LiFePO₄ synthesized with CA or EG chelating agents are inversely related with carbon

content and hence complex stability constants. This finding indicates that the smaller the complex constant, the more beneficial complex compounds would be in order to achieve better electrochemical performances of LiFePO₄. This finding agrees well with the report by Yang et al. who studied LiFePO₄/C prepared with acetic acid, ethanediol, oxalic acid, and ethylenediamine [27]. The stability constant with the carbon containing chelates of choice should be greater than that with hydroxyl group if the synthesis occurs in aqueous solution. As the stability constant is too small, the complex with iron ions is mainly in hydroxide forms. Consequently, no carbon could be retained on the surface of LiFePO₄ and poor performance was seen in this study when only iron

oxalate was used in the precursors with no other chelating agents added. When EG-CA mixed chelating agents are simultaneously added in the precursors, it was observed that the carbon content deviated from mixing principle and was higher than the computed value. This may suggest that CA dominates in the competitive complexing process with iron agents due to its high stability constant.

4. Conclusion

It is found that types and the amounts of chelating agents have significant impacts on the electrochemical performances of LiFePO_4 when all the other experimental factors are controlled to the favorable conditions. The pH value of the sol and hence its viscosity are also critical for rapid complex gelation to ensure uniform distribution. The choice of the complex agent with relatively low complex constant will render easy decomposition in the sintering process and determine the carbon content. Specifically, in this experimental formulation, EG is better than CA for its approximate complex stability constant with ferrous ions and about 1.0 wt% carbon is sufficient for achieving high-performance LiFePO_4/C cathode.

Competing Interests

The authors declare that they have no competing interests.

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