# Chemical Design for Both Molecular and Morphology Optimization toward High-Performance Lithium-Ion Batteries Cathode Material Based on Covalent Organic Framework

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In most cases, to obtain high-performance electrode materials for lithiumion batteries (LIBs), it is necessary to optimize both their molecular structure and morphology. Normally, the molecular structure of covalent organic frameworks (COFs) can be well engineered by chemical design, while their morphology is mainly optimized by post-processing. Herein, by introducing a flexible building unit containing sp<sup>3</sup> N redox-active centers, a bipolar-type TP-TA COF assembled by uniform 2D hexagonal nanosheets is synthesized in a one-step reaction without any post-processing, achieving the highly challenging simultaneous optimization of both molecular structure and morphology required for high-performance electrode materials. Thus, when used as cathode material for LIBs, its combined optimized chemical structure and favorable morphology of TP-TA COF synergistically render a high capacity (207 mA h g<sup>-1</sup> at 200 mA g<sup>-1</sup>), excellent rate performance (129 mA h g<sup>-1</sup> at 5.0 A g<sup>-1</sup>), and cycling stability (93% capacity retention after 1500 cycles at 5.0 A g<sup>-1</sup>).

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## 1. Introduction

In recent years, with the increasing demands for large-scale energy storage systems and electric vehicles, many efforts have been devoted to developing high-performance electrode materials for lithium-ion batteries (LIBs).<sup>[1]</sup> In theory, the performance of electrode materials, such as theoretical capacity, redox potential, kinetics, and cycling stability, are essentially determined by their molecular structure.<sup>[2]</sup> However, in practical terms, the morphology of electrode materials also has significant influence on their practical behaviors, especially for rate performance.<sup>[3]</sup> For instance, compared to bulk electrode materials, their nanoscale counterparts with the same chemical structures usually expose more redox-active sites and also have minimized diffusion length

for the transport of ions/electrons, thus delivering a higher actual capacity and enhanced rate performance.<sup>[4]</sup> Therefore, to obtain practical high-performance electrode materials, both their structure and morphology need to be optimized. In most cases, the optimization or tuning of chemical structure and morphology are carried out separately. For example, the desired morphology of electrode materials is mainly achieved by post-treatment.<sup>[2b,4b,5]</sup> Thus, it is still a great challenge to tune the molecular structure and morphology simultaneously through chemical design to obtain ideal electrode materials with overall desired performance.

At present, in practice cathode materials are probably the main bottleneck for LIBs in terms of capacity due to their low theoretical limit compared to that of anode materials.<sup>[3b]</sup> With the advantages of high capacity, abundant resource, and low-cost, various organic compounds have been developed for high-performance cathode materials.<sup>[6]</sup> Among them, covalent organic frameworks (COFs) show a distinctive advantage for improving cycling performance owing to their good framework stability.<sup>[7]</sup> Moreover, benefiting from the structural diversity and designability of COFs, the electrochemical performance of COFs can be easily tailored by rational chemical design.<sup>[5a]</sup> For instance, our group and many other studies have shown that



Figure 1. The molecular structure and morphology of TP-TA COF and its application as a high-performance cathode material for LIBs.

maximizing the content of redox-active groups in COFs is an effective strategy to design cathode materials with high theoretical capacity and good cycling stability.<sup>[8]</sup> In addition, a few bipolar-type COFs cathode materials with integrated p- and ntype redox moieties have been reported to expand the operating potential windows of them, leading to high specific capacity and increased energy density of LIBs.<sup>[9]</sup> However, despite numerous advantages mentioned above, most of COFs don't display outstanding overall performance in practice. One major reason is that many of them are constructed by rigid aromatic building units, and the resulting strong interlayer  $\pi$ - $\pi$  interactions usually lead them to exhibit undesired bulk morphology with tightly stacking layers.<sup>[4b,10]</sup> As a result, it is hard for these 2D COFs to fully utilize their interior redox-active sites and exhibit desired ions/electrons transport, thus leading to a low actual capacity and poor rate performance.<sup>[4b]</sup> Thus, to improve their practical performance, some post-processing strategies have to be taken to optimize the morphology of COFs, such as exfoliating pristine COFs into few-layer COF nanosheets (CONs).<sup>[4b,c,7c,8b,10,11]</sup> Above the tedious process and likely high cost, the yield of CONs of such processes is generally low, and furthermore, it is difficult to precisely control the thickness and size of the as-prepared CONs.<sup>[4b,12]</sup> All these could be a big hinder for their large-scale preparation and application. So far, it is still a challenge to directly obtain CONs with uniform size without post-processing.

In this study, by introducing a flexible building unit containing sp<sup>3</sup> N redox-active centers and constructed C=N redoxactive linkages, we have achieved the simultaneous optimization of both structure for high theoretical capacity and morphology for practical performance. By the well-known one-step simple solvothermal method, a 3D flower-like COF (namely TP-TA COF) assembled by uniform and ultrathin 2D hexagonal nanosheets, containing both C–N and C=N redox-active groups, was synthesized directly without any post-treatment. When used as the cathode material for LIBs, the bipolar-type feature and the favorable morphology of TP-TA COF work cooperatively, endowing TP-TA COF superior overall electrochemical performance, delivering simultaneously a high capacity (207 mA h g<sup>-1</sup> at 200 mA g<sup>-1</sup>), excellent rate performance (129 mA h g<sup>-1</sup> at 5.0 A g<sup>-1</sup>), and superior cycling stability (93% capacity retention after 1500 cycles at 5.0 A g<sup>-1</sup>).

## 2. Results and Discussion

#### 2.1. Design and Synthesis

According to the discussion above, to develop ideal COF-based cathode materials with overall high performance, the main design criteria should be: 1) high theoretical capacity, meaning that the COF should contain maximized redox-active groups per repeating unit;<sup>[8a]</sup> 2) high redox potential, one effective strategy is to introduce p- or bipolar- type redox-active moieties in the molecular structure;<sup>[13]</sup> 3) favorable morphology to materialize high actual capacity and good rate performance, ideally obtained from a one-step reaction without any post-treatment; 4) long-term cycle life, requiring the COF with a stable framework structure.

Taking the above requirements into consideration, we chose N,N,N',N'-tetraphenyl-1,4-phenylenediamine (TP) and terephthalaldehyde (TA) as the building units, successfully designing an ideal TP-TA COF cathode material that can meet the criteria mentioned above at the same time. Notably, as shown in **Figure 1**, the molecular structure of TP unit contains two sp<sup>3</sup> N (C–N group) redox-active centers, and the TA unit can offer reaction-active sites to construct redox-active C=N linkages into TP-TA COF skeleton during the polycondensation of TP and TA. Therefore, the as-obtained TP-TA COF contains multiple C–N and C=N redox-active groups, which effectively increases the content of redox-active groups in the molecular structure and thus improves the theoretical capacity of TP-TA



COF. In addition, when TP-TA COF is used as cathode for LIBs, the C-N groups can experience p-type doping reaction and the C=N linkages can undergo n-type doping process. The synergistic utilization of C-N and C=N groups endows TP-TA COF a bipolar-type feature, expanding the operating voltage of TP-TA COF, which enables to deliver high energy density for LIBs. Moreover, the sp<sup>3</sup> N in TP molecule renders it spatial flexibility.<sup>[14]</sup> which is beneficial to weaken the interlayer  $\pi$ - $\pi$  interactions of TP-TA COF and suppress the over stacking of layers, promising to display a desired morphology directly. As shown in Figure 1, TP-TA COF could be obtained by the well-known one-step solvothermal method without any postprocessing and more details are given in Experimental Section (Section S1, Supporting information). Moreover, the as-obtained TP-TA COF is found to be assembled directly by uniform and ultrathin 2D hexagonal nanosheets in the reaction process with a 3D flower-like morphology, which is important to achieve high actual capacity and rate performance. Besides, the stable framework of TP-TA COF renders it excellent cycling stability. Therefore, when used as cathode material for LIBs, TP-TA COF is capable of delivering high capacity and energy density, outstanding rate, and cycling performance all together.

#### 2.2. Structural and Morphological Characterization

The obtained TP-TA COF was first characterized by Fourier transform infrared spectroscopy (FTIR) (Figure S1, Supporting Information). The emerging absorption peak at 1616 cm<sup>-1</sup> in the FTIR spectrum of TP-TA COF indicates the formation

of the C=N linkages.<sup>[14c]</sup> In the solid-state <sup>13</sup>C NMR spectra (Figure S2, Supporting Information), the chemical shift of around 156.38 ppm further verifies the formation of the C=N linkages.<sup>[15]</sup> Moreover, the X-ray photoelectron spectroscopy (XPS) spectrum and elemental analysis of TP-TA COF (Figure S3, Table S1, Supporting Information) also confirm its chemical composition and structure.<sup>[8a]</sup> Besides, TP-TA COF shows an outstanding thermal stability approaching 450 °C in N<sub>2</sub> (Figure S4, Supporting Information).

As shown in **Figure 2**a, the powder X-ray diffraction (PXRD) pattern of TP-TA COF (blue curve) exhibits prominent peaks at 2.42°, 4.16°, 4.76°, 6.36°, and 8.69°, which are assigned to the (100), (110), (200), (210), and (300) facets, respectively, indicating the good crystallinity of TP-TA COF. In addition, the peak at around  $2\theta = 21.03^{\circ}$  is ascribed to the (001) plane representing a relatively large  $\pi$ - $\pi$  stacking distance of 4.22 Å, matching with the AA stacking model with *d* spacing of 4.22 Å. The expanded interlayer distance of TP-TA COF should be attributed to the integration of flexible (sp<sup>3</sup> N) building unit TP into the skeleton of TP-TA COF.<sup>[14]</sup> The unit-cell parameters in Figure 2a are obtained by Pawley refinement for TP-TA COF (Section S4, Supporting Information). Besides, the PXRD pattern (blue curve) is well consistent with the Pawley-refined XRD profile (black curve), as evidenced by their negligible difference (grey curve).

High-resolution transmission electron microscopy (HRTEM) images (Figure 2c; Figure S5a, Supporting Information) exhibit clear lattice fringes with an interlayer distance of 2.3 Å that is close to the interspacing of (1600) plane, and the SAED pattern (Figure S5b, Supporting Information) shows obvious diffraction spots, both of which indicate a good crystallinity of TP-TA



**Figure 2.** a) The experimentally observed PXRD pattern of TP-TA COF (blue), Pawley refinement (black), the difference curve (gray), and the crystal structure of the AA stacking with an interlayer distance of 4.22 Å. b,c) HRTEM images of TP-TA COF. d) Pore size distribution profile and chemical structure of TP-TA COF with distinct dual-pore skeleton. e,f) SEM images of TP-TA COF.



COF. As shown in Figure 2d and Figure S5, Supporting Information, the micropore of 1.27 nm and mesopore of 2.52 nm in diameter are consistent with the dual-pore skeleton of TP-TA COF comprised of theoretical triangular (1.25 nm) and hexagonal (2.77 nm) pores, respectively. The micro-meso-porosity of TP-TA COF forms a hierarchical pore characteristic, which can facilitate the ion/electrons transport and provide enough space to buffer the volume change during ions insertion/extraction process, enhancing the rate and cycling performance.

Besides, as mentioned above, the introduction of flexible molecule TP renders TP-TA COF to have weakened the  $\pi$ - $\pi$ stacking interaction and thus achieves the desired morphology for practical high performance. Indeed, the scanning electron microscopy (SEM) image (Figure 2e) of TP-TA COF exhibits a distinctive 3D hierarchical flower-like morphology assembled by 2D nanosheets. Furthermore, Figure 2f shows that most of these 2D nanosheets display a uniform hexagon shape with clear edges, which is consistent with the TEM image of TP-TA COF in Figure 2b. The lateral size of these 2D hexagonal nanosheets is mainly distributed in 1.15-1.20 µm obtained by SEM statistical analysis (Figure S7, Supporting Information), and the thickness of the 2D nanosheets is observed around 4 nm using AFM (Figure S8, Supporting Information). As far as we know, this is the first time that such a 2D COF assembled by uniform 2D hexagonal nanosheets is directly prepared by a one-step solvothermal method, which is of great significance

for large-scale preparation and practical application of ultrathin CONs with uniform size and thickness. Particularly, compared to most of COFs with bulk morphology, pristine TP-TA COF can be used directly as cathode material without exfoliation or other post-treatments. The 3D flower-like morphology assembled by ultrathin 2D nanosheets in TP-TA COF can provide more redox-active sites and also minimize the diffusion length for ions/electrons, delivering a higher capacity and enhanced rate performance.

#### 2.3. The Formation Process of 3D Flower-Like Morphology

In order to investigate the formation process and mechanism of the unique morphology of TP-TA COF, a series of timedependent products were isolated from the polycondensation of TP and TA at different reaction stages (**Figure 3**; Figure S9, Supporting Information), and the morphological change of TP-TA COF during the growth process was investigated by SEM and TEM. As shown in Figure 3a, after reaction for 3 h, small aggregated nanosphere crystallites were formed with a size of ≈100 nm, and after 6 h, the aggregated nanoparticles with larger size were observed (Figure S9b, Supporting Information). However, after 12 h, the starfish-shaped aggregates are observed in Figure 3b. After reaction time of 48 h, it was worth noting that some starfish-shaped aggregates gradually



Figure 3. The proposed formation mechanism of 3D hierarchical flower-like TP-TA COF, and the corresponding SEM and TEM images of the obtained products recorded at different reaction stages: a) 3, b) 12, c) 48, and d) 120 h.



transformed into 2D hexagonal nanosheets (Figure 3c). After 72 h, as shown in Figure S9e, Supporting Information, almost all of starfish-shaped aggregates transformed into 2D hexagonal nanosheets with a scattered distribution. After 120 h, the obtained products exhibited a 3D hierarchical flower-like morphology constructed by 2D nanosheets (Figure 3d).

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Based on above results, we propose the following growth mechanism for the morphology of TP-TA COF (Figure 3; Table S2, Supporting Information). Initially, small nanospheres crystallites appear (Figure 3a), corresponding to the initial imine-condensation products.<sup>[16]</sup> With increasing reaction time, larger and aggregated nanoparticles generate (Figure S9b, Supporting Information), which can be explained by the fact that the initial crystallites further grow by condensation of unreacted monomers.<sup>[16]</sup> After 12 h, these nanoparticles transform into the starfish-shaped aggregates (Figure 3b), which might be attributed to that the adjacent nanoparticles fusion occur by reactive aldehyde and amino groups at their interfaces.<sup>[16]</sup> After 48 h, some regular 2D hexagonal nanosheets (Figure 3c) appear, indicating the crystallinity growth of as-obtained products.<sup>[17]</sup> The PXRD characterization at different reaction times (Figure S10, Supporting Information) also indicates that the crystallinity of the as-obtained products is enhanced gradually over time. As the crystallinity increases, the skeleton of TP-TA COF continues to expand in the in-plane direction, and the 2D layers gradually stack parallelly in the out-of-plane direction with the formation of the pore channels. However, the introducing of flexible building unit into the COF skeleton efficiently weakens the interlayer  $\pi$ - $\pi$  stacking interactions of TP-TA COF.<sup>[14b,14c]</sup> As a result, the further stacking of the 2D layers along the out-ofplane direction is suppressed, while the growth of crystallites in the in-plane dimensions continues, thus promoting the formation of 2D nanosheets. The hexagonal shape of 2D nanosheets is presumably induced by the high stability of the hexagonal geometric structure.<sup>[18]</sup> In addition, we note that the obtained 2D nanosheets after 72 h are scattered and have not yet observed a 3D flower-like morphology (Figure S9e, Supporting Information). After 120 h, the 3D flower-like morphology constructed from 2D nanosheets appears (Figure 3d), and the lateral size of these 2D nanosheets is almost the same as that of nanosheets obtained after 72 h. Thus, we think that the formation of the 3D flower-like morphology could be explained by a self-assembly process for minimizing the surface energy.<sup>[14a]</sup> As can be seen from the above experimental results and analysis, the flexible building unit in the COF skeleton has an important role on their morphology and the reaction time also plays a great role on the crystallinity and morphology of COFs.

#### 2.4. Electrochemical Performance

With the desired molecular structure and favorable morphology in nanoscale, TP-TA COF is expected to show overall optimized electrochemical performance. The electrochemical performance of TP-TA COF for LIBs was evaluated as cathode material by using coin type cells, where metal lithium was used as counter/ reference electrode and 1 mmm LiPF<sub>6</sub> in EC: EMC (3:7, vol%) was used as electrolyte. As shown in **Figure 4**a, the bipolar-type feature of COF enables it to perform multi electron/stage transfer redox reactions in the broad potential window of 1.2–4.3 V versus Li/Li<sup>+</sup>. When the potential is above 2.8 V versus Li/Li<sup>+</sup> (open-circuit voltage), TP-TA COF cathode shows clear two pairs of redox peaks at 3.60/3.51 and 3.88/3.84 V versus Li/Li<sup>+</sup>, respectively, which are assigned to the reversible p-type doping



**Figure 4.** a) The cyclic voltammetry curve at a scan rate of 0.5 mV s<sup>-1</sup>. b) The initial three-cycle capacity–voltage profile of TP-TA COF at a current of 200 mA h g<sup>-1</sup>. c) Cycling performance of TP-TA COF at a current of 200 mA g<sup>-1</sup> for 250 cycles. d) Rate capabilities of TP-TA COF at different current densities. e) Long-term cycling performance of TP-TA COF at a current of 5 A g<sup>-1</sup> for 1500 cycles.





process of C–N active centers.<sup>[19]</sup> The redox peaks in the potential range of 1.2–2.8 V versus Li/Li<sup>+</sup> are attributed to reversible n-type doping reactions of C=N groups.<sup>[20]</sup> Benefiting from the bipolar-type feature of TP-TA COF, a high specific capacity of 207 mA h g<sup>-1</sup> can be delivered at a current density of 200 mA g<sup>-1</sup>, and the corresponding average redox potential can reach up to 3 V, thus rendering a measured high energy density of 621 W h kg<sup>-1</sup>. We further evaluated the cycling performance of TP-TA COF under a current of 200 mA g<sup>-1</sup>, as shown in Figure 4c, the result exhibits a high capacity of 145 mA h g<sup>-1</sup> after 250 cycles, which implies the good redox reversibility of TP-TA COF cathode for LIBs.

Furthermore, as stated earlier, the morphology of TP-TA COF assembled by ultrathin 2D nanosheets is capable of affording fast ion/electrons diffusion and more accessible redox-active sites, which is beneficial to achieve excellent rate performance for TP-TA COF. Thus, the rate capabilities of TP-TA COF were investigated by the galvanostatic charge/ discharge tests (Figure 4d). Notably, as the current increased from 1 to 5 A  $g^{-1}$ , the capacities of TP-TA COF decay only slightly, and it can deliver a high reversible capacity of 129 mA h  $g^{-1}$  even at a high current density of 5 A  $g^{-1}$ , which is superior to many previous related reports (Figure S11, Table S3, Supporting Information).<sup>[4c,9a,13,21]</sup> The corresponding power density can be as high as 14.5 kW kg<sup>-1</sup> (Figures S12,S13, Supporting Information). The remarkable rate performance of TP-TA COF convincingly demonstrates the advantages of its morphology. To investigate the kinetics of TP-TA COF, electrochemical impedance spectroscopy of TP-TA COF was

measured. As shown in Figure S14, Supporting Information, the charge-transfer resistance ( $R_{ct}$ ) of the half-cell is as small as 24.2  $\Omega$ , which suggests a fast and stable reaction kinetic of TP-TA COF as cathode material for LIBs.<sup>[8c]</sup>

In addition, compared to other organic electrode materials, the stable framework of TP-TA COF can effectively suppress its dissolution in electrolyte and improve its cycling stability. Moreover, the 3D flower-like morphology of TP-TA COF provides enough space to buffer the volume change during ions insertion/extraction process, which is also contributed to its cycling performance. Indeed, as shown in Figure 4e, even at a higher current density of 5 A g<sup>-1</sup>, TP-TA COF can still afford a reversible capacity of 122 mA h g<sup>-1</sup> after 1500 cycles with a capacity retention of 93%, revealing a superior cycling stability of TP-TA COF.

#### 2.5. Redox Mechanism

The high specific capacity and broad potential window of TP-TA COF is closely related to its bipolar-type feature. Thus, we proposed a bipolar redox mechanism for TP-TA COF cathode material during its electrochemical process. The C–N redox-active groups can reversibly accept/release anions in the electrolyte, and C=N redox-active groups can reversibly insert/ extract metal cations such as Li<sup>+</sup>.<sup>[13a,22]</sup> As for TP-TA COF, in **Figure 5**, the structural evolution of the whole reversible redox process of TP-TA COF can be divided into four stages. In the initial charge process, TP-TA COF undergoes an oxidization



Figure 5. The proposed redox mechanism during the reversible electrochemical process of TP-TA COF.





reaction by losing two electrons from its pristine state, during which two C-N redox-active groups combine with two PF<sub>6</sub><sup>-1</sup> anions from the electrolyte to remain electroneutral, forming N-PF<sub>6</sub><sup>-1</sup> interaction. For the next two stages, continuous discharge processes are carried out. TP-TA COF is first reduced to the neutral state from its oxidized state by accepting two electrons, accompanying the extraction of two  $PF_6^{-1}$  anions. Subsequently, TP-TA COF is further reduced with the acceptance of four electrons, forming the fully reduced state, and four C=N redox-active groups are transformed into C-N-Li bonds with the insertion of four Li<sup>+</sup>. In the fourth stage, during the charging process, TP-TA COF is oxidized by losing four electrons and recovers to the initial state from its reduced state, which is accompanied by the de-intercalation of four Li<sup>+</sup>. In conclusion, the first two stages are related to the p-type doping redox processes of the C-N moiety, while the latter two processes are attributed to the n-type doping reactions of the C=N groups. When used as the cathode material for LIBs, the C-N and C=N redox-active groups work synergistically, providing a high theoretical capacity of 241 mA h g<sup>-1</sup> (corresponding to 6 electrons transport per repeating unit of TP-TA COF, Section S6, Supporting Information).

To gain more insight into the redox mechanism of TP-TA COF cathode for LIBs, ex situ XPS was performed to analyze its structural changes during the redox process. As shown in Figure S15a, Supporting Information, after charged to 4.3 V, a new N peak emerges at 402.45 eV, revealing the interaction between the C-N redox centers and  $PF_6^{-1}$  anions at the fully charged states.<sup>[13a]</sup> When discharged to 1.2 V, the peak of the N-PF<sub>6</sub><sup>-1</sup> interaction at 402.45 eV diminishes, while a peak at 398.6 eV corresponding to C-N-Li bond is observed.<sup>[23]</sup> This suggests that the removal of  $PF_6^{-1}$  anion from the C–N redox-active center as well as the insertion of Li+ into the C=N bond occurred during the discharge process. Besides, the peak assigned to PF6-1 in the high-resolution F 1s spectra (Figure S15b, Supporting Information) appears at charged state but diminishes at discharged state, and the P 2p spectra (Figure S15c, Supporting Information) also show the similar trend, which indicates the reversible insertion/de-insertion of PF<sub>6</sub><sup>-1</sup> ions into TP-TA COF during the charge/discharge process, revealing the p-type feature of TP-TA COF.<sup>[13a]</sup> At the same time, the change of the high-resolution Li 1s spectra (Figure S15d, Supporting Information) is opposite to that of the F 1s spectra, which suggests the insertion/de-insertion of Li<sup>+</sup> during the discharge/charge process, showing the n-type characteristic of TP-TA COF. These ex situ XPS results further prove the proposed bipolar-type redox mechanism of TP-TA COF as cathode material for LIBs.

## 3. Conclusion

In summary, by introduction of a flexible building unit, we synthesized a high-performance bipolar-type TP-TA COF assembled by ultrathin 2D hexagonal nanosheets, achieving simultaneous optimization for the structure and morphology of TP-TA COF. Furthermore, such a COF assembled by uniform 2D hexagonal nanosheets is directly prepared by a one-step solvothermal method without any post-processing, which is of

great significance for the large-scale preparation and application of COFs. When used as cathode material for LIBs, the bipolartype feature and favorable morphology of TP-TA COF synergistically afford a high capacity of 207 mA h g<sup>-1</sup> at 200 mA g<sup>-1</sup>, excellent rate performance of 129 mA h g<sup>-1</sup> at 5.0 A g<sup>-1</sup>, and good cycling stability of 93% capacity retention after 1500 cycles at 5.0 A g<sup>-1</sup>. This work is expected to offer an efficient way to develop high-performance electrode materials for the next generation LIBs by simultaneously tuning the molecular structure and morphology of COFs.

## 4. Experimental Section

Experimental procedures can be found in the Supporting Information.

## **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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## **Conflict of Interest**

The authors declare no conflict of interest.

## **Data Availability Statement**

Research data are not shared.

## Keywords

2D material, bipolar-type cathode material, covalent organic frameworks, hexagonal nanosheets, lithium-ion batteries

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