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Review Paper

Recent Development in $LiFePO_4$ Surface Modifications with Carbon Coating from Originated Metal-Organic Frameworks (MOFs) to Improve the Conductivity of Cathode for Lithium-Ion Batteries: A Review and Bibliometric Analysis

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Abstract

Using lithium-ion batteries has emerged as a viable approach to lessen the negative effects of fossil fuel use. LiFePO4 (LFP) is one of the lithium-ion batteries that are eco-friendly and safer than others. However, LFP has a main limitation with the poor rate performance due to its low electronic conductivity number. This study aims to present a bibliometric review of the analysis using VOSviewer of surface modification using carbon coating of metal-organic frameworks (MOFs) to improve the challenge of synthesis, structure, electrochemical stability, and performance of LFP. The results of this study showed that surface modification of LiFePO4 electrodes using carbon compounds produced from MOFs improved the efficiency of electrochemical energy storage and conversion technologies. High levels of porosity and customizable characteristics are offered by metal-organic frameworks (MOFs) ideal for surface modification which improves the battery conductivity. The bibliometric analysis showed that research on lithium-ion batteries is currently receiving attention, a sign of its significance and rising popularity. It is suggested for researchers especially Indonesian researchers to contribute more to this field.

Keywords: Carbon coating; $LiFePO_4$; Lithium-ion batteries; Metal-organic frameworks; Surface modification

1. Introduction

The use of vehicles is now a problem that has a significant impact on the environment and sustainability. The continuous use of energy from fossil fuels is responsible for global climate change due to exhaust emissions from their combustion [1]. In addition, it can lead to various health problems, such as asthma, allergies, cancer, and premature death [2]. Consequently, a fuel that can displace fossil fuels is required. Electric vehicles (EVs) powered by batteries have replaced fossil fuel-driven automobiles in a number of nations [1]. Nowadays, a wide range of vehicles, including cars, trucks, airplanes, and even goods- and passenger-transporting vehicles, use lithium-ion batteries (LIBs) [3].

There are several cathodes of lithium-ion batteries, including $LiFePO_4$ (LFP) [4]. $LiFePO_4$ (LFP) has a lower risk of thermal runaway compared to other cathodes of lithium-ion batteries, such as lithium cobalt oxide (LCO), lithium manganese oxide (LMO), and lithium nickel cobalt aluminum oxide (NCA). Therefore, LFP is a better alternative for applications requiring precise safety standards [5]. In addition, LFP offers a wireless monitoring system that can gather battery data and wirelessly communicate it to notify the end user about the battery status while collecting historical data on the battery profile. By warning the end user of any abnormal conditions, this method can ensure safer battery consumption [6].

$LiFePO_4$ has limitations that mean solutions are being searched for, including inaccurate state-of-charge (SOC) estimates, energy density, and ionic conductivity. Several research has been carried out in order to solve the problem of SOC estimation, such as the construction of an auxiliary particle filter [7], hybrid DNN-KF model [8], and full polynomial parameters-mode [9]. Some experiments have also been conducted to build a dual battery system (DBS) on the use of LFP batteries to increase their energy density [10]. In addition, many previous studies have been conducted to improve its ionic conductivity, such as using additives [11], a cationic exchange approach [12], and carbon coating [13].

Carbon coating has been one of the most effective techniques for increasing LFP's electrical conductivity. It is a great electrical conductor and can be helpful in preventing the formation of a solid electrolyte interface (SEI) layer on the surface of LFP particles, which can degrade battery performance over time [14]. Because of its multifunctionality, one kind of carbon that has been investigated can be utilized to coat LFPs generated from metal-organic frameworks (MOFs) [15]. Xu et al. [16] discovered that carbonization of zeolite imidazolate-8 (ZIF-8) framework synthesized into mesoporous carbon (MC) can improve the ion diffusion coefficient and electronic conductivity of LFP. In addition, Wang et al. [17] have found that MOFs can enhance ion transport in polymer electrolytes that act as ionic liquid-decorated MOFs in solid-state lithium batteries.

In this research, bibliometric methods were employed to identify and analyze an encompassing range of previously published studies. The bibliometric analysis offers valuable insights into the progression of research within a specific domain and provides information on the study limitations and areas necessitating further investigation [18]. This analysis includes information on the most relevant publications, active and cited authors, and the most used keywords in a particular research field [19].

VOSviewer is the most common tool used for bibliometric analysis [20]–[22]. In this case, VOSviewer focuses on accurate visualization, which allows bibliometric maps to be examined in detail. VOSviewer allows the creation of country maps based on networks (co-citation), creating keyword maps based on shared networks, and creating maps with multiple items [19]. Vosviewer has been widely used in bibliometric analysis for various fields, including science. Chen et al.'s [23] research focuses on future research regarding the use of LIB in electric vehicles analyzed using VOSviewer. In addition, Lan et al.'s [24] study also used VOSviewer analysis as part of its bibliometric analysis of current trends and research in fault diagnosis of lithium-ion batteries (LIB). These success studies illustrate the versatility and effectiveness of VOSviewer as a tool for knowledge discovery and trend analysis in battery studies.

Thus, the aim of this study is to provide a comprehensive bibliometrics overview and analysis using VOSviewer (https://www.vosviewer.com/) of olivine's LiFePO4 (LFP) cathode of lithium-ion batteries in the context of enhancement performance by surface modification with carbon-based coating. It is our purpose to report challenges of synthesis, structure, electrochemical stability, and performance, special attention is given to the metal-organic frameworks (MOFs) based carbon as the promising surface coating for the LFP of large-scale Li-ion batteries. This study also summarizes the current status of lithium-ion battery technology, discusses the challenges and opportunities associated with their use in electric vehicles (EVs), and explores potential advancements and future directions for high-power applications.

2. Methods

The research method combines bibliometric analysis with a review of the literature. For bibliometric analysis, the articles were taken from Scopus database (https://www.scopus.com) which were exported on June, 19th 2023 with the keywords "lifepo4" OR "lithium ferro phosphate" OR "LFP" OR "lithium-ion battery" OR "LIB" OR "surface modification" OR "carbon coating". The articles considered for this study were limited to those published in journal type publication from 2002-2022 and in English. The total number of articles used in this study was 2369 documents. Those data were analyzed with VOSviewer (https://www.vosviewer.com/) to visualize the data in order to see the trends of the research.

3. Results and Discussion

3.1. The Basic Cell Configuration

Lithium-ion batteries are composed of three main components based on their function as electrical power generating and storing elements, namely anode, cathode, and electrolyte [25]. However, when considered as a whole, other than packaging, lithium-ion batteries consist of copper (Cu) sheets as current collectors on the anode side, anode material, electrolyte, separator, cathode material, and aluminum (Al) sheets as current collectors on the cathode side [26], [27]. The simplified illustration of an electrochemical battery is shown in **Figure 1**.

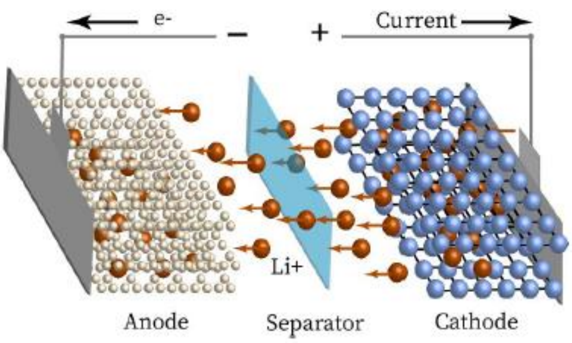

Figure 1. The main components of lithium-ion batteries [25]

3.1.1. The Anode Material of Lithium-Ion Batteries

The anode material in lithium-ion batteries functions as a host, which releases lithium ions to migrate internally to the cathode when the battery is in the process of discharging, along with the release of electrons externally [28]. Thus, electrical devices attached to this battery can be electrically switched on. During the recharging process, electrons are pumped into the anode so that lithium ions are attracted and move to the anode, and the anode material is intercalated with these lithium ions.

The Li+ extraction reaction that occurs in the graphite-based anode during the discharging process is formulated in Eq (1).

$$LiC_6 \rightarrow (1\text{-}x)\ LiC_6 + xC_6 + xLi+ + xe- \quad (1)$$

The Li+ insertion reaction that occurs in the graphite-based anode during the recharging process is formulated in Eq (2).

$$C_6 + xLi+ + xe- \rightarrow (1\text{-}x)\ C_6 + xLiC_6 \quad (1)$$

where $x < 1$

Therefore, when a lithium-ion battery is being discharged and recharged, usually based on graphite which undergoes a dynamic process of lithium extraction and insertion.

3.1.2. The Cathode Material of Lithium-Ion Batteries

The electrochemical performance of lithium-ion rechargeable batteries depends greatly on the cathode material [29]. In the cathode, which made of lithium, frequently used Lithium-iron-phosphate (LFP) as its material [25]. The use of olivine $LiFePO_4$ as a cathode material for lithium-ion batteries is proposed by Goodenough et al. [30]. The Li-ion extraction and insertion reactions work at 3.5 V ($FePO_4$ vs. Li/Li+) at 0.05 mA/cm^2 and are limited to 0.6 Li per formula unit. The reactions are provided on the Eq (3) and (4) [30].

The Li^+ insertion reaction that occurs in the $LiFePO_4$ cathode during the discharging process is formulated in Eq (3).

$$FePO_4 + xLi^+ + xe- \rightarrow xLiFePO_4 + (1\text{-}x)\ FePO_4 \quad (3)$$

The Li^+ extraction reaction that occurs in the LiFePO4 cathode during the charging process is formulated in Eq (4).

$$LiFePO_4\ (1\text{-}x)\ LiFePO_4 + xFePO_4 + xLi^+ + xe- \quad (4)$$

where $x < 0.6$

Based on the provided reactions, Lithium ions (Li+) are added to a $LiFePO_4$ cathode's cathode material during the cathode's discharge process, but they are removed from the cathode material during the cathode's charging process.

3.1.3. The Separator and The Electrolyte

The separator in lithium-ion batteries is not included directly in the redox reaction processes to generate electricity or store electricity, but the existence of the separator ensures safety in terms of battery use by preventing direct physical contact between the cathode and anode [31], [32]. Early in the lithium-ion battery manufacturing process, a separator is added to the battery to mechanically separate the anode from the cathode [33]. This is related to efforts to prevent short-circuiting of the internal battery [25], [34]. Polyethylene and polypropylene are frequently employed as separator materials due to their excellent mechanical strength, chemical resistance, and great thermal stability [35]. Besides, the electrolyte primarily functions as an ion route between the cathode and the anode [25]. Liquid electrolyte based on a lithium salt mixed in a combination of organic solvents is the frequently utilized electrolyte in modern lithium-ion batteries. Lithium hexafluorophosphate (LiPF6) is normally the most popular salt, and ethylene carbonate (EC), dimethyl carbonate (DMC), and diethyl carbonate (DEC) are common substances utilized in the solvent system [36]–[38].

3.2. The $LiFePO_4$ Cathode Materials and Its Preparation

$LiFePO_4$ crystals for lithium-ion battery cathode materials are included in the olivine family having a *Pnma* space group orthorhombic lattice structure. The crystal structure of delithiated $FePO_4$ and lithiated $LiFePO_4$ has the same structure except for the volume, which is 272.433 Å^3 for $FePO_4$ and 292.333 Å^3 for $LiFePO_4$ [39], [40], as presented in **Figure 2**.

One of the apparently most popular methods for researchers to make $LiFePO_4$ is by solvothermal and hydrothermal methods [41]. These methods are simple, some researchers proceed with the calcination process by making a precursor solution in a solvent that is usually a mixture of water and ethylene glycol (including ethylene glycol, diethylene glycol, and polyethylene glycol) in a certain ratio, then heating it for several hours.

3.3. The Research Trend on $LiFePO_4$ (LFP) Cathode of Lithium-Ion Batteries

$LiFePO_4$ (LFP) cathode for lithium-ion batteries has been the focus of continuous research due to its outstanding level of safety, inexpensive price, and good cycling stability. A number of studies have been conducted to improve the electrochemical performance of LFP. Surface coating, ion doping, and material nano crystallization are some of the research areas [42]. In this study, the research trends on $LiFePO_4$ (LFP) are categorized according to the articles published annually.

Figure 3 showed that since 2002 to 2022 there has been a continuous increase in the number of articles on current developments in carbon-coated $LiFePO_4$ surface modifications. Early in the 2000s, there were only a few publications; from 2006 onward, the count increased rapidly, reaching a high point in 2011. Furthermore, in 2019 and 2020, there was a noticeable increase in publications, and the trend continued in 2021 and 2022. This rising trend demonstrates the expanding significance and interest of $LiFePO_4$ surface modification highlighting their potential to advance energy storage.

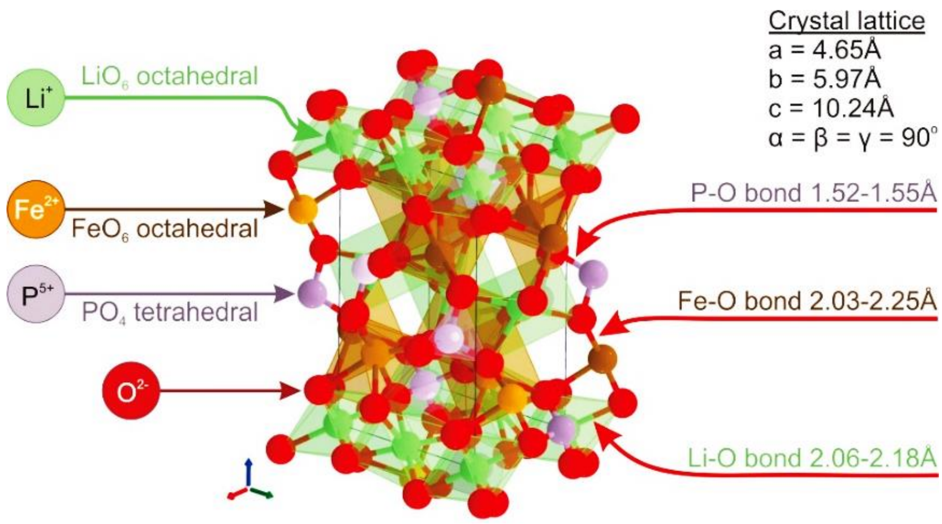

Figure 2. $LiFePO_4$ crystal in the orthorhombic *Pnma* space group

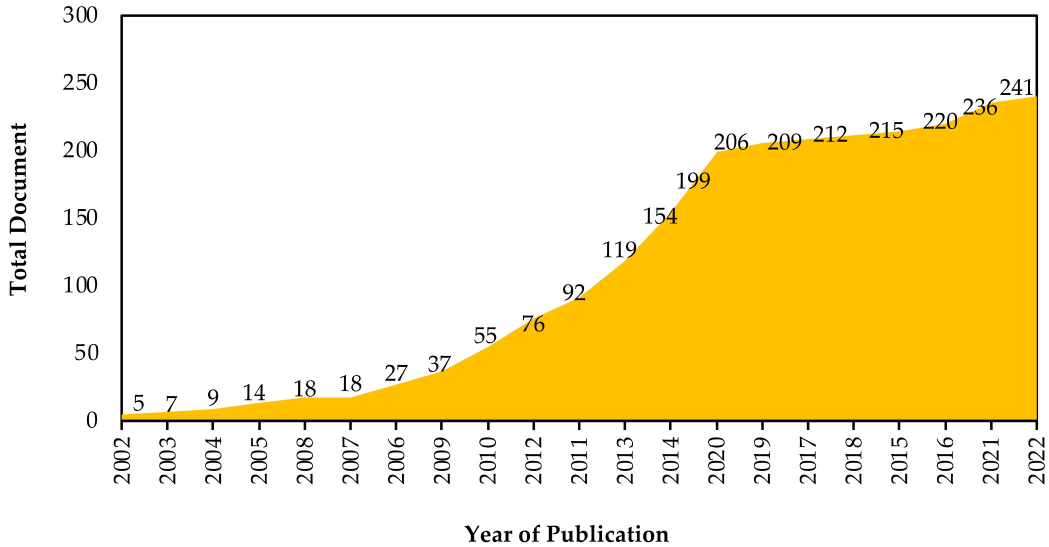

Figure 3. The Cumulative of annual published article related LiFePO4 (LFP) cathode of lithium-ion batteries

The considerable benefits provided by this battery technology have a significant impact on the growing trend in research on lithium iron phosphate (LFP) batteries from 2002 and 2022. Due to its safety, long cycle life, and affordability, LFP batteries are attracting more and more research attention [43]–[45]. In addition, due to their thermal and chemical stability, LFP batteries have an improved safety profile that tackles important issues in a variety of applications, especially those involving electric vehicles and energy storage [14], [46], [47]. In especially in sectors requiring numerous charge and discharge cycles, the extended cycle life of LFP batteries—which is greater than that of many other lithium-ion counterparts—is an important driver for research activities [43]. Additionally, LFP batteries are a desirable subject for research into economically efficient energy storage because of their long lifespan and low maintenance requirements [44].

Moreover, the increasing interest in research related to LFP batteries is further fueled by the recognition that significant opportunities for future research and advancements in battery technology still exist. Improving the volumetric energy density and specific energy/power, adding inherently safer chemistry, creating faster charging, and utilizing less expensive batteries with competitive/near-competitive performances are some of the difficulties that need to be addressed [48]. Therefore, because of their safety, high cycle life, and affordability, and the room for future research and advancements in LFP batteries are gaining more and more research interest.

3.4. *Bibliometric Analysis Based on the Co-authorship, Citation and Keywords Occurrence*

A bibliometric analysis is carried out in this study based on three important criteria. Co-authorship, citations, and keyword occurrence. The objective of this investigation is to recognize and comprehend the current state of knowledge regarding lithium-ion batteries that have had their surface modified with a layer of $LiFePO_4$ carbon. Therefore, by incorporating these three parameters, thorough grasp of the research trends, it can be acquired the significant contributors, and increasingly focused areas on study of lithium-ion batteries with $LiFePO_4$ carbon coating surface modification.

3.4.1. *Document Citation*

Figure 4 represents the most highly cited documents based on author citations. The top cited author is Zheng G. [49] with 1170 citations and has the highest number of citations among the listed authors, which indicates that their work has been more frequently referenced or cited by other authors.

3.4.2. *Co-Authorship Country*

Based on the data in **Figure 5**, it is shown that China has highest count of co-authorship country which demonstrates that China has made significant strides in this research topic. Besides,

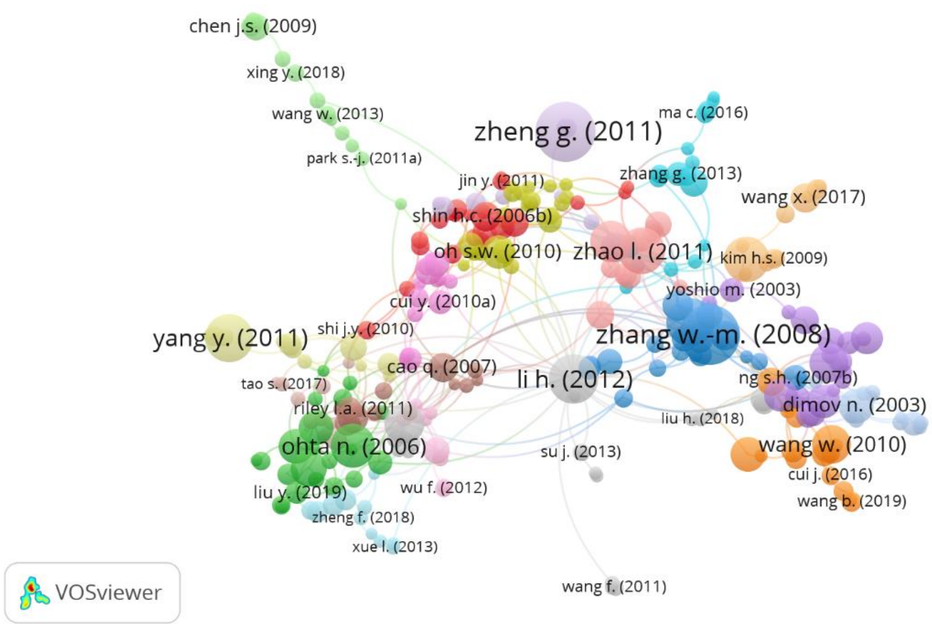

Figure 4. The most prominent cited documents based on the author citation

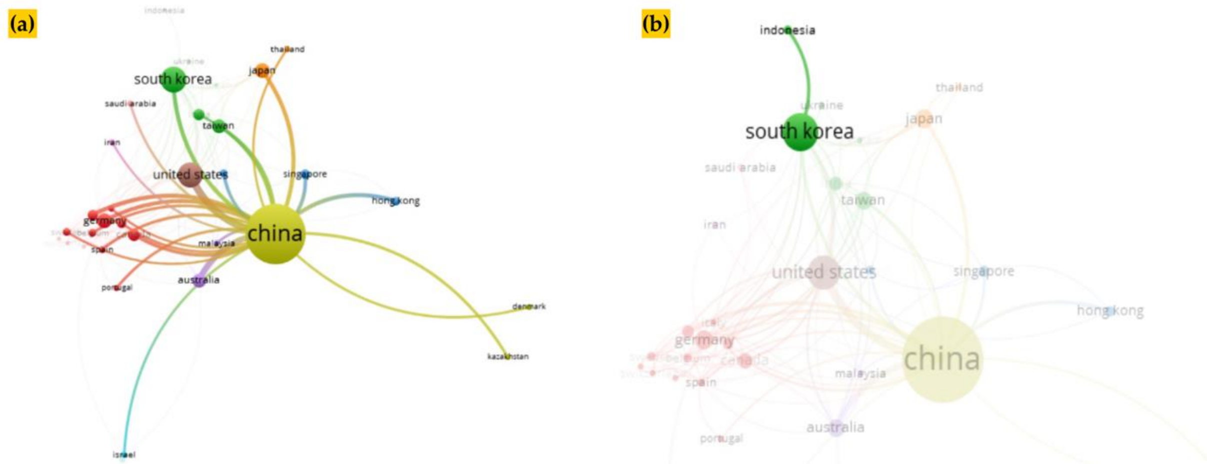

Figure 5. The most prominent Co-Authorship Countries, China (a), Indonesia (b).

the inclusion of Indonesia on the list suggests that country is actively engaged in this research topic, although there aren't as many publications as in China and South Korea. This finding suggest that Indonesian researcher should have more co-authorship or research collaboration in this regard of research topic.

3.4.3. *Keywords Occurrence*

Based on the keywords co-occurrence in **Figure 6**, it is evident that research on lithium-ion batteries has recently received considerable interest (a). In particular, when focusing on $LiFePO_4$, there is a notable relationship with carbon layers (b). Moreover, There has been a lot of research conducted regarding carbon coating in the context of lithium-ion batteries, in which these coatings serve as surface modifications to improve its performance.

3.5. *Surface Modification through Carbon Materials*

Currently, research is focused on using carbon as the primary component of $LiFePO_4$ coatings to carry out surface modifications to improve the electrochemical efficiency of the cathode. To enhance the electrochemical performance of $LiFePO_4$ cathode materials, numerous investigations have been conducted. The most pertinent studies on $LiFePO_4$'s carbon coating were listed in **Table 1**.

According to **Table 1** concerning the pertinent studies, the research on surface modification using carbon materials in lithium-ion batteries with $LiFePO_4$ cathodes has made substantial strides in improving battery performance. Various carbon materials, including Zeolitic Imidazolate Frameworks-8 (CZIF-8), core-shell composite nanospheres (C-PVDF), asphalt/soft carbon-coating (SCC), Super P (SP), polycyclic aromatic hydrocarbon coronene (C24H12), sodium maleate coating, fluidized bed chemical vapor deposition (FB-CVD) process, Paleozoic iron carbonate ($FeCO_3$), carbon nanotubes (CNTs), and conductive polymer polyaniline (PANI), have been explored. These modifications have resulted in improved of volume change flexibility, a better lithium-ion diffusion coefficient, higher conductivity, a high specific capacity, improved rate capability, and long-term cycling behavior. Besides, Phytic Acid (PhyA) synthesis, carbon-coated $LiFePO_4$ Nano-hollow Spheres (LFP@C HSs), PEDOT: PSS coating, Polydopamine-derived Nitrogen-doped Carbon (N-doped Carbon), Reduced Graphene Oxide (RGO), and Uniform and Ultrathin carbon coating have all shown notable improvements in capacity and performance ratings. These results encourage current attempts in research and development that focus on improving the effectiveness, stability, and general performance of battery systems based on $LiFePO_4$.

3.6. *LFP Surface Modification by MOFs Originated Carbon*

Metal-organic frameworks (MOFs) are crystalline substances with a significant amount of porosity of coordination compounds. It usually consists of a metal ion as the central atom that has the function of a node and an organic ligand with the chelating function as a linker, known as a secondary building unit (SBU) [50]. MOF applications represent clean energy potential, most significantly as a storage medium for gases such as hydrogen and methane, and as a high-capacity adsorbent to fulfil various separation needs [51].

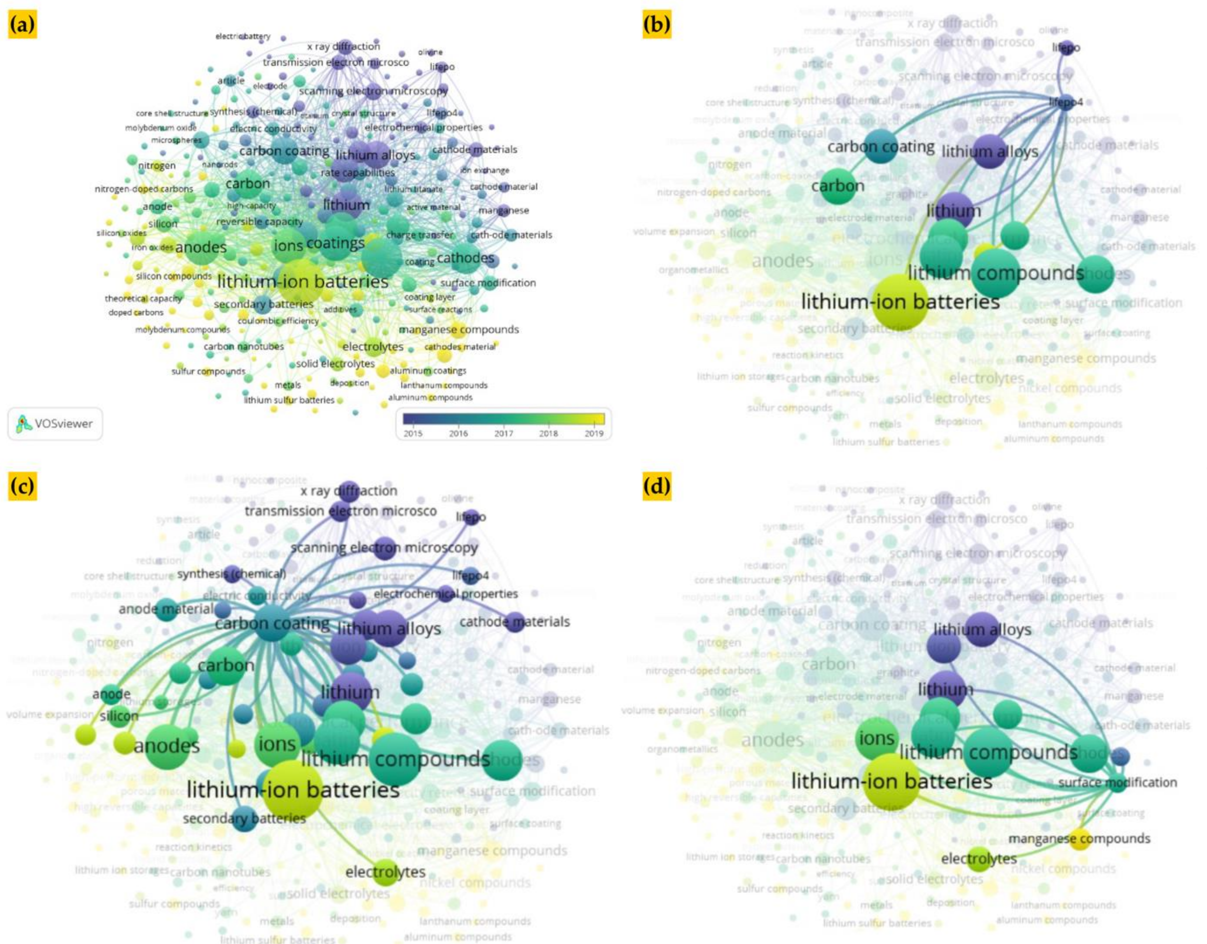

Figure 6. The Co-occurrence of: (a) all keywords; (b) $LiFePO_4$; (c) Carbon coating; (d) Surface modification

Table 1. The most relevant studies on surface modification through carbon materials

No	Author	Ref	Surface modification	Result
1	Xu X.L., et al.	[16]	Zeolitic Imidazolate Frameworks-8 (CZIF-8)	LFP/CZIF-8 significantly enhances the conductivity, the diffusion coefficient of lithium-ion batteries, and the degree of freedom for volume change.
2	Ponnamma., et al.	[52]	Core–shell composite nanospheres using polyvinylidene fluoride (LFP/C-PVDF)	PVDF is a simple and possible carbon precursor for $LiFePO_4$
3	Jiang W., et al.	[53]	Asphalt/Soft carbon- coating (SCC) and glucose/hard carbon-coating (HCC)	Asphalt can be used as an affordable and effective carbon source material for $LiFePO_4$ cathodes for LIBs.
4	Huang C.-Y., et al.	[54]	Super P (SP)	LIB with SP-coated has improved high-rate charge/discharge qualities.
5	Ye S., et al.	[55]	Polycyclic aromatic hydrocarbon coronene ($C_{24}H_{12}$)	CLFP cathode had a high specific capacity.
6	Heng S., et al.	[56]	Adding sodium maleate onto the surface of lifepo4 cathode	The $LiFePO_4$ cathode's rate capability and long-term cycling behavior are considerably enhanced by the sodium maleate coating.
7	Sarbarze S.A., et al.	[57]	Fluidized bed chemical vapor deposition process (FB-CVD) to coat carbon on LFP nanoparticles.	Overcome obstacles for conventional carbon coating methods
8	Dose W.M., et al.	[58]	Paleozoic iron carbonate ($FeCO_3$)	Increased optimization enhances the material.
9	Li W., et al.	[59]	Uniform and ultrathin carbon coating	Lithium migration into the active material and charge transfer kinetics were enhanced by a uniform and ultrathin carbon covering
10	Lian J., et al.	[60]	Conductive polymer polyaniline (PANI), active carbon and $LiFePO_4$ (C-LFP/PANI)	Demonstrate a notable improvement in capacity and performance ratings.
11	Lu J., et al.	[61]	Carbon-coated $LiFePO_4$ nano-hollow-spheres (LFP@C HSs)	The hollow-spheres structure and carbon layer coating are both crucial for enhancing $LiFePO_4$'s electrochemical performance.
12	Huynh L.T.N., et al.	[62]	Carbon nanotubes (CNTs)	Improve the $LiFePO_4$/C nanocomposite's electrochemical performance.
13	Raj H., Sil A.	[63]	PEDOT: PSS coating on pristine and carbon coated $LiFePO_4$	It is effective to coat the insulating $LiFePO_4$ cathode material with PEDOT: PSS, a conducting polymer.
14	Oh J., et al.	[64]	Nitrogen-doped carbon (N-doped carbon) and reduced graphene oxide (RGO) produced from polydopamine.	Enhancing the high-performance lithium-ion batteries.
15	Li Y., et al.	[65]	Phytic acid (PhyA) to synthesize $LiFePO_4$	PhyA is successfully used for producing $LiFePO_4$ materials in a variety of forms.

MOF can be synthesized by various methods and approaches to obtain pathways that can produce the expected topology, morphology, and activity, as well as surface area, size, and pore volume. The currently used in MOF synthesis include sonochemical, electrochemical, and mechanochemical methods, as well as hydrothermal and solvothermal approaches [66]. The sonochemical method or the ultrasonic irradiation-assisted method is applied with the aim of reducing the reaction time, but to produce a smaller particle size, organic-contaminant removal efficiency, and high crystallinity [67]. Mechanochemical synthesis is the chosen strategy to obtain a high surface area and quantitative yields, as well as adjustable pore size, and has just become a scalable green method for making MOFs [68], [69].

3.7. *MOFs and Their Applications to Electrochemical Energy Storage and Conversion*

Due to their superior chemical and physical properties, as well as the various methods that can be adapted to synthesize them, for electrochemical energy storage and conversion technologies, MOFs are desirable functional materials and precursors [58]. In particular for hierarchical nanostructures, MOFs are great precursors for designing and creating nanostructured porous carbons and metal oxides [70]. However, the use of MOFs as a source material for N-doped carbon for electrode coating is still very limited, especially for olivine $LiFePO_4$.

Based on the previous research, it is shown that the use of MOF as an N-doped carbon material to coat $LiFePO_4$ gives very significant results, especially in improving LIB conductivity and Li^+ ion distribution, which gives a very high specific charge/discharge capacity, close to the theoretical value. The organic ligands with one or more electron-withdrawing groups are the best ones to use in redox processes. Therefore, using quinone derivatives functionalized with coordination groups as organic linkers in combination with the proper metal ions is workable method of creating MOFs for energy storage applications [71].

Furthermore, the selection of suitable synthetic parameters can improve the electrochemical stability of MOFs [72]. Studies show that the utilization of MOFs for energy conversion and storage indicates that rich porosity states, abundant redox sites, and high surface area can improve the effectiveness of electrode materials MOF-based materials can be nanostructured to have components with active sites that are both highly active and stable [73]. Another advantage of using MOFs is that MOFs can be derived into nanostructured carbon, it is carbon–metal/metal oxide hybrids that can be simply produced.

Additionally, by picking the appropriate MOF precursors, it is simple to change the composition and shape of carbon products; alternatively, the carbon materials produced can even achieve in situ heteroatom doping (such as N, P, S, and B) with a regular and uniform atomic distribution matrix. These exceptional structural benefits allow MOF-derived carbon, which has so far been used in the field of energy storage and conversion systems, to realize its immense promise as a high-performance energy material [74].

3.8. *Future Works Recommendation*

The extent of the bibliometric analysis is one of this study's limitations. The analysis is based on current papers as of a certain date; however, new scientific advancements may have since appeared, thereby affecting the analysis's conclusions. Therefore, to acquire a more thorough assessment of the research trends in this sector, future studies might think about enlarging the scope of study and updating the data.

4. Conclusion

This review encourages current attempts in research and development that focus on improving the effectiveness, stability, and general performance of battery systems based on $LiFePO_4$. The effectiveness of electrochemical conversion and storage of energy methods can be improved through the surface modification of $LiFePO_4$ electrodes utilizing MOFs-derived carbon materials. Carbon materials generated from MOFs have great promise for high-performance energy storage and conversion devices due to their outstanding structural advantages. Therefore, it can enhance the cathode conductivity of lithium-ion batteries.

The bibliometric analysis offers a thorough comprehension of the present state of study on lithium-ion batteries with $LiFePO_4$. It is clear that research on lithium-ion batteries is receiving attention, a sign of its significance and rising popularity recently. Regarding co-authorship, China came out on top as the nation with the highest number of co-authors, demonstrating their considerable dedication and outcomes in this study field. As a result of this study, Indonesian researchers may be able to contribute more to the field by increasing the number of co-authors on this particular topic.

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Author's Declaration

Authors' contributions and responsibilities

Apang Djafar Shieddieque, Iman Rahayu, Sahrul Hidayat, and Joddy Arya Laksmono, wrote and revised the manuscript. All authors agreed to the final version of this manuscript.

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Availability of data and materials

All data are available from the authors.

Competing interests

The authors declare that the publishing of this paper does not involve any conflicts of interest. This work has never been published or offered for publication elsewhere, and it is completely original.

Additional information

No additional information from the authors.

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