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Covalent organic frameworks for next-generation of lithium-ion batteries

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Abstract

The demand to develop lithium-ion batteries (LIBs) is growing as their consumption in various applications rises. In this regard, it is very important to pay attention to materials with suitable properties for use in LIBs. The energy storage capacity, cycle life, weight, and recycling of aging batteries are expected to improve with these materials. Recently, covalent organic frameworks (COFs) have shown great promise for use in cathode, anode, electrolyte, and separators of LIBs due to their numerous properties, including porosity and excellent physical and chemical stability. This review discusses the application of COFs in the next-generation of LIBs. First of all, the Main components, working principles, and characteristics of LIBs are described. After that, the advantages, applications, and synthesis methods of COFs are discussed. Finally, the application of COFs in various parts of LIBs by key features and the associated challenges is highlighted. In this review, we focus on improving batteries by introducing and developing COFs as a new-generation of materials for energy storage applications to overcome the current limitations and provide promising alternatives for current LIBs.

Keywords: *Lithium-ion batteries, Covalent organic frameworks, Energy storage applications, Electrolytes, Electrode materials*

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1. Introduction

Innovative energy storage technologies are required due to the continual rise in global energy consumption and a move toward renewable energy sources. Batteries can decrease the need for power supply during periods of peak usage and increase the use of renewable energy. Batteries lead the transportation network towards using electric vehicles and provide options for storing energy from renewable sources, such as solar and wind [1,2]. Since lithium-ion batteries (LIBs) were commercially available in 1991, their use has expanded from specialized industries to include portable devices, electric vehicles, and grid energy storage systems. Technological advancements

have increased energy density, going from 80 Wh/kg for first-generation commercial cells to around 300 Wh/kg for LIBs. Despite the significant technological advancement, LIBs still have many disadvantages, such as expensive lithium sourcing, short cycle lifetimes, and safety concerns [3].

Some substances, including carbon, silicon, and organic polymers, have so far demonstrated exceptional electrochemical characteristics in LIBs [4]. Because of their advantages of lower toxicity, recyclability, structural variety, and enormous reserves, organic electrodes are regarded as attractive energy storage materials for the next generation [5]. A new type of crystalline porous materials called covalent organic frameworks (COFs) are entirely made of organic building blocks and displays outstanding

porosity, ordered channels, and great resistance in most solvents [6]. In recent years, numerous functional COFs have been developed with tunable physicochemical features for a variety of applications, including catalysis for gas adsorption and separation [7,8], optoelectronics [9,10], water and wastewater treatment [11,12], sensing [13–15], and energy storage [16,17]. COFs are covalently connected lightweight elements with several outstanding inherent benefits, such as structured crystalline structures, many configurable pores, high surface areas, low densities, and strong chemical and thermal stabilities [3]. The well-aligned π -columns of 2D COFs allow for significant electronic contacts between nearby atomic sheets, realizing effective carrier transfer. Due to the 2D COFs exceptional designability, interactions between molecules, ions, holes, electrons, excitons, and spins are also probable to occur. Therefore COFs would have enormous potential for energy storage and conversion applications [6].

COFs have been widely researched in batteries, including LIBs, lithium-sulfur batteries, sodium-ion batteries, potassium-ion batteries, zinc-ion batteries, zinc-air batteries, lithium CO₂ batteries, and lithium-silicon batteries. To change the energy density of electrode materials, the redox-active sites can be built into COF precursors with precisely controlled densities and locations [1]. To overcome the slow ion transport issues in batteries, organic functions that support metal ion mobility can also be properly anchored in the structure of COFs [18]. In LIBs, COFs could include many active Li⁺ embedding sites. However, COFs have a high relative molecular quality, which makes them difficult to dissolve in electrolytes and can improve the LIBs cycle life. Furthermore, Li⁺ movement and charge exchange in LIBs are promoted by porous structures, open channels, and π -conjugated systems of COFs [4].

Recently, many excellent reviews have discussed the application of COFs in different batteries, but still few reviews have accurately and completely introduced various aspects of LIBs, COFs, and the application of COFs in different parts of LIBs [1,3,4,6,19–21]. This review first discusses the main components, working principles, and characteristics of LIBs. The advantages, applications, and synthesis methods of COFs are also presented. Then, the COF-based

cathode, anode, electrolyte, and separator in LIBs are described. Finally, the challenges of using COFs in LIBs are discussed.

2. LIB fundamentals

Researchers have been motivated to replace fossil fuels with sustainable energy converters because of the energy crisis and global warming, so sustainable energy converters become the key parts of any new electronic device or electric vehicle. Therefore, these devices and vehicles require proportionally suitable energy storage systems [22]. The main types of energy storage devices are fuel cells, LIBs, capacitors, and supercapacitors [23].

In recent years, LIBs being the best choice and have gained great attention. LIBs are used in some applications such as computers and cell phones [24], and also for electric or hybrid vehicles [25]. They show excellent performance and energy density for all those applications [26].

LIBs have a high energy density, long service life, and no memory effect [27,28]. So, they are one of the first choices for electric vehicles (EVs) and renewable energy systems (RESs) [29]. Although they have improved recently, but there is still a capability for further investigation. LIBs will play an important role in the future because of the concept of low-carbon and excellent properties. The application of LIBs has also expanded to various areas like robots, automated guided vehicles (AGVs), and consumer electronics.

Battery aging is divided into two main forms: calendar aging and cycling aging [30–33]. Calendar aging is related to the high temperature and SOC [34–36] and cycling aging is related to the charging and discharging process of LIBs. This battery contains the cathode, anode, and separator for transferring the Li ion to charge and discharge battery [37]. There are some researches that focused on improving LIB performance by a different method such as developing new catalysts [38–40]. The highest energy density their major drawback is safety and operational issues [41].

2.1 Main components of LIBs

By insertion/extraction of lithium ions, these batteries can store/provide energy in/from the structure of the

electrode in successive charge/discharge cycles. Energy and power densities determine the battery's performance. In order to improve the energy/power density and cyclic life of a LIB, its electrode, and electrolyte must be properly investigated. Cathode materials through conversion reactions can store energy and also anode materials through intercalation, conversion reactions, or alloying/dealloying store energy. The electrode material is one of the important parameters which directly affect the battery performance. Each group of electrode materials has its own advantages.

2.1.1 Cathode

One of the most important components in LIBs is the cathode. during the charging large compositional changes because of the oxidation reaction so the cathode material must have a stable crystalline over wide ranges of composition [42]. During discharging cycle, a reduction of the transition metal ions in the cathode is carried out by the electrons from the anode. The electrode microstructure and morphology effects cathode performance [43].

Energy can store in cathode material through two mechanisms: (1) In the intercalation mechanism cathode material act as a host for Li ions and the ions can insert in or extract from the material reversibly such as: FeF_2 , CoFe , and NiF_2 . They have a lot of challenges because of the high volume, expansion, poor electron conductivity, and hysteresis issues.

Transition metal oxides due to their higher operating voltage and the resulting higher energy storage capability are the best examples of intercalation material [44]. LiCoO_2 is the most commonly used cathode material in LIBs. LCO has a high cost of cobalt and safety issues [45]. (2) Conversion electrodes undergo a solid-state redox reaction during lithiation/delithiation, in which there is a change in the crystalline structure, accompanied by the breaking and recombining of chemical bonds. The main cathode materials used in LIBs are listed in Table 1 with their advantages and disadvantages.

The cathode materials are classified according to their energy storage mechanism and they can store energy by intercalation or conversion reaction mechanism. Each group of electrode materials has some disadvantages that researchers try to solve them.

2.1.2 Anode

Another component of LIBs is the anode which they made from three distinguished groups of materials. In the intercalation-based group, Lithium ions are electrochemically intercalated into the space between the layers of materials, such as graphite which is widely used in LIBs for mobile devices. One of the downsides of this group is irreversibility during the first charge (lithiation) process cause a cathodic decomposition of a number of constituents of the electrolyte [46]. Therefore, the solid-electrolyte interface (SEI) is a stable passive layer that is formed

Table 1. Main cathode materials used in LIBs and their advantages and disadvantages.

Material	Advantage	Disadvantage
LiCoO_2 (LCO)	High structural stability and ease of mass production	High cost of cobalt and safety issues [45]
LiNiO_2 (LNO)	Does not have the drawbacks of LCO. Delivers 20 to 30% more reversible capacity than LCO [128].	-
$\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ [24]	High specific capacity / enhanced Rate capability / cycle-ability [46]	-
Over-lithiated Oxides (OLOs)	High capacity LIBs [46]	-
LiMn_2O_4 (LMO)	Three dimensional framework which promotes the movement of lithium ions [46,129]	Reversible capacity is less than that of LCO or LNO.
LiFePO_4 (LFP)	Thermal stability and high-power capabilities [130,131]	-

to Charge/discharge reversibility which is essential for long-term cycle-ability. Some solvents which form a satisfactory SEI layer are ethylene carbonate (EC), dimethyl carbonate (DMC), and methyl ethyl carbonate (MEC).

Moreover, a major drawback of graphite anodes is their low capacity [47] so developing carbonaceous material to obtain better performance. Increasing the surface area is one way to increase their capacity. Carbon nanofibers (CNF), carbon nanotubes (CNT), and graphene are studied as an alternative to graphite because of their larger surface area as well as higher electron conductivity which makes them suitable for high rate charging/discharging [48].

Conversion-reaction-based is a second group of anode materials that are produced by combining candidates of M and X even without considering multiple oxidation states of M. Their range is from the low range of 350 mAhg^{-1} for Cu_2S to the high range of 1800 mAhg^{-1} for MnP_4 [49]. Some drawback of this group is related to their irreversibility and pulverization.

They are not as good as graphite in their cyclability and during discharge to charge they drastically decrease their cyclability. This means that the conversion reactions in these materials would have intrinsically limited reversibility. Moreover, they become pulverization or electric isolation due to the lithiation and the following delithiation [50]. The reaction potentials of that materials are relatively higher than that of graphite, so it leads to lower cell potential which results in a lower energy density than expected only from capacities [49].

Alloying-reaction based materials are another group of anode materials in this group some metals such as silicon (Si), germanium(Ge), tin (Sn), and their alloys have been combined with lithium [51–53]. If the lithium ion is not well dissolved in the anode material, a phase change occurs in the anode so the alloy metal has different from its pure structure. The alloying-dealloying mechanism in Si, Sn, Al, and Sb is accompanied by phase change [54]. Extremely large volume change during charge and discharge is their problem [55]. Most of the research worked on improving the performance of silicon as an anode material because of its highest capacity and its most serious detrimental volumetric change [56].

Each group of anode materials has different energy storage mechanisms and store energy by intercalation, conversion reaction, and alloying/dealloying mechanisms. Each group of electrode materials has its own drawbacks, such as a very high-volume change in the conversion or alloying/dealloying based anode materials. Even considering the much research spent on enhancing the performance of LIBs in recent decades, these batteries still have numerous challenges which need to be overcome.

2.1.3 Electrolyte and separator

A LIB uses liquid electrolytes. It allows lithium ions (Li^+) to move between anode and cathode, stabilizes cathode and anode surfaces, extends battery lifespan, and improves cell performance. It plays a pivotal role as one of the major four components of a battery. The electrolyte is another crucial component of an electrochemical battery. The electrolyte conducts ions between the anode and cathode, but it is insulative to electrons [57–62]. LIB research has heavily focused on liquid electrolytes. Although high ionic conductivity and easy-accessibility could be achieved by liquid electrolytes, they often suffer from poor electrochemical and thermal stabilities, as well as low ion selectivity. They have many intrinsic drawbacks that are summarized in the following:

- Liquid electrolytes have caused many safety concerns due to their low voltage windows and flammability, especially under high power operations or in large-size batteries [59,61,63].
- Dissolution of active materials and side reactions often occur in liquid electrolytes, which deactivates the electrode materials and decreases the cycle stability and rate performance [59–62].
- Liquid electrolytes usually have low lithium transport numbers around 0.5 [59–62].

To avoid these issues, we can use solid-state electrolytes (SSEs). Chalcogenides, and solid-state polymers are commonly used as SSEs [59].

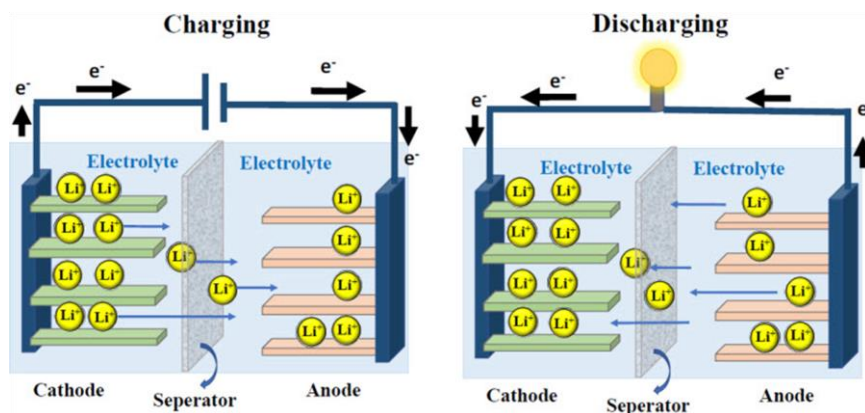


Figure 1. Schematic presentation of a LIB during charging and discharging [132].

2.2 Working principles and characterization

A battery is made up of an anode, cathode, separator, electrolyte, and two current collectors (positive and negative). The anode and cathode store the lithium. The electrolyte carries positively charged lithium ions from the anode to the cathode and vice versa through the separator. The movement of the lithium ions creates free electrons in the anode which creates a charge at the positive current collector. The electrical current then flows from the positive current collector through a device being powered (cell phone, computer, etc.) to the negative current collector. The separator blocks the flow of electrons inside the battery. Discharging, the lithium ions travel from the anode to the cathode through the electrolyte, thus generating an electric current, and, while charging the device, lithium ions are released by the cathode and then go back to the anode [64]. Figure 1 shows the process of charging and discharging in LIBs.

The two most common concepts associated with batteries are energy density and power density. Energy density is measured in watt-hours per kilogram (Wh/kg) and is the amount of energy the battery can store in mass unit. Power density is measured in watts per kilogram (W/kg) and is the amount of power that can be generated by the battery per unit mass. To draw a clearer picture, think of draining a pool. Energy density is similar to the size of the pool, while power density is comparable to draining the pool as quickly as possible [65].

3. COF-based materials

3.1 Advantages of COF

COFs have a variety of advantages, including chemo-stability and thermo-stability, porosity, and highly organized structures, that could be very helpful in battery applications. A variety of COF topologies, couplings, and functionalities could be achieved by carefully choosing the monomers. Using a bottom-up synthetic technique, redox active groups or lithium ion coordinating groups, which are essential to LIBs functionality, can be added right into COF structures. For better performance in LIB applications, post-synthesis modification of COFs has also frequently been employed as a substitute strategy. This involves adding new functional groups or changing the structure of the COFs backbone [3].

One of their most outstanding features is the flexibility to modify the structures and functionalities of COF materials for particular applications. By using a range of easily accessible monomers, COFs can be built through rational design. Studying the structure-property relationship of COFs is made possible by fine tuning their topology, linkage, and functional groups. Typical examples of connections that serve as vertices to form hexagonal pores in COFs include the boroxine and triazine rings. Multitopic monomers are necessary as vertices to create a 2D structure when links are linear. In addition to topologies, scientists have shown great control over the pore size and porosity of COFs by adjusting the length and symmetry of the constituent parts [3]. Through monomer design and

linkage formation, desirable functionalities for LIB applications can easily be included in COFs. Post-synthesis alteration or de novo design can improve COF performance [66]. Functional groups have also been routinely introduced into pores using post-synthetic alteration techniques. Another popular method for creating COFs for LIB applications is through the formation of functional connections. Linkages can form that are non-conjugated, conjugated, ionic, or neutral. Although highly ordered materials have been created using non-conjugated connections, their electron delocalization is poor because of bond polarization [67]. Conjugated connections have good electron delocalization characteristics and are often stable [68]. Next-generation LIB materials can be designed using COFs, which offer an ideal platform due to their customizable porosity and great structure tunability.

For LIBs to have practical uses, material stability is essential. Because of their sturdy frameworks, COFs can withstand various working situations without decomposing. Certain linkages, including boronate, boroxine, and amine, are vulnerable to hydrolysis in acidic environments. However, other ultra-stable COFs have been produced immediately or after the initial synthetic process [69]. TGA has demonstrated that the COF material is thermostable up to 400 °C [3]. It is expected that the strong thermal and chemical stability of COFs will improve LIBs functionality. Another advantage of COFs is their proper porosity. Porosity is particularly desirable for the movement of lithium ions or the accommodation of sulfur in the cathode in sulfur-lithium batteries [3]. The pore size and distribution reveal more details on the framework layering and regularity of the channels generated in COFs. The adsorption isotherm data can be used to determine them. COFs with a single pore model or a variety of distinct pores can be produced, depending on the geometry and symmetry of the monomer construction blocks [70].

3.2 Applications of COFs

3.2.1 CO₂ Capturing

The increased atmospheric CO₂ due to anthropogenic activity is a significant global problem. Future industrial and urban development, which constantly

adds CO₂ to the atmosphere, could exacerbate atmospheric CO₂ levels. Materials that are effective at capturing CO₂ from the atmosphere are needed due to the significance of the topic [71]. High durability and affordability should be characteristics of the material. High porosity and functionalized COFs make great candidates for efficient CO₂ capture. When CO₂ is converted into carbonates, the cycloaddition of propargylic alcohols is catalyzed by the nano-Ag catalyst, which COF can support. Because nitrogen atoms have a higher interaction affinity for carbon dioxide, nitrogen-rich COFs are a good choice for CO₂ collection [72].

3.2.2 COFs in Sensors

As was previously mentioned, COFs are porous crystalline substances made of light elements bound together by covalent bonds [73]. Because of their configurable functionality, large surface areas, low densities, and organized pores, COFs can be utilized for various sensing applications, including humidity sensing, electrochemical, and fluorescence sensors [71]. These sensors are used to identify nitroaromatic explosives, volatile organic chemicals, and tiny organic molecules. It has been reported that a magnetic COF-based immunoaffinity probe may identify cancer biomarkers during clinical diagnosis [21]. The application of COFs for the sensitive and focused detection of chemicals is intriguing given all these characteristics of COFs. Some COF-based sensors are electrochemical, Immuno, and antibiotic sensors [71].

3.2.3 COFs for environmental remediation

3.2.3.1 Adsorption in aqueous solution

Heavy metal and radionuclide contamination pose a significant concern to the environment and human health because of their toxic consequences. Therefore, they must be eliminated using the appropriate treatments before being discharged into the aquatic environment [74]. Using COFs to remove metal pollutants by adsorption may be highly efficient. Numerous COFs have been found to effectively remove metal-containing ions from water while exhibiting promising selectivity between various water-based ions and favorable adsorption kinetics. Ion exchange and coordinative adsorption using ligand sites on the COF surface are the two main adsorption

processes [75]. Rare earth elements have also been captured using COFs, particularly 3D ones, following post-synthetic alteration to include carboxyl groups. This was accomplished through the succinic anhydride-ring-opening reaction between the OH groups on the pore surface of the 3D COFs [76]. The addition of succinic anhydride reduced the porosity of the COFs. However, the carboxyl functionalized 3D COFs displayed strong selectivity for lanthanide ions and could be used repeatedly without noticeably degrading their adsorption efficiency [77].

3.2.3.2 Adsorption of organic pollutant

The widespread use of pesticides, organic dyes, and PPCPs is a big concern; most of these substances are organic substances that can contaminate water [78,79]. Often, only a tiny bit of the pollutant is required to have a significant negative impact on both human health and the environment. Therefore, organic pollutants should be removed before being discharged into a water system [80,81]. Due to their broad, rigid, and stable porosity, COFs have demonstrated promise for the adsorption of large organic compounds. Different adsorption methods have been reported, depending on the pollutants that are being targeted and the type of COF functioning. The pore size effect, H-bonding, hydrophobic interaction, and π - π interaction are some of the main adsorption mechanisms [81,82], [83].

3.2.3.3 water treatment

The most popular membrane-based separation techniques that COF-based membrane materials can be utilized for include microfiltration, reverse osmosis, nanofiltration, and ultrafiltration. The potential of COFs for water desalination was demonstrated by computational research [84]. Numerous studies on COFs as effective water filtration membranes have shown that they can be used for water treatment [75]. Interfacial polymerization created a 3D COF membrane and incorporated it into a polyamide layer. The covalent joining of COFs with many secondary amino groups changed the membrane's hydrophilicity and stability. Additionally, after the alteration, better desalination performance was observed in terms of increased pure water flux and long-term running stability without compromising salt-rejection ability [85].

3.2.3.4 Gas separation

Gas separation has also been accomplished with success using COF-based membranes. Several COFs with varied pore apertures and various covalent connections have been studied as elements in mixed-matrix membranes. These membranes contained fillers made of COFs, which function similarly to those used in polymer matrices. For this work, there must be a high affinity between the dispersed filler phase and the continuous polymer matrix phase. The separation performance is greatly influenced by how much COF is loaded into the mixed-matrix membrane. Due to the contribution of evenly spaced-out channels, low COF impregnation exhibits improved membrane performance. However, excessive COF loading might clog the gas transport channels in the membrane and reduce performance [86].

3.2.4 COFs for biomedical applications

The biocompatibility and bio-functionality of a biomaterial are its two essential characteristics. Depending on the type of application, other properties like strong corrosion resistance, high wear resistance, the capacity to withstand or interact with host reactions, and extended fatigue life should also be considered. As a result, biomaterial design and selection are crucial for the intended application. COFs offer outstanding design and functionalization possibilities, just like polymeric biomaterials. The use of COFs as biomaterials is still relatively new; thus, it's critical to identify the particular fields in which they can be useful [87].

3.2.5 COFs-based catalysts

In ideal circumstances, the holes created in COFs offer a contained space, allowing the growth of nanoparticles to be regulated. The applications of COFs can be expanded by incorporating other functional materials into COFs. COF-supported nanoparticles demonstrate outstanding catalytic characteristics towards numerous reactions, such as oxygen evolution reaction, water splitting, and organic pollutants degradation [71]. Due to their broader size distributions, COF-supported nanoparticles have significantly higher catalytic activity than unsupported nanoparticles. These catalysts exhibit good dispersity and more significant catalytic activity and are stable in aqueous and organic environments. Materials made of

COFs have been utilized to assist organic processes such as nitro reduction, Suzuki coupling, and glycerol oxidation [88].

3.2.6 COFs in batteries

The performance of the current batteries is being improved, and COFs are also intended for battery applications. COFs are being investigated as advanced organic materials for use in rechargeable batteries. In order to produce flexible Zn-air batteries, LIBs, Li-CO₂ batteries, lithium-sulfur batteries, and potassium ion batteries, COFs are being researched. Materials based on COFs have shown to be promising materials for rechargeable batteries since they are simple to functionalize and have open channels [1]. COFs can be widely used as anodes, cathodes, electrolytes, and separators in LIBs.

3.3 Design and synthesis of COFs

Due to its unique geometries, pore shapes, compositions, and uniformities, COF synthesis is more difficult than standard polymers. As a result, it is not easy to synthesize a rationally designed COF with specified features, and successful COF production necessitates the integration of several research fields, including organic chemistry, topochemical polymerization, and reticular assembly [75]. Linkers (Building units) and linkages (Bonds created between those units during reticulation), which regulate pore size, are the two distinct modules that make up a COF framework. To achieve the design strategy, the building units must have specified geometry and hard structures. Consequently, the synthesis of the building components begins the process of the organic synthesis of COFs, after which they are connected to form an extended framework using reticulation. The chemical reactions in creating COFs need to be reversible to enable the growth of lattice structures. Reversible reactions provide COF systems the potential to self-heal and correct structural flaws, which is essential for developing highly structured frameworks. The creation of the building units (topology design concept) and the synthesis method are the first and second issues that must be considered in the design and synthesis of COFs, respectively [89].

3.3.1 Design strategies

For energy storage applications, 2D COFs are frequently used. Because 2D COFs perform superior in terms of charge transfer, conductance, and redox coupling capabilities, their eclipsed structural alignment is widely believed to be ideal for energy storage applications. The symmetrical monomers may effectively control microscopic features, such as pore sizes, shapes, environments, and porosity. Symmetrical building blocks are typically categorized as C₂, C₃, C₄, and C₆. Hexagonal, tetragonal, trigonal, kagome, and rhombus polygons can be created by combining these basic elements, according to the topological diagram illustrated in Figure 2. Although a growing variety of asymmetrical building blocks are gradually being developed, these design diagrams have been widely used to construct 2D COFs. The connection type, in addition to monomer structures and topologies, significantly impacts the physicochemical characteristics of COFs. The boronic ester, imine, imide, olefin, and dioxin bonds are the most widely employed linkages because of their availability, repeatability, and stability [6].

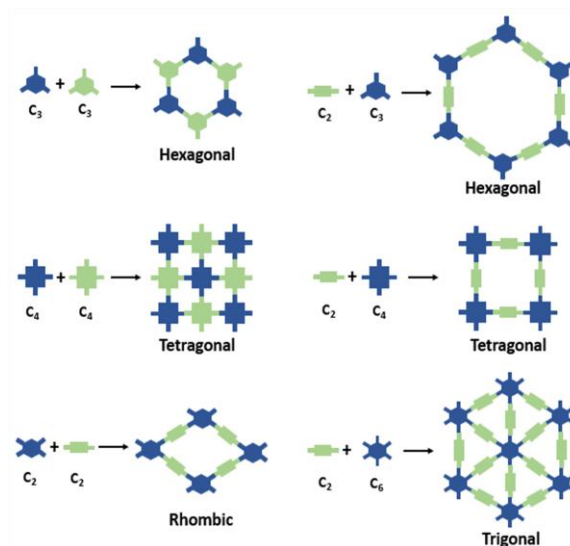


Figure 2. Topological diagram for 2D COFs [6].

3.3.2 Synthesis methods

3.3.2.1 Solvothermal synthesis

Solvothermal conditions in a sealed vessel have been used to synthesize the majority of known COFs. Nevertheless, throughout the synthetic process,

numerous parameters, such as the pressure in the sealed vessel, temperature, reaction time, and the volume ratio of solvent combinations, play important roles in the crystallinity of COFs. The COF films cannot be transferred to preferred substrates for characterization and specific application because they depend on specified supports when prepared. Additionally, because COF particles in a solution phase are the main product, the yield of film production is low. Furthermore, it is not easy to regulate the thickness of COF sheets under solvothermal conditions. The quality of COF crystallites and films is influenced by various factors from the standpoints of synthesis techniques and circumstances [90]. Solvothermal synthesis of COFs is comparable to the autoclave-based synthesis of Metal Organic Frameworks (MOFs). The pyrex tube is first filled with the reactive substance, and a degassing procedure is carried out. The tube is then sealed and heated to the predetermined temperature. The precipitate is then collected, cleaned using the proper solvents, and dried in a vacuum to produce solid powder COFs [89].

3.3.2.2 Synthesis of COFs in ionic liquids

Ionic liquids have recently been investigated for the mild-condition synthesis of imine-linked 2D and 3D COFs with increased crystallinity. Molten organic salts with low melting points are known as ionic liquids. Ionic liquids have physicochemical features that can be changed by the counter anions and are nonvolatile, nonflammable, and ecologically friendly. This might result from the idea that an ionic environment makes it easier for aromatic building components to align into an ordered structure. Ionic liquid synthesis is quick, easy, and economical compared to solvothermal synthesis [91].

3.3.2.3 Microwave synthesis

Chemical reactions can be accelerated using microwave heating. It has already been discovered that crystalline COF materials can be synthesized using microwaves [89]. In 2009, crystalline boron-based two- and three-dimensional COFs were first synthesized using microwave synthesis. The produced COFs had surface areas comparable to or even more significant than those obtained using the solvothermal approach and the reaction time was significantly

reduced. COFs could only be easily produced after 20 minutes of irradiation. Compared to the solvothermal synthesis, the reaction rate is almost 200 times higher [90].

3.3.2.4 Mechanochemical synthesis

In addition to being used for the synthesis of MOFs, mechanochemical synthesis has also been shown to be successful for the synthesis of COFs [90]. Only basic experimental setups like mortar and pestle are required to synthesize COF materials using this approach. In contrast to the solvothermal method, the mechanochemical process is restricted to the synthesis of COFs, which have low crystallinity and porosity. The synthesized COFs are resistant to boiling water, powerful acids, and bases [91]. According to these results, synthesizing COFs from highly polar monomers may be replaced by mechanical synthesis [90].

3.3.2.5 Template surface synthesis

The solvothermal process was used to synthesize the COF materials, which typically resulted in powder. This may have limited their application areas in some circumstances, such as when used in device and interface combinations. The thin film materials increased the crystallinity more than the powder samples. This technique can also be used to create other 2D COF films, each exhibiting distinctive qualities. The process resulted in building functioning COFs and enhanced their performance in actual applications. Moreover, some crystalline metal surfaces can also be used to synthesize COF sub-monolayers [89].

3.3.2.6 Sonochemical synthesis

This approach involves applying ultrasound to the reaction system to cause bubbles to form and burst in the solution, which can hasten the crystallization process due to acoustic cavitation. The synthesis of 2D and 3D COFs using this technique is possible in powder and film form. Due to the lack of an induction period, sonochemical synthesis can reduce reaction time. Furthermore, compared to the usual batches utilized in solvothermal synthesis, the reaction can be performed on an immense scale. In this regard, sonochemical synthesis provides an affordable and

energy efficient method of producing high quality COFs [91].

4. COFs for LIBs

Serious global warming and dangerous environmental consequences from using fossil fuels led developing of some alternative clean and renewable energy resources and utilizing modern technologies. In this journey, by arising electrical vehicles and taking advantage of wind and solar energies, LIBs have emerged as promising systems for energy storage and some other relative applications [92,93]. There have been numerous studies for the exploration of more applicable and beneficial battery systems of Li-ion generation in which all-solid-state Li batteries and Li metal batteries have been known as great candidates for post-LIBs [94].

Recent LIBs are using liquid electrolytes in which there are lots of organic solvents producing thermal instability practically. They have some other drawbacks including short circuits, explosion threats, and other issues as well [95]. To avoid the subsequent deleterious safety problems, many efforts have been doing to develop desirable replacement(s) for liquid electrolytes. Solid electrolytes such as ceramic electrolytes were introduced to probably be high potential candidates because they have more efficient properties theoretically; for example, they were considered safer with higher energy density and have a wider electrochemical window than liquid electrolytes. However, the low interfacial contact of solid-based electrolytes kept them behind and solid polymer electrolytes were proposed for their higher flexibility, consistent interfacial contact, and are suitable for undergoing industrial processes. Meanwhile, most solid polymeric electrolytes had a special condition for their high ionic conductivity as they need a glassy or melted environment. Furthermore, their ionic conductivity is dependent on the amorphicity degree [96,97]. Then, lower discharge voltage and lower practical ability which come from lower active groups in polymeric organic materials resulted in the emergence of a group of porous organic structures named covalent organic frameworks [98]. They are constructed by organic building blocks taking advantage of strong covalent bonds which can bring promising merits including improved stability and good electronic ions infiltration [1,93].

Furthermore, they have a rigid conformation and 1D periodic channels making them suitable to be ion conductors [99]. As a group of rapidly developing porous and crystalline organic compounds, they are formed as two or three-dimensional (2D/3D) structures based on some light atoms comprising C, N, H, O, and Si. It will result in making appropriate lightweight conformations (low density) which can be utilized largely for storage and separation processes (e.g. gas storage and gas separation), semiconductors, sensors, photo/electro catalytic materials, optoelectronic applications, proton conduction, and supercapacitors. COFs can provide a highly accessible surface area with readily adjustable conformation in addition to a low price for the synthesis procedure [100,101]. Here, the most popular applications of COFs in LIBs that are pursued to improve their performance will be described.

4.1 COF-based cathode

As before mentioned, COFs can be used for cathode applications. In 2015, Xu et al. introduced a new category of cathodes for Li batteries as redox-active, crystalline, mesoporous COF on carbon nanotubes. It was made of redox-active naphthalene diimide walls and triphenylene knots and boronate linkages. As a result, boosted electrode stability, appropriate ion transport, and increased conductivity were achieved [102,103].

Wei Zhang et al. reported a truxenone-based COF (COF-TRO) that was able to be applied as cathode materials for all-solid-state LIBs. In this structure, truxenones were used as node building blocks that made small pores with 1.3 nm. Moreover, there were high-density carbonyl groups integrated with the COF structure which provided an ultrahigh specific capacity (268 mAh g⁻¹ experimental capacity at a discharge rate of 0.1 C) and desirable storage by redox reactions. The performance evaluation of these COF-TRO cathodes represented significant cycling stability [101].

A study by Xueliang Sun et al. combined an atomic-layer COF cathode as E-TP-COF with a dual-active-center of C=O and C=N group to boost the capturing and diffusion properties in addition to the capacity output of LIBs. It provided a high initial capacity of 110 mAh g⁻¹ and good retention of 87.3% after 50 cycles [115]. Some of the COF-based cathode

materials in LIBs and their performance are presented in Table 2.

4.2 COF-based anode

Literature illustrated that few COF-based anodes have been reported to now. In fact, most COF-based anodes are still on the experimental scale and they are mainly comprised of conjugated COFs with high-density redox-active sites, COF-based hybrids mostly with

carbon-based compounds and exfoliated COF nanosheets [1]. Although organic electrode materials include lots of advantages like environmentally friendly, valuable theoretical capacity, cost-efficient, and recyclability, some problematic issues such as unsatisfactory stability and low active sites still remained unmet.

Yanli Zhao et al. were able to develop two N-COFs (N₂-COF and N₃-COF) that as anode electrodes for Li

Table 2. Some COF-based cathode materials in LIBs

COF cathode materials	Electrolyte	Potential range (V)	Capacity (mA h g ⁻¹), Current density (mA g ⁻¹)	Number of cycles	Capacity retention (%)	Energy density (W h kg ⁻¹)	Ref
TP-TA COF	1.0 M LiPF ₆ in EC: EMC (3:7, vol%)	1.2–4.3	207, 200	1500	93	621	[104]
HATN-AQ-COF	1.0 M LiTFSI in DOL/DME (1:1)	1.2–3.9	319, 358	3000	80	640	[105]
DAPQ-COF50	1.0 M LiTFSI in DOL/DME (1:1 v/v)	1.5–3.2	162, 500	3000	76	-	[106]
TPPDA-CuPor-COF	LiPF ₆ in EC/EMC (3:7, v/v)	1.7–4.0	142, 60	3000	82	371	[107]
TPPDA-COF	1 M LiPF ₆ in EC/DEC	2.6–4.1	47, 200	3000	65	-	[108]
HATN-HHTP@CNT	1.0 M LiTFSI in dimethoxyethane	1.8–3.5	210, 50	6900	100	-	[109]
USTB-6	1.0 M LiTFSI in DOL/DME (v:v=1:1)	1.2–3.9	285, 200	1000	91	-	[110]
DAPO-COFs	-	1.5–4.2	81.9, 100	1000	72	-	[111]
Phos-COF-1	3 M Na ₂ SO ₄ aqueous solution	-	-	5000	90	32.4	[112]
DAAQ-ECOF	-	1.5–4.0	145, 20	1800	98	-	[113]
DAAQ-TFP-COF	-	1.5–4.0	110, 20	1800	73	161.04	[113]
DAPH-TFP-COF	1.0 M LiPF ₆ in ethylene carbonate/diethyl carbonate (v:v=1:1)	1.4–3.6	70, 100	500	70	221.26	[114]

batteries provided a high charge capacity of up to 700 mA h g⁻¹ with 82% retention after 500 cycles [116]. Moreover, in another study, Huibiao Liu et al. synthesized 2D COF polyporphyrin (TThPP) linked by 4-thiophenephenyl groups to be used as anodes for

LIBs. The satisfying results showed high performance with a reversible capacity of 666 mAh g⁻¹ [117].

Xueliang Sun and coworkers for the first time proposed a special Tp-Azo-COF with a dual active site of N=N and C=O as anode materials for Li batteries.

Table 3. Some COF-based anode materials in LIBs

COF anode materials	Electrolyte	Potential range (V)	capacity (mA h g ⁻¹), Current density (mA g ⁻¹)	Number of cycles	Capacity retention (%)	Energy density (W h kg ⁻¹)	Ref
TA-COF	solid electrolyte interface	0.01-3	473, 500	100	100	-	[133]
CoTAPc-PDA	1 M LiPF ₆ solution in DEC/EMC/EC	0.01-3	204, 100	600	98.5	-	[134]
CoTAPc-BDA	1 M LiPF ₆ solution in DEC/EMC/EC	0.01-3	251, 100	600	99	-	[134]
CoTAPc-TDA	1 M LiPF ₆ solution in DEC/EMC/EC	0.01-3	382, 100	600	98.8	-	[134]
S-COF	LiTFSI-based solid electrolyte	1.7-2.8	110.3, 500	250	88.9	-	[135]
PA-TA-COF	-	0.01-3.0	543, 1000	400	38	216	[136]
SnO ₂ @NCNR-1	solid electrolyte interface film (SEI)	0.005-3.0	694.2, 100	200	-	-	[137]
NA-NiPc	1 M LiPF ₆ in EC/DMC/DEC (1:1:1 by volume)	0.01-3.0	422, 100	700	131.8	-	[138]
PPDA-NiPc	1 M LiPF ₆ in EC/DMC/DEC (1:1:1 by volume)	0.01-3.0	469, 100	700	142.9	-	[138]
DAB-NiPc	1 M LiPF ₆ in EC/DMC/DEC (1:1:1 by volume)	0.01-3.0	566, 100	700	166	-	[138]
COF@CNT	1.0 M LiPF ₆ in EC/DEC 1/1 (v/v)	0.01-3.0	570, 100	2000	65	-	[139]
Tf-TAPA-COF	1 M LiPF ₆ in EC/DMC/DEC (1:1:1 by volume)	0.01-1.5	583, 2000	1500	96	-	[140]
Tp-Ta-COF	solid electrolyte interphase (SEI)	0.01-3.0	413, 200	800	-	-	[141]
Tf-Ta-COF	solid electrolyte interphase (SEI)	0.01-3.0	264, 200	500	-	-	[141]
Tp-Tb-COF	solid electrolyte interphase (SEI)	0.01-3.0	217, 200	500	-	-	[141]
COF _{TpPa}	solid electrolyte interphase (SEI)	-	152.3, -	300	96.8	-	[142]

This system was able to produce high electrochemical kinetics and have a stable structure with a specific capacity of 305.97 mAh g⁻¹ at a density of 1000 mA g⁻¹ after 3000 cycles. In this structure, synergistic functions of azo and carbonyl units helped in improving the structural stability of Tp-Azo-COF when charge and discharge processes were applied [118]. In Table 3, some of the COF-based anode materials in LIBs and their performances are presented.

4.3 Electrolyte materials based on COFs

Nowadays, developing novel and improved performance solid-state electrolytes (SSEs) is crucial to find various applications for LIBs. Traditional liquid electrolytes used in LIBs put dangerous threats such as overheating and combustion [119]. COFs are a group of high-potential candidates for assembling SSEs in LIBs. When they were applied as a solid electrolyte, they represented more advantages because of their high stability and widely tunable structure in comparison to inorganic ion conductors. However, there was a big challenge with the low level of ion conductivities of dry organic compounds. To tackle this issue, a good solution based on the infiltration of COFs with Li salts was proposed to improve ion conductivity. In this line, Kian Ping Loh et al. designed an SE based on COF using hydrazine. This organic porous structure was able to get an ion conductivity of 10⁻⁵ S cm⁻¹ at -40 °C and resulted in a Li⁺ transference number of 0.92. Furthermore, it suppressed the dissolution that occurred by a small organic molecular electrode in all-solid-state batteries [99].

COFs also will provide high electrochemical stability and functional diversity and will be cost-efficient in synthesis processes. Special types of COFs which are developed for SSEs so far include 2D and 3D, cationic and anionic, rigid and flexible, and a composite type namely post-synthetic polymer composite [119] COFs also count as an emerging class of porous polymers useful as electrolytes which can be a good solution for safety issues in a traditional liquid electrolyte in LIBs. Solid polymer electrolytes (SPEs) recently have attracted widespread attention from scientific society to be utilized in LIBs for their high flexibility, lightweight and easy scalability. Since those conventional SPEs are making some drawbacks such as low thermal stability and unsatisfying Li

transference capacity and also are not able to stop dendrite propagation, a new generation of safe-fire COFs is being investigated for SPEs. Zhenjie Zhang et al. designed and fabricated a thermal-responsive imide-linked COF based on a thermal rearrangement protocol that exhibited significant flame retardant characteristics. Furthermore, when polyethylene glycol and Li salt were combined with COF the solid polymeric electrodes will be achieved that have promising flame retardancy with 6.42×10⁻⁴ S cm⁻¹ ionic conductivity and suitable Li-ion transference number (0.95) [120,121].

Research done by Wei Zhang showed a novel example of crystalline imidazolate-containing ICOFs as single-ion conducting COF solid electrolyte materials. Its conductivity was raised up to 7.2 × 10⁻³ S cm⁻¹ and the activation energy was decreased to 0.1 eV. Both the porous structure of ICOF and weak imidazolate binding interactions resulted in promising practical characteristics [122]. In Table 4 some COF-based electrolytes in LIBs and their performance characteristics are summarized.

4.4 COFs as separator

Using COFs for simple and affordable separator applications is also valuable [123]. To prevent an electrical short circuit in batteries, separators keep the anode and cathode from coming into direct contact. Moreover, they operate as electrolyte retention areas to promote ion mobility. Efficient separators are essential for enhancing the rate performance, cycle stability, safety, and other aspects of LIBs [124,125]. Due to their excellent stability, uniform porosity, outstanding mechanical properties, and structural tunability, COFs make excellent prospective separator materials. During the charge and discharge operation, COFs' high stability helps to prevent their breakdown or reactivity with the electrolyte. The capacity to transfer ions, retain electrolytes effectively, and maintain a uniform current density are all made possible by high and uniform porosity. Superior mechanical strength avoids breakage during battery construction and operation, while structural tunability guarantees electrolyte wettability and effective Li⁺ transport [126,127].

In Li-S batteries, the shuttle effect has been successfully reduced, and the use of COFs or COF composites as separators has improved the cycling

Table 4. Summary of COF-based electrolyte properties in LIBs

COF electrolyte materials	Operating temperature (°C)	Metal salt	Activation energy (eV)	Conductivity (S cm ⁻¹)	Transference number	Ref
PVDF/H-COF-1@10	-	LiClO ₄	-	2.5×10 ⁻⁴	0.66	[143]
COF-NUST-7	30	LiTFSI	0.317	4.4×10 ⁻⁷	0.11	[144]
COF-PEG-B6	60	LiTFSI	0.60	3.4×10 ⁻⁶	0.30	[144]
Ge-COF-1	30	LiPF ₆	0.29	2.5×10 ⁻⁴	0.67	[145]
COF-5	-	LiClO ₄	0.037	2.6×10 ⁻⁴	-	[146]
TPB-DMTP-COF	40	LiClO ₄	0.96	1.4×10 ⁻⁷	-	[147]
Q-COF	30	LiTFSI	0.15	7.5×10 ⁻⁵	0.72	[148]
Im-COF-TFSI@Li	30	LiTFSI	0.32	2.9×10 ⁻⁵	0.62	[149]
COF-PEO-3	200	LiTFSI	-	9.7×10 ⁻⁵	-	[150]
COF-PEO-6	100	LiTFSI	-	7.9×10 ⁻⁶	-	[150]
COF-PEO-9	200	LiTFSI	-	1.3×10 ⁻³	-	[150]
Tp-PaSO ₃ Li-COF	20	-	0.13	1.6×10 ⁻³	0.94	[151]

stability of the batteries. To increase the Li⁺ transfer rates, COFs were effectively coated on a polymer separator in LIBs. This prevented transition metal ions from penetrating the separator while also enhancing the transportation of Li⁺. In [Li_{0.2}Mn_{0.55}Ni_{0.15}Co_{0.1}]O₂/Li cell, pure commercial polymer separator demonstrated a low Li⁺ transfer number of 0.38 whereas COF coated commercial polymer separator exhibited a high Li⁺ transfer number of 0.76. [1].

4.5 Challenges of COF-based LIBs

Due to the harsh reaction conditions and the fact that solvothermal techniques are often used in closed environments, it is challenging to synthesize COFs on a large scale for industrial purposes. Although many room temperature synthesis techniques have been suggested, more research on a large base is still required to determine how general these techniques are. Thus, creating large-scale synthesis methods without losing the crystallinity and porosity of COFs is crucial for their actual applications. Furthermore, COFs are polycrystalline powders with numerous structural flaws. Numerous COFs are challenging to control in terms of porosity, crystal domain size, and level of periodicity [1].

On the other hand, Battery performance is limited by COFs' weak electrical conductivity. Enhancing electronic conductivity might be accomplished effectively by including a conductive substance.

Nevertheless, the active components are less concentrated due to this method, which lowers the energy density and gravimetric specific capacity. COF materials with inherent conductivity should be developed to reduce the use of inert material. Designing COFs with conductive connections between monomers through post-synthesis alteration is a method to be investigated. Additionally, it is necessary to enhance the structural stability of COFs or electrodes generated from COFs. If not, the battery will eventually be harmed by the irreversible processes between the electrode contacts [3].

Large numbers of active sites could be buried by bulk COFs with several stacking layers, making it difficult to access and use redox-active sites. Furthermore, stacking layers results in slower Li-ion diffusion, increased routes for lithium diffusion, and reduced lithium transfer kinetics. Several organic electrodes, including COFs, still have lower energy densities than many inorganic electrodes. The energy density of batteries can be increased by rationally designing COF

structures and optimizing the density of redox-active sites on both backbones and substituents. Even though COFs are far more stable than small organic molecules, their capacity to discharge and charge over extended periods is still limited by the insufficient stability of COFs in organic electrolytes [1]. COFs for battery applications are still in the initial phase, and it is hoped that by improving the properties of these materials and methods, these challenges will be resolved as soon as possible.

5. Conclusion and prospect

With an emphasis on the fundamentals of LIBs and COF design and synthesis principles, we have outlined the most recent developments in COFs and COF-derived materials for LIBs in this review. Additionally, we investigated the application of COFs in the cathode, anode, electrolyte, and separator of LIBs. COFs have much promise for use in battery applications. The sturdy framework and porous channel structure provide exceptional cycle stability and rate performance. The various adjustable components allow COFs to perform better than their competitors. The uniform 1D channels with accessible active sites allow COFs to achieve outstanding performance and quick electrochemical responses. Furthermore, frameworks with improved chemical and physical stability can give LIBs greater longevity. The numerous applications of COFs in LIBs have been amply illustrated by the instances mentioned earlier. These accomplishments are extremely motivating for the ultimate use of COFs in practical and commercial energy storage and transformation devices, despite some issues and challenges. More theoretical and experimental works are needed to be done in the future to solve the existing challenges of using COFs in LIBs.

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