



Review

Hotspots and trends of covalent organic frameworks (COFs) in the environmental and energy field: Bibliometric analysis

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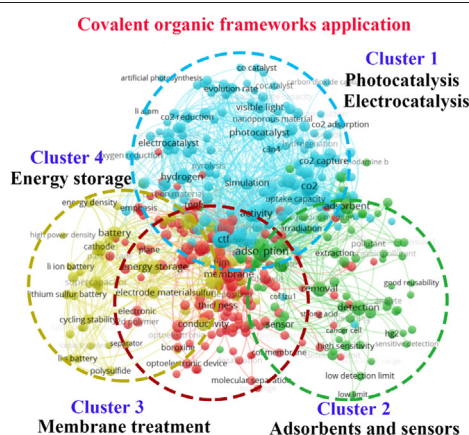
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HIGHLIGHTS

- For the first time, research status of COFs is presented by bibliometrics method.
- The high frequency of “photocatalysis” reflects the current research hotspot.
- The linkage, topology, and synthesis method of COFs was summarized.
- The environmental and energy application of COFs were discussed in details.
- Prospects and challenges for COFs field are put forward.

GRAPHICAL ABSTRACT



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ABSTRACT

Covalent organic frameworks (COFs) have attracted extensive attention due to their low density, adjustable structure, functionalization, and good stability. This paper systematically and comprehensively describes to qualitatively and quantitatively the progress, trends, and hotspots of COFs in the environmental and energy fields from the perspective of bibliometrics. Herein, based on the Web of Science database, a total of 2589 articles from 2005 to October 6, 2020, were collected. Thereafter, co-occurrence, co-citation analysis, and cluster analysis were conducted using CiteSpace and VOSviewer software. The results indicated that COFs research shows the characteristics of rapid growth. The active countries were mainly USA, Germany, Japan, China, and India. More than half of the top 20 active institutions were from China. The research hotspots in this field were systematically elaborated, including synthesis, adsorption, catalysis, membrane, sensor, and energy storage. Research has shown that various COFs are reasonably designed, synthesized, and used in different applications. For example, when COFs are used for photocatalysis, groups containing photocatalytic active sites are integrated into COFs to improve photocatalytic activity. Finally, some challenges were proposed, that are beneficial to the rapid and balanced development of the COFs field. For instance, the preparation methods still need to be further improved for mass production and there is an imbalance in environmental applications such as fewer sensor and membrane applications. We believe that this study provides a comprehensive and systematic overview of the environmental and energy applications of COFs for future investigations.

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1. Introduction

Covalent organic frameworks (COFs), a type of novel crystalline organic porous material, are composed of light elements (H, B, C, N, O), which have broadly attracted the interest of many researchers owing to their advantages of low density, dominant crystallization, periodic pore channels, designable structures and adaptable functionalization (L.H. Li et al., 2017; Zhou et al., 2019). In 2005, the first COFs (COF-1, COF-5) were synthesized for the first time by Yaghi et al. (Cote et al., 2005). In 2009, their research group proposed for the first time that COFs could be used as good adsorbents for gases such as dihydrogen, methane, and carbon dioxide, which paved the way for COFs field applications (Furukawa and Yaghi, 2009). Afterwards, COFs have given rise to extensive applications in gas storage (Gao et al., 2018; (Braunecker et al., 2020)), adsorption (Y. Yang et al., 2020), and so on. In recent years, with the rapid development of COFs, applications such as catalysis (Jin et al., 2020; C.-C. Li et al., 2020; H. Liu et al., 2020), sensor (Tian et al., 2020; X.Y. Wang et al., 2020), and energy storage (Sun et al., 2020a) (; Wu et al., 2020) have received widespread attention. Many strategies, including controlling the sequence of reactions (post-synthesis (Segura et al., 2019), multi-component one-pot reaction (X.T. Li et al., 2020)) and dominating structure (chiral induction (Dong et al., 2020), defect engineering (Luo et al., 2020)), are effective strategies to satisfy the demands of functional diversity.

Some review literatures have reported the development, application, and future directions of COFs (Ding and Wang, 2013; Segura et al., 2016; Cao et al., 2019; X. Liu et al., 2019; Vardhan et al., 2019; X. Cui et al., 2020; Jarju et al., 2020; J. Li et al., 2020; Z. Wang et al., 2020). However, COFs, as a new material, urgently requires an analysis of its development trajectory, research status, and trends of rapid development in recent years, which helps readers quickly determine subsequent research directions. To clarify the multi-factor analysis of COFs

area, a bibliometric method was applied to review the evolution in the field of COFs. By applying bibliometrics to the research in the field of COFs, one can obtain a visual analysis in this field, which would help researchers to grasp the overview of the subject of COFs, determine the areas of inter-disciplinary and collaborative research, identify the most active research group, and promote the development of the subject. To the best of our knowledge, an overview of the research trends in the field of COFs not been reported.

Bibliometric is a hotspot in current research, and it is defined as the quantitative analysis of scientific publications via statistical methods. It not only can be used to estimate the impact of research areas and identify emerging trends (Pritchard, 1969), but also can depict the characteristics, hotspots, and emerging trends of a given field (Zhi et al., 2015; Y. Zhang et al., 2017). This method has several advantages such as the following: evaluation of the productive journals that publish research in a particular field; providing information on the most active institutions, countries, authors; identifying the collaboration between authors, countries, and institutions, and determining the research hotspots and future directions (Li and Zhao, 2015); further, bibliometric analysis has recently played an increasingly significant role in scientific research (Alexandre-Benavent et al., 2017). Thus far, a bibliometric method has been developed in the field of environmental science (Zhao et al., 2018; (Blinova et al., 2020); Teixeira et al., 2020), material science (Fonteyn et al., 2020; Yilmaz et al., 2020), chemistry (Z. Wang et al., 2019; Yang and Ho, 2019), and energy (Yu and He, 2020). As it is very difficult to obtain valuable information from tens of thousands of literatures, this requires computer assistance. Since the introduction of auxiliary visualization in bibliometrics, many useful scientific knowledge mapping tools (e.g., VOSviewer CiteSpace, CitNetExplorer, BibExcel, HistCite, etc.) have also been generated. CiteSpace and VOSviewer are the most commonly used tools owing to their characteristics of easy operation and multifunction.

Herein, we review the research on COFs fields via bibliometric analysis using CiteSpace and VOSviewer software. A total of 2589 published articles are discussed. The co-occurrence analysis of countries, institutions, and keywords are visualized, and the co-cited analysis of authors, journals, and references are discussed. The above analysis is used to understand the research developments in the field of COFs. Furthermore, current research hotspots are also investigated.

2. Method and content

2.1. Data collection

The data were obtained from the SCI-expand database, which is a part of the Web of Science (WOS) Core Collection. The WOS Core Collection was chosen since it is considered the most comprehensive database, embodying the most influential and relevant journals (Olawumi and Chan, 2018). An advanced search was performed on the literature, and the specific requirements were as follows: (1) the search topic (TS) = ("covalent organic framework*" OR "covalent organic-framework*" OR "covalent-organic framework*" OR "covalent triazine framework*" OR "covalent triazine-based framework*"). It is noteworthy that abbreviations such as COF(s) and CTF(s) were not considered. The abbreviated words were used as the search topic to provide the number of documents in irrelevant fields such as biology and automation systems, which make the data unrepresentative and inaccurate. Moreover, the full form of an abbreviation can be found when it is used for the first time in the article. This means that the literature can still be searched, even if the full form is used. (2) The search data was

set from 2005 to October 6, 2020 because the first article related to COFs was published on 2005. (3) The language of the article was set to English. (4) In order to obtain accurate articles related to COFs materials, nine WOS categories were finally chosen by excluding irrelevant categories. They are "Engineering Environmental, Environmental Science, Water Resources, Green Sustainable Science Technology, Chemistry Multidisciplinary, Materials Science Multidisciplinary, Nanoscience Nanotechnology, Energy fuels, Electrochemistry." Then, a second round of manual screening was conducted based on the title, abstract, and content. The document selection and flow diagram of COFs are illustrated in Fig. 1.

2.2. Analysis method

The data from the WOS Core Collection were further analyzed by bibliometrics. The document types and numbers, co-occurrence analysis (e.g., countries, institutions, and authors), research hotspot analysis (keywords co-occurrence and cluster), and co-cited analysis (e.g., journals, authors and references) were conducted using CiteSpace and VOSviewer software. In addition, Microsoft Excel 2020 and Origin 9.0 were used to analyze the data and plot graphs. The co-cited analysis describes the relationship of two or more authors (journals, references), which were cited by the same authors (journals, references) at the same time (Kolahi et al., 2017). In the network of co-occurrence and cluster analysis, the nodes represent the specific key terms such as countries, institutions, or keywords, where the larger the node size, the higher the frequencies. The color of a node indicates the frequencies every year. The bigger the purple circle, the greater the

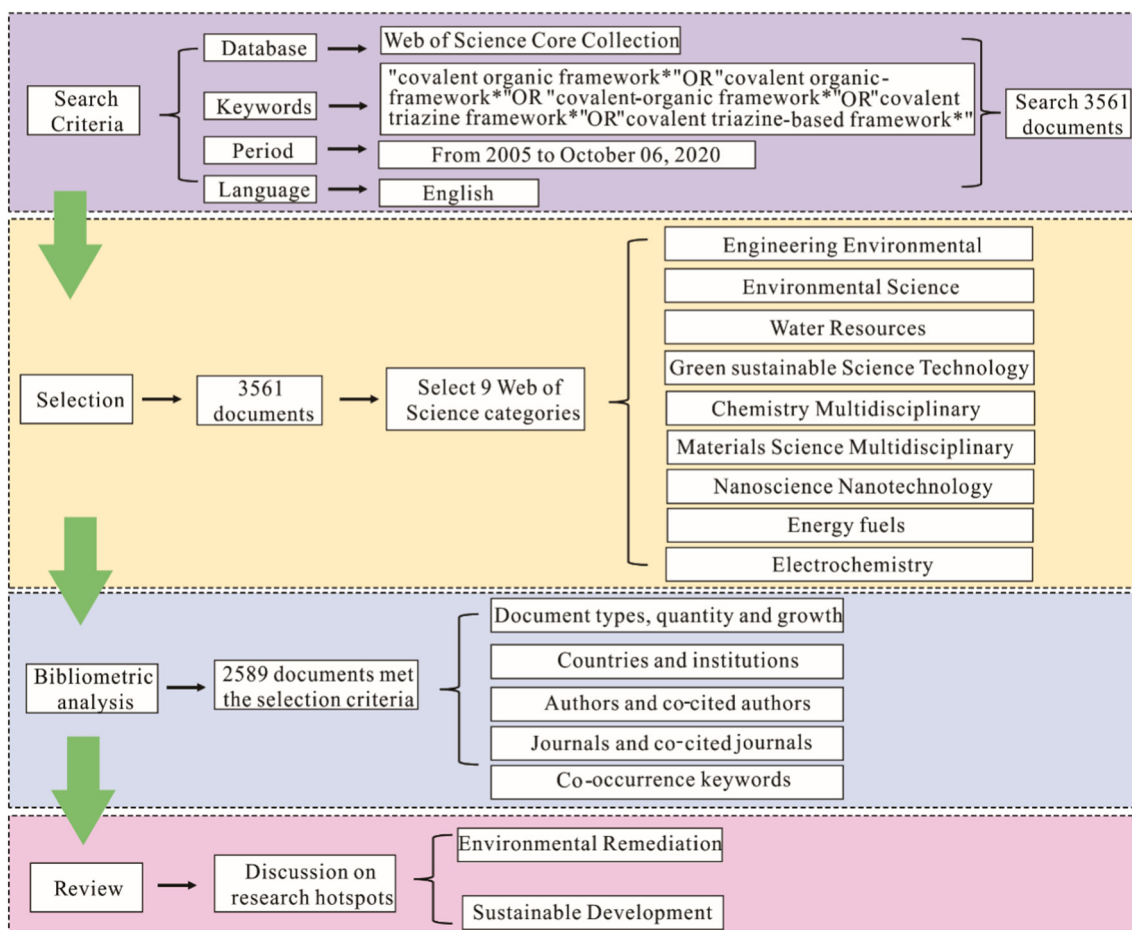


Fig. 1. Document selection and flow diagram of COFs.

importance of the node. The thicker the line, the closer the collaboration relationship. Additionally, the cluster network was formed by grouping closely connected keywords into one category based on VOSviewer software.

3. Results and discussion

3.1. Document types and numbers

A total of 2589 documents were obtained from the SCI-expanded database of the WOS. Depending on the type of documents, the documents were divided into nine types including article, review, meeting abstract, early access, editorial material, correction, proceedings paper, news items and book chapters (Fig. 2). Notably, the “article” type ranked first with a proportion of 81.22%, followed by the “review” type with a proportion of 10.11%. It was worth noting that the proportion of “review” type on COFs field was higher than other areas such as microplastics (8.01%), groundwater remediation (3.60%) (S. Zhang et al., 2017), biodiesel (6.91%) (Zhang et al., 2016), metal-organic frameworks (4.5%) (Ho and Fu, 2016), or industrial wastewater treatments (9.90%) (Mao et al., 2020). This indicates that related researchers in the field of COFs began to summarize and draw conclusions based on the current research status and promoted the development and progress in the field. This also implies that more articles in the future will likely focus on research in the field of COFs.

The numbers of the nine research directions were also studied (Fig. 2b). Chemistry multidisciplinary, materials science multidisciplinary and nanoscience nanotechnology were the top three fields of COFs. This was because COFs, as a new type of porous crystalline material, have attracted wide attention in the direction of chemistry, materials, and nanoscience. Additionally, the research direction of COFs gradually shifts to the application direction including the typical research directions of environmental science, engineering environmental, green sustainable science technology, and water resource. This indicates that COFs occupies an important position in the environmental application.

The numbers of documents every year (green histogram) and total numbers of documents accumulated year by year (red line) over time are summarized (Fig. 2c). The number of publications in COFs area has progressively increased each year, with the number of articles published increasing from 1 in 2005 to 592 on October 06, 2020. Before 2011, a total of 124 documents were published, showing that the initial stage of development in the COFs field was relatively slow because the structural characteristics, preparation methods, and application areas of COFs were still unclear, requiring some time to explore. It is worth mentioning that pronounced growth was observed from 2016 to 2018 compared to the previous years. A total of 1160 documents were published in the last two years, accounting for 44.80% of the total number. These results indicate that an increasing number of researchers are dedicated to the field of COFs.

3.2. Active countries, institutions, and authors

Understanding the number of articles published in different countries helps to quickly identify the country that has made significant contributions to the COF area. These papers come from a total of 67 countries. China published the highest number with a value of 1373, accounting for 53.03%, followed by USA (507, 19.58%), Germany (262, 10.12%), India (182, 7.03%), and Japan (160, 6.18%). A visual diagram can indicate the node co-occurrence effectively. A visual co-occurrence analysis of the top 20 countries is shown in Fig. 3. Among the top 20 countries, there were only two developing countries, namely China and India. This explains why, except for China and India, research in developing countries is relatively lagging behind when compared to the trend in developed countries. Thus, developing countries should strengthen research in the field of COFs and collaborate with researchers in developed countries to improve their respective national scientific research capabilities. It was observed that China, USA, Germany, Spain, and Sweden play an important role in the research on COFs.

The network of co-occurrence can not only identify the number of documents published by institutions, but also help researchers find

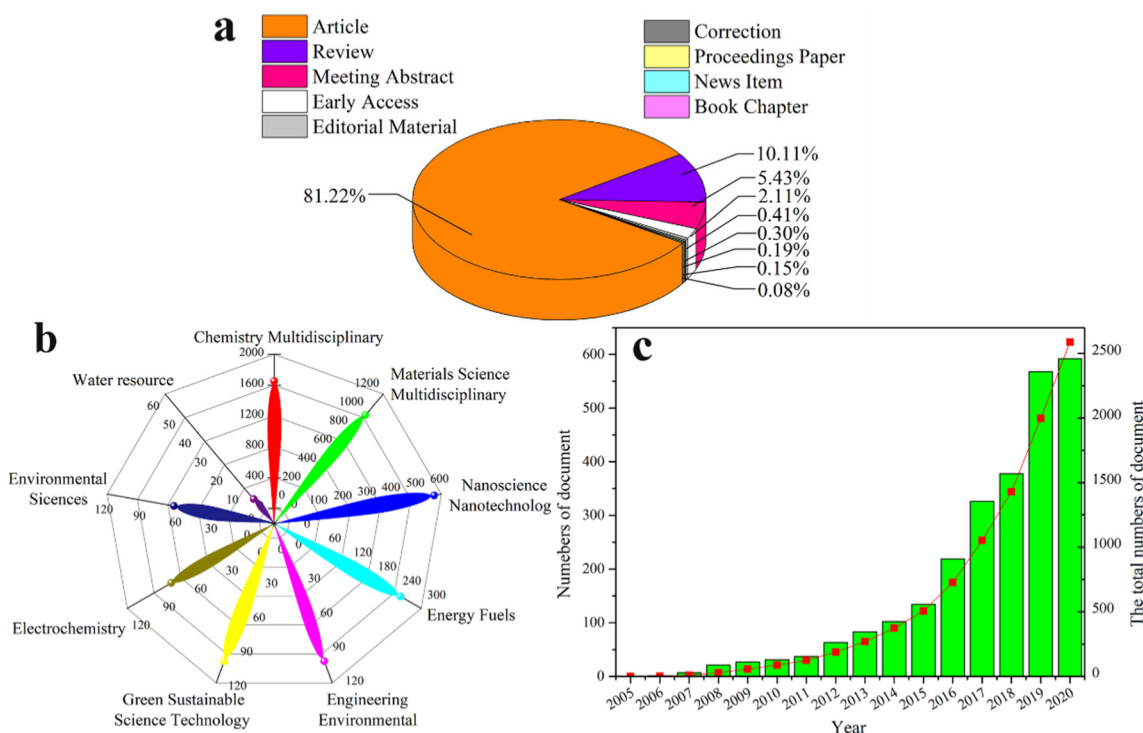


Fig. 2. Proportion of document types on the field of COFs (a); Distribution of research directions (b) and document numbers (c) during the years 2005–2020.

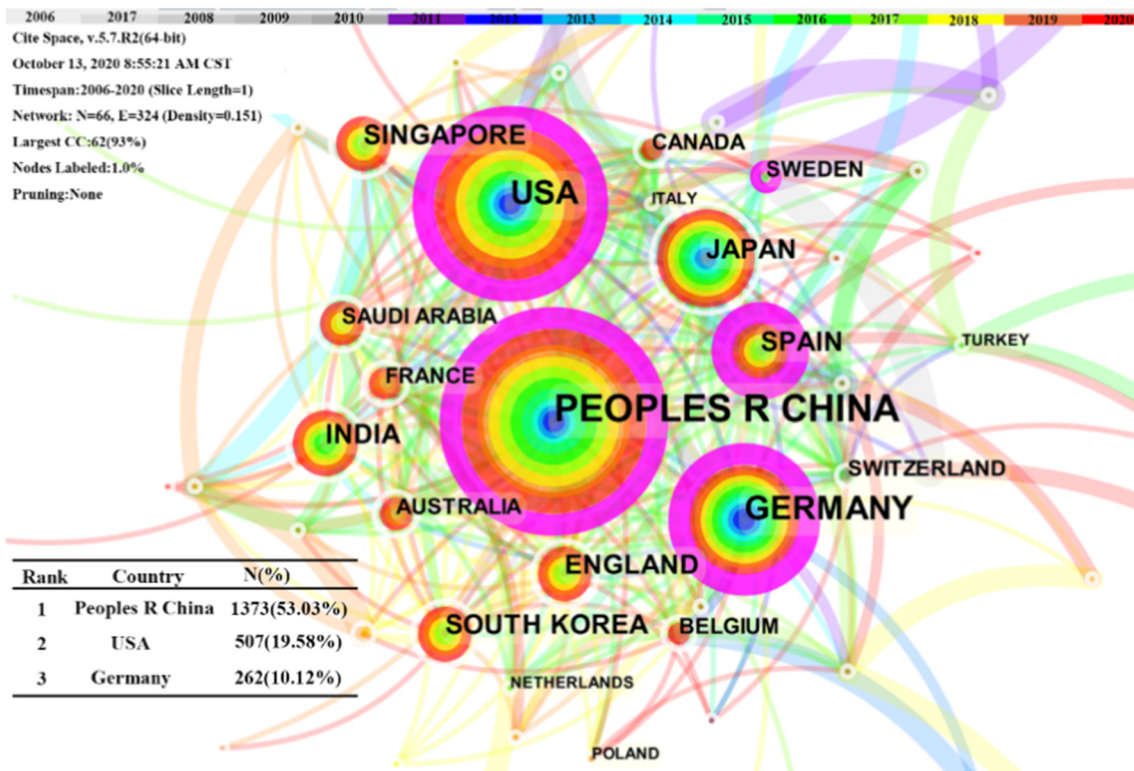


Fig. 3. Visual co-occurrence analysis of top 20 conductive countries.

important institutions and potential collaborators. A total of 403 institutions were found and the top 20 conductive institutions were visualized (Fig. 4). Collaboration between institutions is displayed through the link

of institutions, where the thicker the connection line, the closer the collaboration of institutions. For instance, China has close ties with 98 institutions. Among them, the Chinese Academy of Sciences ranked first

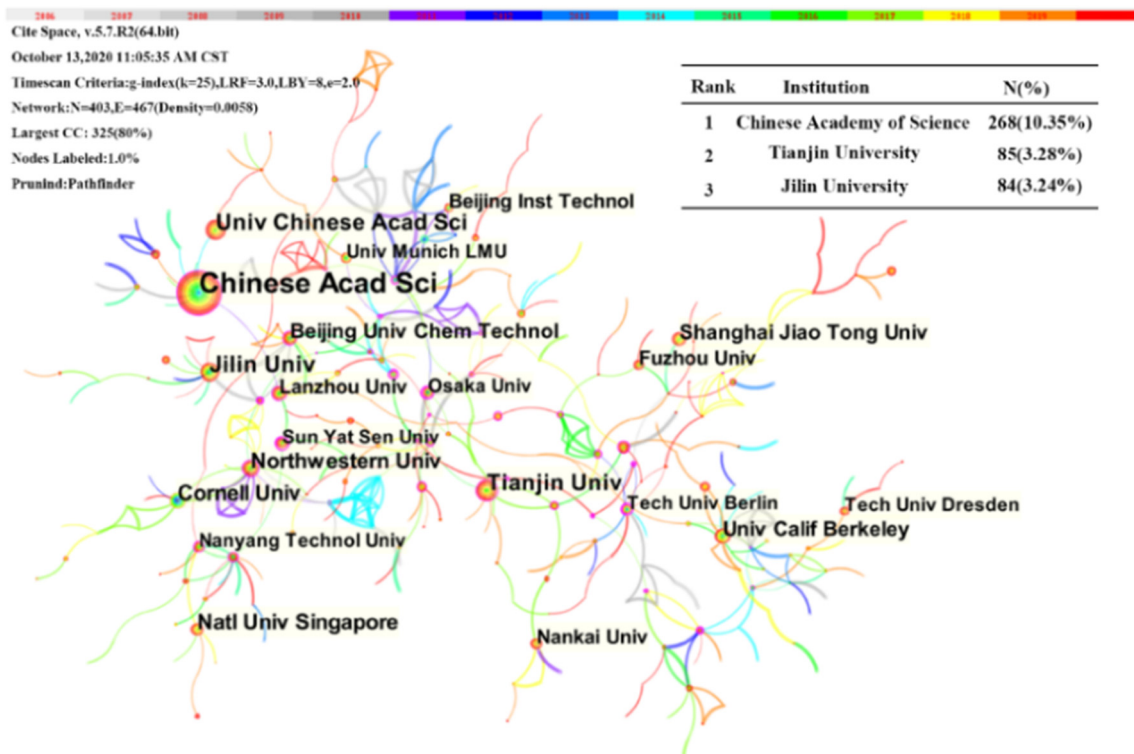


Fig. 4. Visual co-occurrence analysis of top 20 conductive institutions.

Table 1
The top 10 active authors and co-cited authors.

Rank	Author	Country	N (%)	Co-cited author	Country	Citations
1	Donglin Jiang	Singapore	70 (2.70%)	Adrien P. Cote	USA	1150
2	William R. Dichtel	USA	54 (2.08%)	Sanyuan Ding	China	1039
3	Rahul Banerjee	India	46 (1.78%)	Xiao Feng	China	722
4	Omar M Yaghi	USA	33 (1.27%)	Sharath Kandambeth	Saudi Arabia	606
5	Xiao Feng	China	32 (1.24%)	Ning Huang	China	599
6	Arne Thomas	Germany	30 (1.16%)	Hiroyasu Furukawa	USA	552
7	Yang Li	China	29 (1.12%)	Pierre Kuhn	Germany	536
8	Thomas Bein	Germany	29 (1.12%)	Sun Wan	China	504
9	Shengqian Ma	China	26 (1.00%)	Hong Xu	China	486
10	Long Chen	China	25 (0.97%)	John W. Colson	USA	476

with 268 articles, accounting for 10.35%, followed by Tianjin University (85, 3.28%) and Jilin University (84, 3.24%). Furthermore, 11 institutions were from China, and they accounted for more than half of the top 20 institutions. The other nine institutions were from USA (three institutions), Germany (three institutions), Singapore (two institutions), and Japan (one institution).

A total of 589 authors were identified in the field of COFs. The top 10 conductive authors and co-cited authors are summarized according to numbers and co-cited frequencies, respectively (Table 1). Donglin Jiang was the author with the largest numbers of articles (70), followed by William R Dichtel (54), Rahul Banerjee (46), Omar M Yaghi (33), and Xiao Feng (32). Adrien P. Cote has higher co-cited frequency of 1150. Sanyuan Ding and Xiao Feng also have higher co-cited frequencies ranked second and third, respectively. As shown in Table 1, highly co-cited authors and high-yield authors mainly came from three countries, namely, China, USA, and Germany, which were consistent with the results of the country-wise analysis. Furthermore, Rahul Banerjee, one of high conductive authors, and Sharath Kandambeth, one of highly co-cited authors, were both from developing countries (i.e., India and Saudi Arabia, respectively).

3.3. Journals distribution, co-cited journals and co-cited references

The sources of COFs articles from 2005 to 2020 can be deconvoluted into 176 journals through the data exported from the WOS. The top 10 journals are summarized in Table 2. Citation/N indicates the average number of citations per document. The *Journal of the American Chemical Society* ranked first with 252 documents, accounting for 9.73% of all documents. Simultaneously, it holds the highest average number of

citations per document; that is, every document in this journal was cited an average of 99.90 times, indicating the importance of this journal. Moreover, half of the journals originate from the USA, followed by England and Germany with values of 3 and 2, respectively. The *Chemistry of Materials* from the USA had the lowest document numbers with 55 articles.

The top 10 co-cited journals are also listed (Table 2). The *Journal of the American Chemical Society*, with the co-cited frequency of 2346, ranked first and its impact was 14.612. The papers published in this journal received the most attention from researchers and enjoyed a high reputation in the field of COFs. The 10 journals were from three countries, namely, USA, England, and Germany. Accordingly, their significant contributions can be inextricably linked to the successive evolution of COFs. Additionally, the co-cited frequencies of *Angewandte Chemie International Edition* and *Science*, two top journals, were ranked second and third, respectively. The high-impact review journals of *Chemical Society Reviews* (42.846) and *Chemical Reviews* (52.758) were also ranked among the top ten most co-cited frequencies, which shows that the review articles published in these journals are more popular in the field of COFs.

The top 10 co-cited references are listed in Table 3. The study published in 2005 by Yaghi et al. received the highest co-citation, which was the first article to depict COFs (Cote et al., 2005). Seven documents were cited more than 400 times. In the top 10 co-cited references, four reviews systematically summarized the design, synthesis, and application of COFs, indicating that material research not only focused on the invention of new materials, but also paid more attention to the application of materials. All these highly co-cited references were published in influential and authoritative journals, such as *Science*, *Chemical Society*

Table 2
The top 10 journals and co-cited journals.

Rank	Journals	N (%)	Citation/N (average citation)	Country	IF (2019)	Co-cited Journal	Co-citations	Country	IF (2019)
1	Journal of the American Chemical Society	252 (9.73%)	99.90	USA	14.612	Journal of the American Chemical Society	2346	USA	14.612
2	Chemical Communications	175 (6.76%)	41.85	England	5.996	Angewandte Chemie International Edition	2190	Germany	12.959
3	Journal of Materials Chemistry A	159 (6.14%)	27.02	England	11.301	Science	1930	USA	41.845
4	Angewandte Chemie International Edition	156 (6.02%)	77.98	Germany	12.959	Chemical Communications	1922	England	5.996
5	ACS Applied Materials Interfaces	149 (5.75%)	23.63	USA	8.758	Chemical Society Reviews	1778	England	42.846
6	Chemistry A European Journal	107 (4.13%)	25.31	Germany	4.857	Advanced Materials	1596	USA	27.398
7	Journal of Physical Chemistry C	67 (2.59%)	21.54	USA	4.189	Chemistry of Materials	1476	USA	9.567
8	RSC Advances	66 (2.55%)	11.68	England	3.119	Nature Communications	1413	England	12.121
9	Advanced Materials	60 (2.32%)	79.83	USA	27.398	Journal of Materials Chemistry A	1294	England	11.301
10	Chemistry of materials	55 (2.12%)	25.27	USA	9.567	Chemical Reviews	1246	USA	52.758

Table 3
The top 10 co-cited references.

Rank	Co-cited references	Type	Journal	Year	Co-citation	DOI	Reference
1	Porous, Crystalline, Covalent Organic Frameworks	Reports	Science	2005	1135	10.1126/science.1120411	(Cote et al., 2005)
2	Covalent organic frameworks (COFs): from design to applications	Review	Chemical Society Reviews	2013	688	10.1039/c2cs35072f	(Ding and Wang, 2013)
3	Covalent organic frameworks	Review	Chemical Society Reviews	2012	632	10.1039/c2cs35157a	(Feng et al., 2012)
4	Construction of Covalent Organic Framework for Catalysis: Pd/COF-LZU1 in Suzuki-Miyaura Coupling Reaction	Article	Journal of the American Chemical Society	2011	563	10.1021/ja206846p	(Ding et al., 2011)
5	Porous, Covalent Triazine-Based Frameworks Prepared by Ionothermal Synthesis	Communications	Angewandte Chemie International Edition	2008	521	10.1002/anie.200705710	(Kuhn et al., 2008)
6	Designed Synthesis of 3D Covalent Organic Frameworks	Reports	Science	2007	461	10.1126/science.1139915	(El-Kaderi et al., 2007)
7	Construction of Crystalline 2D Covalent Organic Frameworks with Remarkable Chemical (Acid/Base) Stability via a Combined Reversible and Irreversible Route	Communications	Journal of the American Chemical Society	2012	453	10.1021/ja308278w	(Kandambeth et al., 2012)
8	The atom, the molecule, and the covalent organic framework	Review	Science	2017	375	10.1126/science.aal1585	(Diercks and Yaghi, 2017)
9	A Crystalline Imine-Linked 3-D Porous Covalent Organic Framework	Communications	Journal of the American Chemical Society	2009	373	10.1021/ja8096256	(Uribe-Romo et al., 2009)
10	Covalent organic frameworks: a materials platform for structural and functional designs	Review	Nature Reviews Materials	2016	344	10.1038/natrevmats.2016.68	(Huang et al., 2016)

Reviews, *Journal of the American Chemical Society*, and *Angewandte Chemie International Edition*.

3.4. Keywords co-occurrence and clusters

The visualization of mainly keywords co-occurrence and the top 10 keywords of frequency are summarized and shown in Fig. 5. The node color from gray to red represents the years from 2006 to 2020. The highest frequency belonged to the “covalent organic framework”

(488) and “covalent triazine framework” (62), which confirmed that these two terms were the search subject words in the field. It was easy to find that the keywords focused on “adsorption,” “photocatalysis,” “heterogeneous catalysis,” and “supercapacitor” have high frequencies and represent the application of COF materials. In addition, other keywords related to applications were displayed, which included “hydrogen evolution,” “electrocatalysis,” “catalysis,” “CO₂ capture,” “electrochemical sensor,” “energy storage,” “lithium-sulfur battery,” and “lithium-ion battery.” In terms of structure design, the

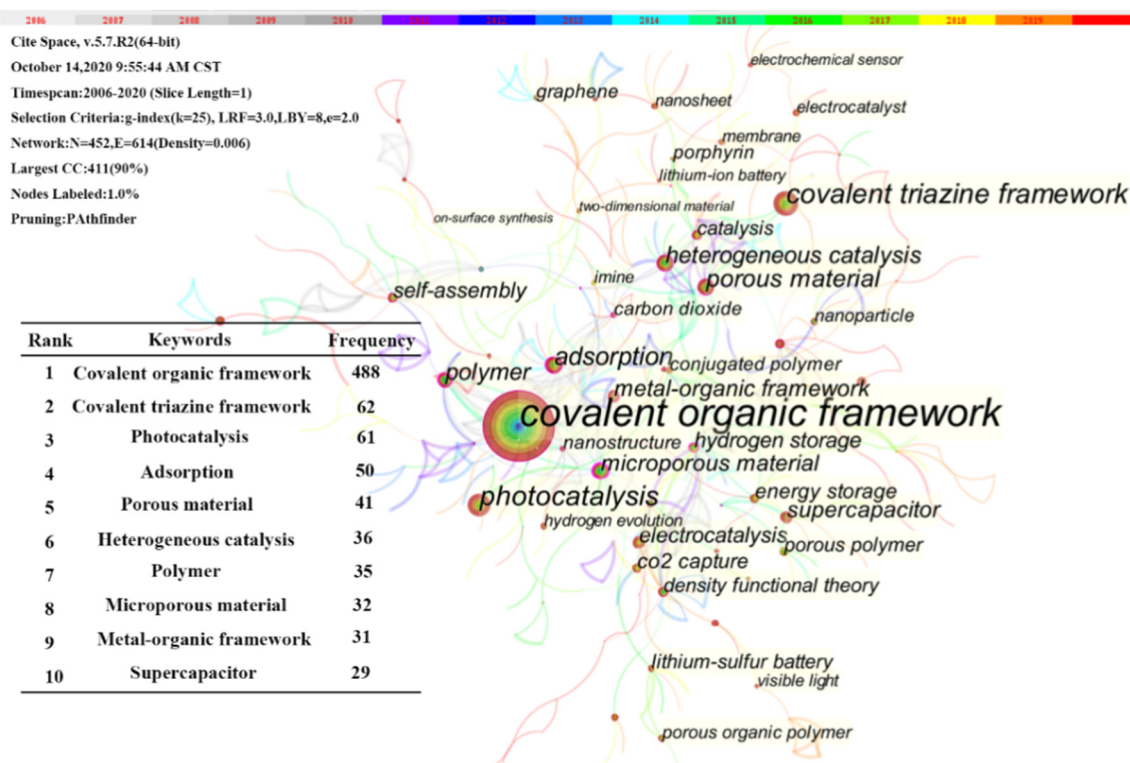


Fig. 5. Visual analysis of keywords co-occurrence.

shape of COFs changes continuously with the depth of research, including nanoparticles, sheets, nanosheets, and membranes. Some words that indicated the characteristics and preparation methods of COF materials were “porous material,” “microporous material,” “self-assembly,” and “on-surface synthesis.” Notably, the keywords of “Metal-organic framework,” “graphene,” and “polymer” hold a higher frequency, demonstrating that COFs hybridize with inorganic and organic materials to form inorganic- inorganic and inorganic-organic hybrid materials, respectively.

To better understand the research hotspots, the keywords of COFs field were divided into four clusters by the VOSviewer software (Fig.6). Cluster 1 focused on photocatalysts and electrocatalysts based on COFs, mainly for CO₂ reduction, O₂ reduction, and hydrogenation. Cluster 2 studied the related research of COFs as adsorbents and sensors, which were expressed through “adsorption,” “sensor,” “low detection limit,” “extraction,” and “removal.” Cluster 3 introduced some reports on COFs membrane, which was the most advanced platform for various environmental and energy-related separation/transport processes. The keywords with the highest frequency in Cluster 4 were “energy storage,” “Li-ion battery,” “lithium-ion battery,” and “supercapacitor,” indicating that the use of COFs for energy storage was also a hot research topic. Based on the above analysis results, the research hotspots in the field of COFs are discussed in depth in Part 4.

4. Research hotspot analysis

4.1. Linkages, topology, and synthesis methods of COFs

4.1.1. Linkages and topology of COFs

Based on the reticular chemistry principle, COFs are porous organic frameworks with different topologies formed by the

connection of covalent bonds via various kinds of linkages. COFs can be designed and functionalized because of the diversity of their building blocks and topological shape. Thus, it is necessary to understand its types of building blocks and topologies. A variety of topology structures by reticular synthesis were formed and listed in Fig. 7. In addition, new topologies of 3D COFs have been developed, such as tfj, cda, pcu, acs, bbt and so on ((Bureekaew and Schmid, 2013); Lan et al., 2018).

The building blocks of the COFs are connected by covalent bonds. Typical covalent bonds are assigned to boronate, triazine, imine, and hydrazine bonds, and the performance of the COFs varies with the type of linkage. For instance, boronate-linked COFs are unstable and easily hydrolyzed in water. The triazine bond has abundant nitrogen atoms and a triazine ring, which could be used as a photocatalytic active site and is beneficial for improving the photocatalytic performance of COFs. In addition to traditional covalent bonds, other types of building blocks (imidazole, sp² carbon-carbon, aryl ether, vinylene, porphyrin, and thiophene) are used to synthesize periodic COFs, which greatly broadened the types of COFs. Different types of building blocks are summarized and displayed in Fig. 8 (H. Wang et al., 2020). According to the type of covalent bond, the reactions used to synthesize COFs can be divided into three types of typical reactions, including the condensation reaction of boric acid, the polycondensation of triazine, formation reaction of imine bond, and the Schiff-base reaction. Schiff-base COFs, as a class of COFs, can be divided into imine- and hydrazine- bond COFs according to the diversity of monomers in the reaction.

4.1.2. Synthesis method of COFs

The methods for preparing COFs were divided into solvothermal method, ion-thermal method, microwave assisted method, mechanochemical method, and other methods. The advantages and

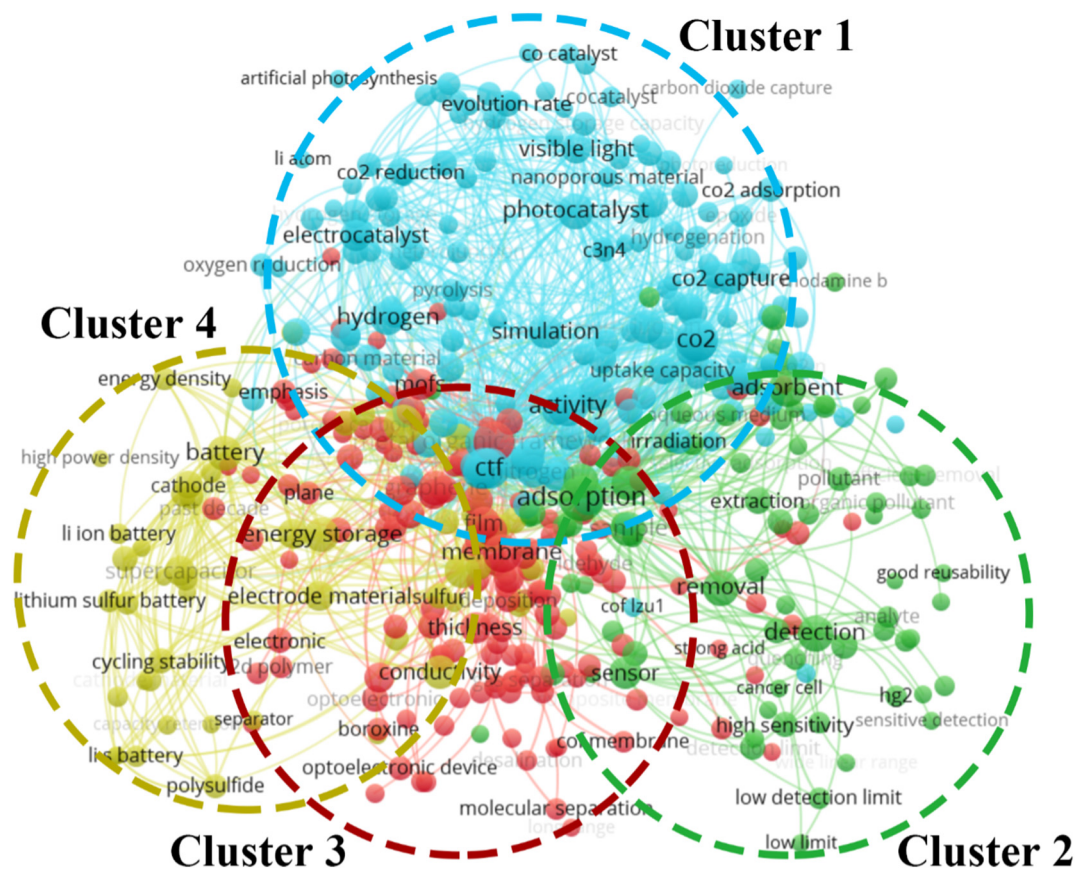


Fig. 6. Visual analysis of keywords co-occurrence. Cluster 1: COFs as catalysts including photocatalyst and electrocatalysts; Cluster 2: COFs as adsorbents and sensors; Cluster 3: COFs membrane; and Cluster 4: Application of COFs in energy storage.

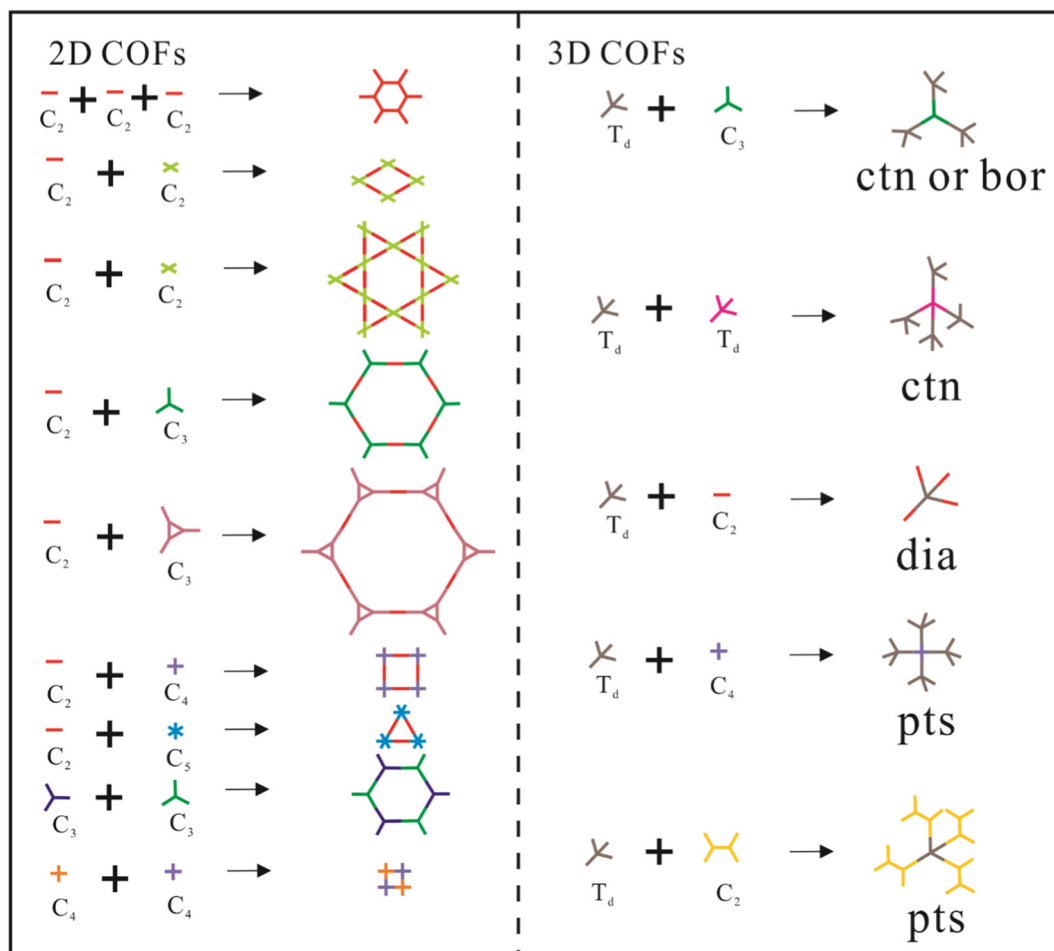


Fig. 7. Topology formation of 2D COFs and 3D COFs.

disadvantages of four typical synthesis methods were summarized in a table (Table 4).

Other methods such as the interface synthesis method (e.g., synthesis on metal (Faury et al., 2012) or pyrolytic graphite surfaces (Xu et al., 2013)) and synthesis under ambient conditions (Medina et al., 2015; Y. Liu et al., 2019) have also been developed to form a variety of multifunctional COFs.

4.2. COFs act as adsorbents

Superior porosity and specific surface area endow COFs with excellent adsorption removal capacity for hazardous pollutants, making COFs as promising application materials. In this field, COFs are supposed to be excellent adsorbents for adsorption removal of heavy metals, organic pollutants as well as for gas adsorption. The main adsorption mechanism can be divided into six types: 1) hydrogen bonding, 2) electrostatic interaction, 3) π - π interaction, 4) size-sieving interaction, 5) hydrophobic interaction, and 6) affinity interaction. The diagram of the adsorption mechanism is displayed in Fig. 9.

4.2.1. Removal of heavy metals

Heavy metal pollution has caused serious hazards to human health and the ecological environment due to its high toxicity and non-degradability. Thus, it is imperative to develop an effective adsorbent for heavy metal removal.

On account of the remarkable affinity between sulfur-containing groups and Hg^{2+} , the elaborate design and synthesis of COFs containing

sulfur groups can achieve the purpose of removing Hg^{2+} . COFs (TAPB-BMTTPA-COF) were synthesized by the co-condensation of sulfur-containing monomers (Huang et al., 2017). More than 99% of the Hg^{2+} was removed in a short time (5 min) and the maximum uptake capability of 734 mg/g by TAPB-BMTTPA-COF was obtained. In addition, 92% Hg^{2+} could be removed by COF after six cycles, which was much superior to other porous materials, such as Cr-MIL-101s, mesoporous silicas, and porous carbons.

A brilliant uptake capacity (1350 mg/g) for Hg^{2+} was realized by another COF owing to the synergy of rapid diffusion and chelation (Sun et al., 2017). COFs with ethynyl, triazole, and thiol groups can achieve an extraordinary adsorption capacity (4395 mg/g) for Hg^{2+} , wherein 98.23% of the Hg^{2+} was removed within 2.50 min (Meri-Bofi et al., 2017). In addition, magnetic nanoparticle-modified COFs (Fe_3O_4 /M-COFs) were applied to adsorb Hg^{2+} ; these not only possessed a high adsorption capacity with a value of 97.65 mg/g, but were not interfered with by other cations (e.g., Na^+ , Zn^{2+} , Mg^{2+} , Ni^{2+} , Pb^{2+} , Cd^{2+} , or Cr^{3+}) in the solution; this led to a high selectivity for Hg^{2+} owing to the complexation interaction between amino groups and Hg^{2+} (Ge et al., 2016). The removal Hg^{2+} efficiency of Fe_3O_4 /M-COFs maintained 76.82% after five cycles.

Ionic COFs were used to remove Pb^{2+} and had a lower uptake capability of 12.68 mg/g (Gupta et al., 2018). To improve the adsorption capacity of Pb^{2+} , two diamine-based COFs (COF-TP, COF-TE) were constructed and showed high uptake capacities of 185.7 and 140 mg/g, respectively (G. Li et al., 2019). Both COFs could remain 95% adsorption capacity for Pb^{2+} after ten cycles indicating the great stability of

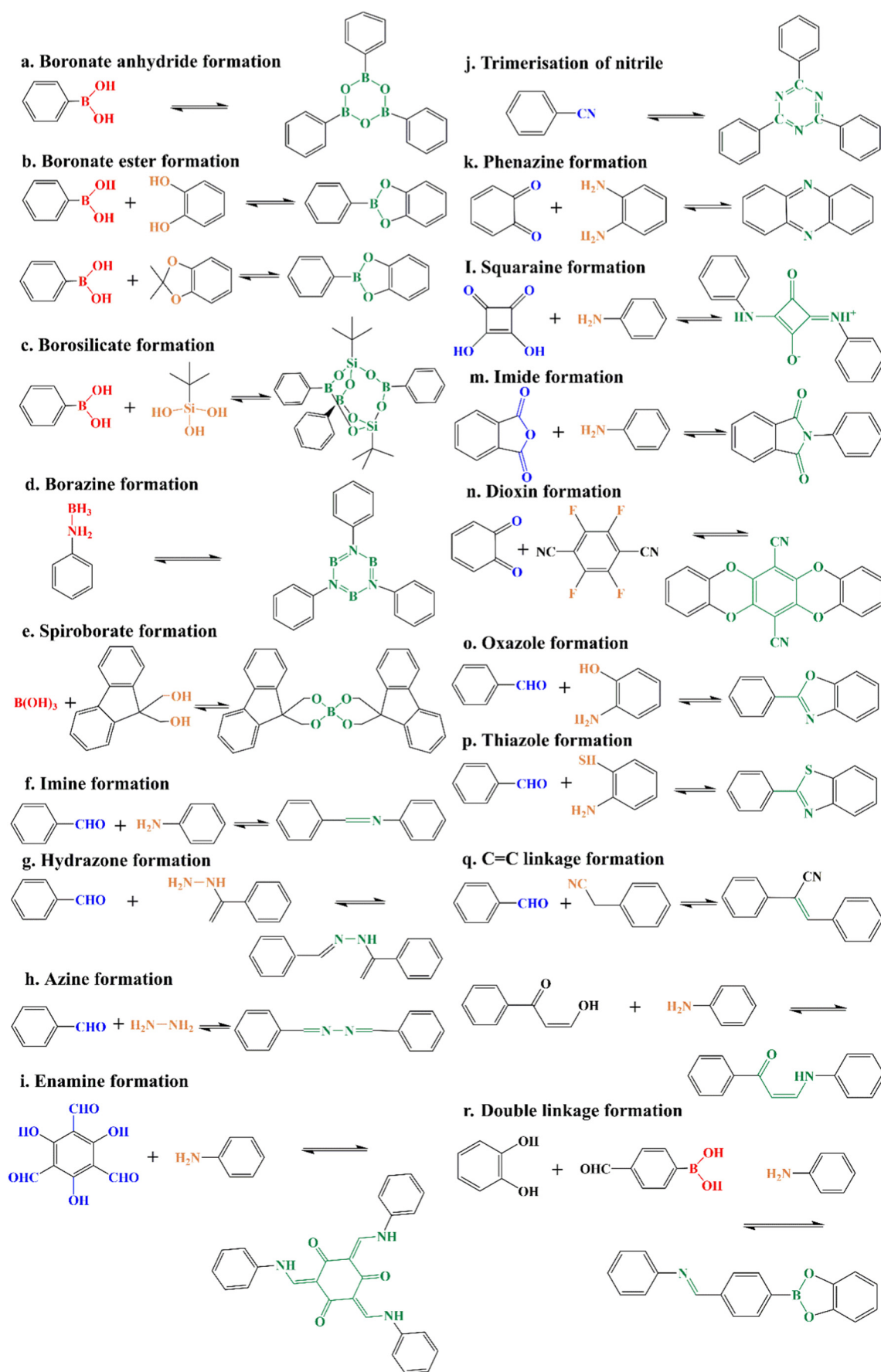


Fig. 8. Various kinds of COFs linkages (H. Wang et al., 2020).

Table 4
The advantages and disadvantages of typical methods.

Method	Advantages	Disadvantages	Reference
Solvothermal method	High crystallinity; uniform particle size; few defects	High temperature (85–120 °C); Long time (2–9 days); Large consumption of organic solvents; High pressure	(Huang et al., 2013; W. Liu et al., 2019; Ding and Wang, 2013; Li et al., 2018)
Ion-thermal method	Mostly CTFs; Large specific surface area; High electron-hole mobility; Potential of large-scale production	High temperature (400 °C); the narrowed range of monomer types of COFs; amorphous porous structure; low crystallinity	(Niu et al., 2020)
Microwave assisted method	Fast energy transfer; Easy control; Short time; High yield	High energy consumption	(Zhang et al., 2012; Ji et al., 2020)
Mechanochemical method	Less time consumption; Less or no organic solvents; Simple operation; Potential of large-scale production	Low crystallinity, porosity, and specific surface area	((Biswal et al., 2013); Das et al., 2014; X. Wang et al., 2018)

adsorption performance of the amide-based COFs materials. The first research on the removal of Cd^{2+} by CTF-1 via adsorption based on electrostatic interaction has been reported (Ghazi et al., 2018). The improvement of the affinity between COFs and Cu^{2+} by introducing hydrogen bonds into COFs, leading to a high adsorption capacity has also been confirmed (Y. Li et al., 2019a). The distribution of surface groups on COFs is closely related to the adsorption capacity of heavy metals. For example, two COFs (COF-1 and COF-2) with different hydroxyl group distributions were formed (Fig. 10). The maximum adsorption capacities for Cr (VI) were 462.96 (COF-1) and 649.35 (COF-2) mg/g, respectively (Zhu et al., 2020).

4.2.2. Removal of organic pollutants

At present, various COFs were developed to adsorb organic pollutants from water such as dyes, endocrine disrupting chemicals (EDCs), pharmaceuticals, pharmaceuticals and personal care products (PPCPs), flame retardant, biotoxin, crude oil, and organic solvents, and so on.

The adsorption of dyes was also studied. Triazine-functionalized polyimide COFs were synthesized, exhibiting great potential for the removal of three kinds of organic dyes (X. Zhu et al., 2017). This COF achieved a high adsorption capacity (1691 mg/g) for methylene blue with a small molecule, which exceeded the performance of the majority of nano-porous adsorbents. The outstanding adsorption capacity originated from the contribution of pore-size effect.

The efficient removal of EDCs via adsorption of COFs was also investigated. Typical core-shell Fe_3O_4 @COF nanospheres reasonably designed and synthesized showed faster adsorption within 5 min and achieved maximum uptake for bisphenol A (160.6 mg/g) and bisphenol AF (236.7 mg/g) (Y. Li et al., 2017). In the adsorption process, π - π interactions and hydrogen bonding were considered to be the main adsorption mechanisms. Another magnetic COFs were used for the adsorption of lipophilic chlorpyrifos and atrazine and obtained maximum adsorption capacities of 270 and 54 mg/g, respectively (Romero et al., 2020). In this process, van der Waals interactions played an important role. Furthermore, the desorption efficiency decreased only ~4% at the end of five cycles indicating the magnetic COFs was recyclable.

The adsorption of pharmaceutical pollutants such as painkillers and antibiotics on COFs was also explored. The β -agonists and fluoroquinolones in milk and pork meat samples were efficiently adsorbed and identified by magnetic sulfonated COFs composites (Fe_3O_4 @ SiO_2 - NH_2 /1,3,5-triformylphloroglucinol/2,5-diaminobenzene-sulfonic acid (200:105:141, m/m)), and the adsorption mechanism was attributed to the synergistic effect of π - π stacking, hydrophobicity, electrostatic attraction, and hydrogen-bonding interactions (Hu et al., 2021). COFs also showed great adsorption and satisfactory reusability for quinolone antibiotics (G. Xu et al., 2020).

PPCPs were determined through the adsorption of COFs. Li et al. synthesized magnetic COFs (Fe elements = 59.62 wt%) for the removal of triclosan and triclocarban (Y. Li et al., 2019b). Results showed that high adsorption capacity and fast adsorption were achieved based on

the space embedding effect, van der Waals forces, and π - π stacking interactions.

It is necessary to remove flame retardants from water through adsorption methods. Three COFs with different pore sizes of 1.81, 2.57, and 3.34 nm were synthesized and assigned to COF-1, COF-2, and COF-3, respectively (W. Wang et al., 2018). The maximum uptakes for aryl-organophosphorus flame retardant (triphenyl phosphate) were 86.1 (COF-1), 387.2 (COF-2), and 371.2 mg/g (COF-3), respectively, which indicated that proper pores designed according to the size of pollutant molecules were conducive to the efficient adsorption of pollutants.

Recently, the adsorption behavior of biotoxins on COFs was studied. Magnetic COFs with crystalline and superparamagnetic features were formed (Romero et al., 2019). Okadaic acid and dinophysistoxin-1 were adsorbed for the first time by this magnetic COF, achieving maximum uptake values of 812 mg/g and 830 mg/g; these values exceed those of non-magnetic macro-porous resins by 500-fold and 300-fold, respectively.

4.2.3. Adsorption of gas

In 2009, the first adsorption studies of H_2 , CH_4 , and CO_2 in COFs was reported by the Yaghi et al. Three kinds of COFs, including 2D COFs with 2D structures and 1D small pores (COF-1 and COF-6), 2D COFs with 2D structures and 1D large pores (COF-5, COF-8, and COF-10), and 3D structures (COF-102 and COF-103), were synthesized and applied to capture gas molecules (Furukawa and Yaghi, 2009). These results indicated that 3D COFs had the largest adsorption capacity at 35 bar (i.e., 72 mg/g at 77 K for H_2 , 187 mg/g at 298 K for CH_4 , and 1180 mg/g at 298 K for CO_2). In addition, the gas absorption capacity of COF-5, COF-8, and COF-10 was higher than that of COF-1 and COF-6. This phenomenon showed that the increase of micropores was the key condition to improve the gas adsorption capacity. Subsequently, many studies have developed different types of COFs for gas capture in air, and the factors affecting the mechanism, including pressure, temperature, surface area, pore volume, and size, were also explored (Olajire, 2017; Chang et al., 2020; Mukhtar et al., 2020; Li et al., 2021). For instance, the uptake capacity of COF-10 for NH_3 was 15 mol/kg at 298 K and 1 bar, which was higher than the values of microporous 13 \times zeolite (9 mol/kg), Amberlyst 15 (11 mol/kg), and mesoporous silica (7.9 mol/kg) (Doonan et al., 2010). In 2017, COFs applied to the removal of SO_2 were reported for the first time (Lee et al., 2017). These COFs with high mesoporosity and large surface areas achieved an adsorption capacity of 6.30 mmol/g (40 wt%) for SO_2 as their porosity allowed SO_2 to enter the internal adsorption site through the channel.

4.3. COFs act as catalysts

COFs catalysts are generally divided into photocatalysts and electrochemical catalysts. The mechanism of pollutant degradation and clean energy generation depends on the type of catalyst used. It is necessary to improve photocatalytic and electrocatalytic performances.

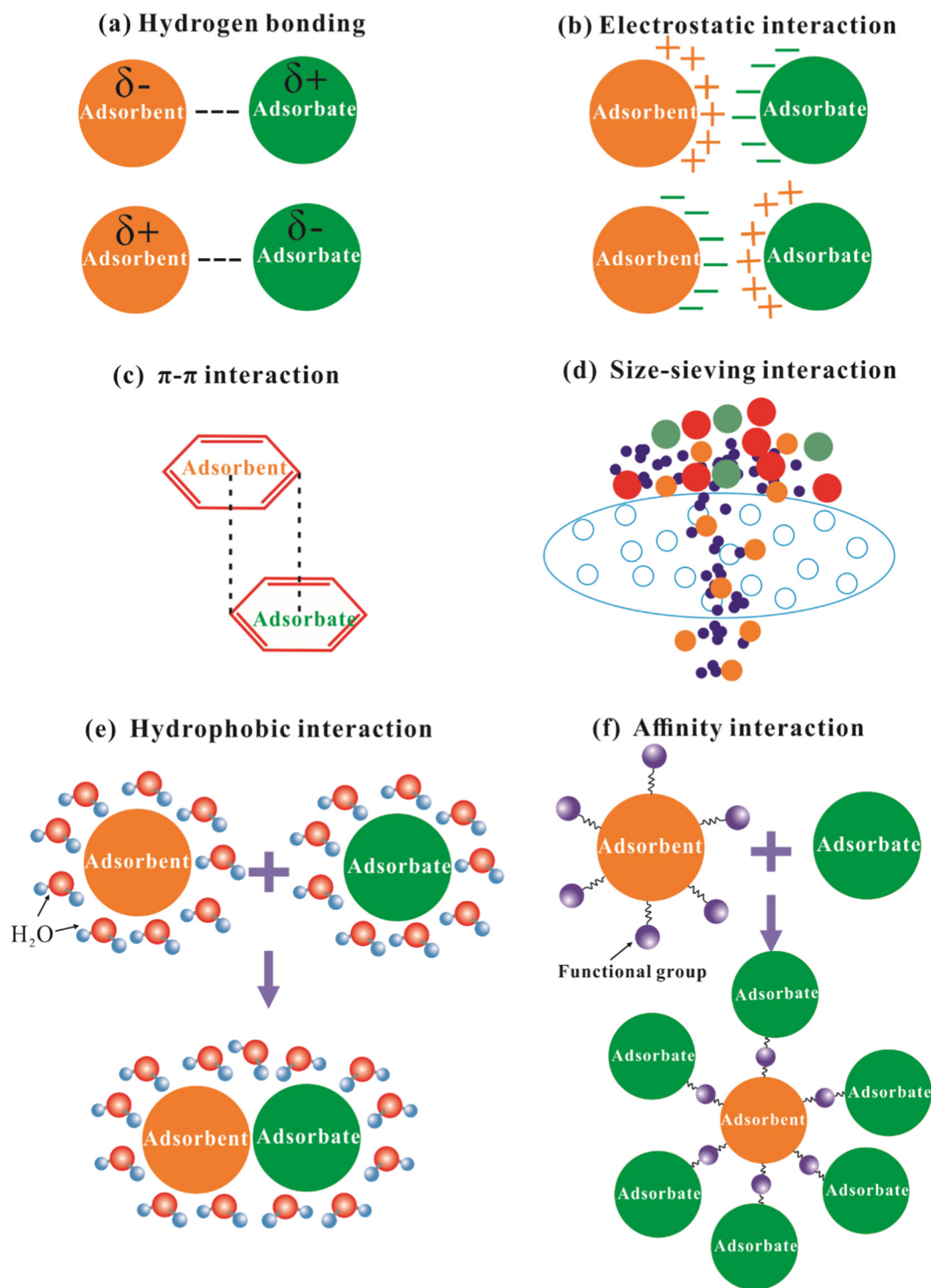


Fig. 9. Diagram of possible adsorption mechanism for removal of pollutants.

4.3.1. Clean energy generation

Photocatalysis is considered a cost-effective and promising method for large-scale clean energy generation, and the significant potential of COFs in this regard has been reported and summarized in journal articles and literature reviews (Ding et al., 2019; Chen et al., 2020; Huang et al., 2020; Q. Yang et al., 2020). The first COF with conjugated A_3B_2 -type poly(azomethine) networks exhibited a steady hydrogen evolution rate (HER) and high chemical and thermal stability (Schwab et al., 2010). The structure of two-dimensional COFs is an important

factor affecting the rate of photocatalytic hydrogen production. Studies have shown that COFs exhibiting the great HER with $750 \pm 25 \mu\text{mol g}^{-1} \text{h}^{-1}$, which benefited from the structure of AA' stacking and promoted exciton migration and charge transport. In addition, the morphology of pre- and post- photocatalysis was not changed due to the chemical robustness of COFs (Ghosh et al., 2020). In the water-splitting process using COFs as catalysts, noble metals are generally used as co-catalysts, which undoubtedly increase industrial costs and limit the actual applications. Banerjee et al. constructed an effective

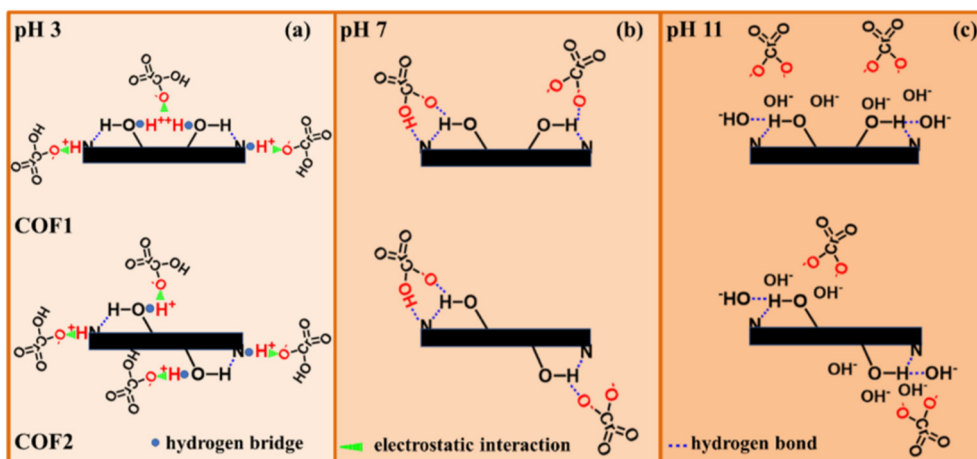


Fig. 10. The proposed Cr (VI) adsorption mechanism on COFs (Zhu et al., 2020).

heterogeneous photocatalytic platform, that is, they used COF as a photosensitizer and cobaloxime as a non-precious metal molecular co-catalyst, and obtained $782 \mu\text{mol g}^{-1} \text{h}^{-1}$ HER. This platform facilitated the transfer of an outer shell electron from the COF to the co-catalyst, causing a monometallic pathway of H_2 generation from the Co^{III} -hydride/ Co^{II} -hydride species (Banerjee et al., 2017). In the presence of electron donors, the photocatalytic reduction of CO_2 into HCOOH catalyzed by mesoporous COFs was determined with the assistance of cobaloximes (Fig. 11a) (Sarkar et al., 2020). Different types of transition metal ions (TMI)-modified COFs can modulate the reduction products of CO_2 . Co-modified COFs and Zn-modified COFs exhibited the highest CO and formic acid production rates of 1.02×10^3 and $152.5 \mu\text{mol h}^{-1} \text{g}^{-1}$, respectively (Fig. 11b) (C. Lu et al., 2019). Furthermore, integrating photocatalytic sites into COFs can effectively

carry out the photocatalytic process without sacrificial agents and co-catalysts.

Electrocatalysis is another effective method for producing clean energy. CTFs with abundant nitrogen atoms exhibited excellent catalytic performance. An electrocatalyst (Nipro-CTF) was prepared and exhibited remarkable selectivity for CO_2 reduction owing to the reduction of the kinetic energy barrier at the active site (C. Lu et al., 2019). The influence of different types of metal loading on the electrocatalytic reduction of CO_2 by CTFs was also explored (Su et al., 2018). Metalloporphyrin-derived 2D COFs were synthesized by regulating the proportion of cobalt/copper porphyrins for the aqueous electrochemical reduction of CO_2 to CO. Research has shown that up to 90% Faradaic efficiency and a high turnover number (TON) of 290,000 are observed at the initial time of 9400 h^{-1} in a neutral solution (Lin

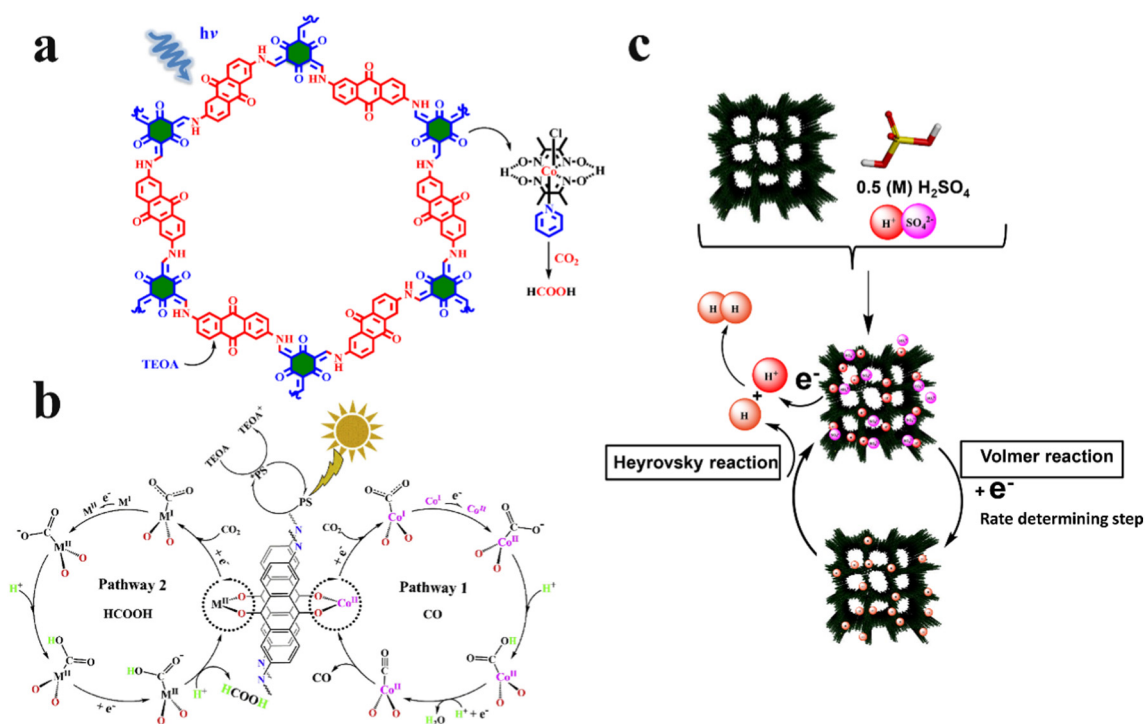


Fig. 11. (a) Proposed reaction cycle for the photocatalytic CO_2 reduction using COF and $\text{Co}-(\text{dmg})_2$ as co-catalyst (Sarkar et al., 2020); (b) Proposed mechanisms for photocatalytic reduction of CO_2 with DQTP COF-M (M. Lu et al., 2019); (c) Possible Volmer–Heyrovsky pathway of hydrogen evolution reaction on SB-PORPy-COF (Bhunia et al., 2017).

et al., 2015). In addition, in the oxygen reduction process, the carbonized COFs used as electrocatalysts showed considerable electrocatalytic activity (Jiao et al., 2017). A theoretical calculation was carried out to determine the oxygen reduction and evolution performance of COF-based electrocatalysts for fuel cells and metal-air batteries. A metal-free hydrogen evolution reaction electrocatalyst based on pyrene-porphyrin was prepared, displaying outstanding charge carrier mobility due to the synergistic effect between the reticular assembly structure and pyrene core (Fig. 11c) (Bhunia et al., 2017). Furthermore, the reasonable design of the porosity of COF induced an improvement in the electrocatalytic activity. For example, macro-COF as electrocatalysis exhibited a Faradaic efficiency of ~98% for the oxygen evolution reaction because the macropores facilitated the substrate entry into the COF pores and reacted with the catalytically active sites (Zhao et al., 2019).

4.3.2. Degradation of organic pollutants

Environmental pollution remains a key problem, thereby restricting the sustainable development of the environment. The photocatalytic process generates redox free radicals and converts mineralized organic pollutants into inorganic substances (e.g., CO₂ and H₂O). To the best of our knowledge, there are very few research reports on COF-based catalysts required for the degradation of organic pollutants, which indicates that it is an area that still needs to be explored and developed by researchers. A novel COFs with a ternary structure of “photoactive centers-photoelectron shift platform-electron withdrawing unit” was constructed using the building blocks of 1,3,5-triformylphloroglucinol and melamine based on reversible Schiff-base reaction and irreversible enol-keto tautomerization (He et al., 2017). Compared to bulk g-C₃N₄, the photocurrent response was 22-fold higher and exhibited a smaller interface resistance and weaker photoluminescence intensity. The COF with such an excellent structure displayed a faster photodegradation rate ($1.02 \times 10^{-1} \text{ min}^{-1}$) than that of bulk g-C₃N₄ ($1.72 \times 10^{-3} \text{ min}^{-1}$) for the degradation of methyl orange; further, it photodegraded 90% of phenol under given conditions as compared to 8% with g-C₃N₄ (Fig. 12a). Semiconductor-modified COFs can enhance the separation of photoinduced carriers, leading to the improvement of photocatalytic activity. A Z-scheme COFs-based heterojunction photocatalyst (COF-PD/AgI) was synthesized via AgI-modified COFs (using 2,5-diaminopyridine and 1,3,5-triformylphloroglucinol as precursors) (F. Liu et al., 2020). Rhodamine B (5 mg/L) can be completely photodegraded by COF-PD/AgI within 80 min under visible light irradiation; the pseudo-first-order kinetic constant was 58.25 times and 2.52 times that of COF-PD and the mixture of COF-PD and AgI, respectively. In addition to the photodegradation of Rhodamine B, COF-PD/AgI also exhibited an effective photocatalytic performance for

the photodegradation of acetaminophen (Fig. 12b). COF-PD/AgI exhibited excellent stability and catalytic activity (>97%) during the four successive reused cycles. Another Z-scheme MOF/COF heterojunction was synthesized and showed outstanding photocatalytic performance for the degradation of methyl orange. A heterojunction photocatalyst (MoS₂/COF) was designed based on MoS₂ modified COFs by a hydrothermal method (Khaing et al., 2020). A total of 98% Rhodamine B and 85.90% tetracycline can be removed effectively by MoS₂/COF under simulated sunlight irradiation for 30 min and 60 min, respectively, and these values are higher than those of pure MoS₂ and COFs. The superior photocatalytic activity was attributed to the construction of the 2D-2D layered structure and the well-matched band position between MoS₂ and COFs. The XRD and degradation efficiency of the catalyst were not obviously changed or decreased after three cycles showing that MoS₂/COF was stable and reusable in the process of photodegradation. The photodegradation of tetracycline hydrochloride and ciprofloxacin was investigated. Three-dimensional COFs and BiOBr nano-flakes (BiOBr/CTF-3D) were compounded via a precipitation method to form composite materials. The photocatalytic activity for 2 wt% of COFs doping in the composite was the greatest due to the expansion of the light absorption range and the improvement of photocarrier separation efficiency. Furthermore, the tetracycline hydrochloride degradation efficiency decreased to around 5.4% compared with the first run after the fourth recycling runs, suggesting the long reusable life of the BiOBr/CTF-3D-2% composite (S.-R. Zhu et al., 2017). Furthermore, morphology control was also used to heighten the photocatalytic performance. COFs (TpMA) were synthesized by co-condensation of 1,3,5-triformylphloroglucinol and melamine via a milling method. The morphology of TpMA varied from the interwoven thread shape to a thin ribbon-like morphology as the volume of solvent increased from 1 mL to 3 mL, that is, TpMA_(1 mL) and TpMA_(3 mL). Further, TpMA_(3 mL) can completely degrade phenol, while only 83.50% of phenol can be degraded by TpMA_(1 mL) under the same conditions (Lv et al., 2019).

4.4. COFs act as membrane treatment

Permeability and selectivity are important indices for evaluating membrane separation performance. Traditional membrane materials such as glassy polymers have high selectivity, while their permeability is reduced and vice versa. Interestingly, COF-based membranes overcome these shortcomings to a certain extent. Periodic and ordered channels of COFs facilitate the transmission of gas and improve pollutant removal efficiency, leading to high permeability and selectivity (Zou and Zhu, 2018). There are two methods for fabricating COF membranes: the top-down approach, such as the assembly of exfoliated COF nanosheets and a bottom-up approach, such as in situ growth and

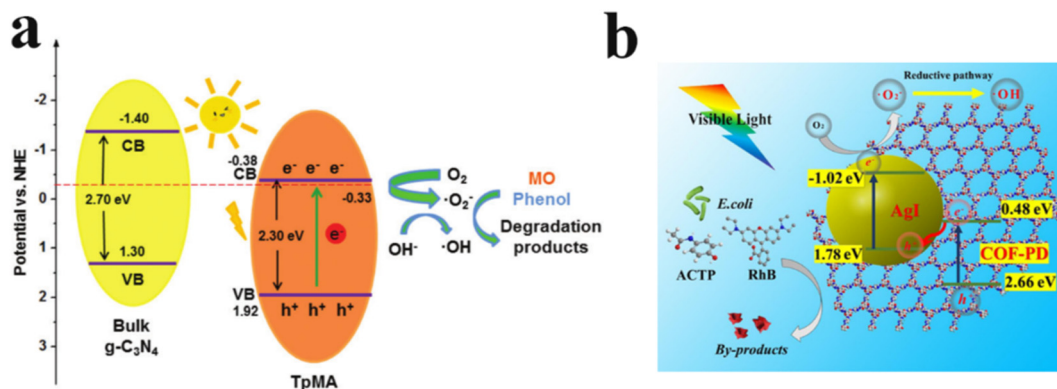


Fig. 12. (a) Schematic of the band gap structures of bulk g-C₃N₄ and TpMA, and the proposed photocatalytic mechanism for the degradation of MO and phenol over TpMA under visible light irradiation (He et al., 2017); (b) the proposed possible mechanism of COF-PD/AgI composite under visible light irradiation (F. Liu et al., 2020).

interfacial polymerization (Kandambeth et al., 2017; G. Li et al., 2017; Fan et al., 2018a; W. Zhang et al., 2018; G. Li et al., 2020).

4.4.1. Water treatment

Membrane treatment technology plays an important role in water treatment because of its simple operation, low energy consumption, high separation efficiency, and no phase change. Various COFs membranes have been developed and applied to water treatment, including molecular separation, oil/water emulsion separation, and desalination.

COFs membranes show great potential for molecular separation. A crystalline two-dimensional COFs membrane (TFP-PDA membrane) with an ultrathin nature of 120 nm was manufactured (Khan et al., 2020). On the one hand, the TFP-PDA membrane showed ultrahigh permeance toward aprotic (acetonitrile and acetone) and protic (water, ethanol, and methanol) solvents, and its performance was dependent on the nature of the solvent. For example, high permeance of water ($411 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) and acetonitrile ($583 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) was achieved because the former could form hydrogen bonds with the membrane, while the latter did not interact with the membrane, leading to a difference in permeance. The TFP-PDA membrane not only showed selectivity to solvents but exhibited a high rejection rate for dye molecules larger than 1.4 nm (99.1% for alcian blue, 98.9% for protoporphyrin IX, 98.9% for Congo red, 98.1% for methyl blue) (Fig. 13a). A COFs-based sandwiched composite membrane was constructed using a perforated polymer-assisted transfer (Xiao et al., 2019). This composite membrane exhibited a high water permeance of $\sim 51 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ and a rejection rate of 98% for dye molecules with a molecular size of $>1.55 \text{ nm}$.

Oil/water emulsion separation is another field of COFs membrane application. Zhang et al. synthesized COF-based composite membranes with a spindle-knotted structure and applied them to oil/water emulsion separation (Zhang et al., 2019). The results showed that COF-based composite membranes possessed superoleophobicity in the presence of water and superhydrophobicity in the presence of oil (Fig. 13b). In addition, they exhibited a high flux ($4229.29 \text{ L/m}^2 \text{ h}$) and separation efficiency ($>99.9\%$).

To improve the efficiency of desalination, hydrophilic carboxylated COF (COF-COOH) were introduced into thin-film composite (TFC)

membranes by interfacial polymerization, causing a thin-film nanocomposite (TFN) membrane (L. Xu et al., 2020). The water flux ($64.2 \text{ L m}^{-2} \text{ h}^{-1}$) and reverse flux selectivity (10.0 L/g) of the TFN membrane (COF-COOH content/TFN membrane solution, 0.5 mg/mL) was much higher than that of the TFC membrane ($15.9 \text{ L m}^{-2} \text{ h}^{-1}$, 2.6 L/g). Therefore, the TFN membrane enhanced the water flux and $\text{H}_2\text{O}/\text{NaCl}$ separation selectivity under given conditions because the introduction of hydrophilic COF-COOH improved hydrophilicity and negative charge properties. Wang et al. reported that a TFN membrane that was modified using COFs improved its water delivery performance ($192.5 \text{ L m}^{-2} \text{ h}^{-1} \text{ MPa}^{-1}$) and high rejection ($>80\%$) to Na_2SO_4 (C. Wang et al., 2017).

4.4.2. Separation of gas

The COFs membrane was expected as a candidate for membrane-based gas separation, owing to its adjustable pore aperture and chemical stability. As such, to date, many studies have reported the research progress and current situation of COFs membranes in gas mixture separation (Wu et al., 2017; Fan et al., 2018c; Das et al., 2020; Liu et al., 2021).

The separation of H_2/CO_2 has been of continued interest to researchers. Fu et al., for the first time, synthesized COF-MOF composite membrane based on the formation of chemical bonds; that is, the COFs are connected with polyaniline by imine groups, and simultaneously, the HN-Zn-imidazole bonds seal the interface between COFs and MOFs. The COF-MOF composite membrane exhibited higher selectivity for H_2/CO_2 mixed gas compared to the individual COF and MOF membranes (Fu et al., 2016). This is because the formation of an interlayer (amorphous MOF layer) enhanced the gas selectivity. A vertically aligned 2D COF (COF-LZU1) membrane was developed through in situ oriented growth inside vertically aligned CoAl-layered double hydroxide (LDH) nanosheets (Fan et al., 2020). This membrane exhibited good H_2 permeance (7200 Barrer at 298 K, 1 bar) and satisfying selectivity for H_2/CO_2 (31.6) and H_2/CH_4 (29.5). The excellent performance was attributed to the precise size-exclusion that occurred in the COF interlayer space (typically 0.3–0.4 nm) (Fig. 13c). COF-COF (i.e., COF-LZU1-ACOF-1) bilayer membranes were fabricated and demonstrated excellent separation selectivity for gaseous mixtures such as H_2/CO_2 , H_2/N_2 ,

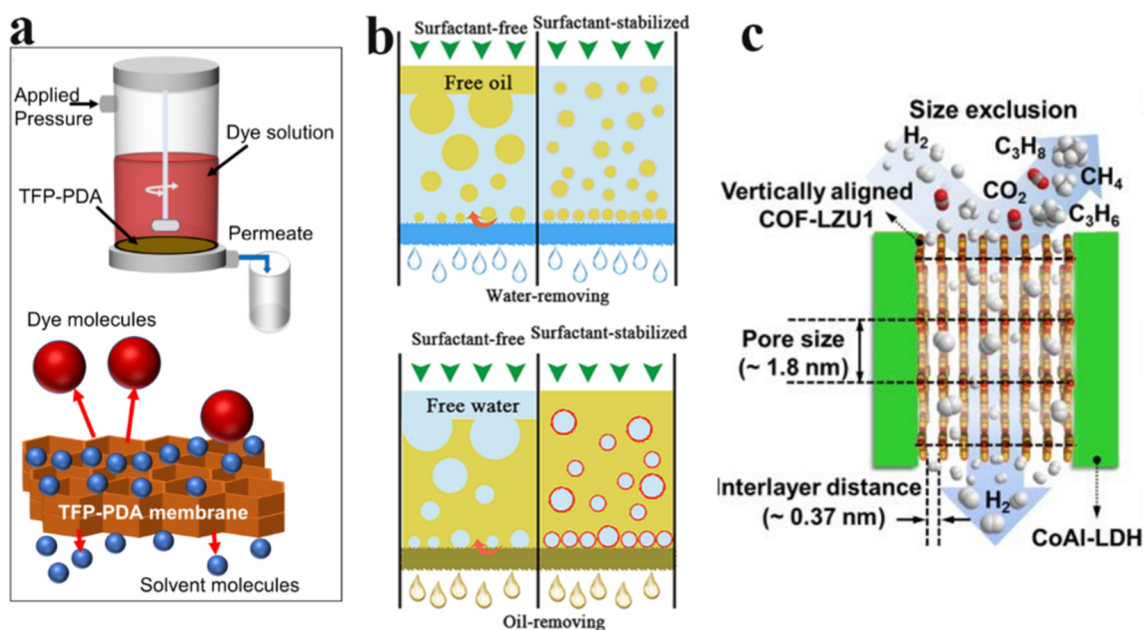


Fig. 13. (a) Schematic diagram of the nanofiltration operation and size exclusion-based separation mechanism (Khan et al., 2020); (b) Elective separation mechanisms for oil/water emulsions and mixtures (Zhang et al., 2019); (c) Schematic illustration of the gas permeation through vertically aligned COF-LZU1 membrane (Fan et al., 2020).

and H₂/CH₄, with values higher than those of monolayer membranes (e.g., COF-LZU1 membrane and ACOF-1 membrane) (Fan et al., 2018b). The key to the improvement of separation efficiency was ascribed to the formation of narrow apertures. The introduction of COFs also improved the interfacial compatibility of the mixed matrix membranes. Jiang et al. for the first time established mixed-matrix membranes based on CTF (Jiang et al., 2019). The mixed matrix membranes with 2 wt% filler loading not only displayed high permeability to CO₂ (7300 Barrer), but also obtained good selectivity (16.6) for a CO₂/CH₄ gas mixture. High permeability and selectivity are due to the affinity of CO₂ with F atoms and triazine and the micro-porosity of the CTF, respectively. Combining COFs with other materials is an effective strategy for making sequential COF-based membranes. The COF/graphene oxide (GO) composite membranes were developed using the vacuum filtration method (Tang et al., 2019). This study showed that H₂ permeance ($1.067 \times 10^{-6} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$) and adequate H₂/CO₂ selectivity (25.57) were obtained for the optimal COFs/GO composite membranes (COFs/GO = 15:1, *m:m*).

4.5. COFs act as sensors

Although traditional methods for detecting pollutants have high selectivity and sensitivity, their widespread application is limited due to the high cost of detection instruments and complex pretreatment. Thus, chemical sensors, especially those prepared with COFs, have attracted wide attention. COFs sensors can detect various pollutants such as heavy metals, antibiotics, EDCs, dynamite, and gas. When a target substance interacts with COFs, it not only causes fluorescence quenching and enhancement but also leads to a change in solution color.

Heavy metals (e.g., Al³⁺, Cu²⁺, Hg²⁺, and Pb²⁺) can be quickly detected using COFs sensors. Considering the affinity between Au and Hg, a reasonable design of sensing materials could achieve the specific detection of Hg²⁺ in water. Cui et al. developed a colorimetric sensor

by anchoring AuNPs onto COFs materials (Cui et al., 2019b). This sensor exhibited a higher detection ability for Hg²⁺ with a detection limit as low as 0.33 nM, which is far below the threshold level of drinking water (10 nM) (Fig. 14b). To effectively detect and remove Hg²⁺, Yu et al. synthesized recyclable fluorescent COFs (AH-COFs) using allyl combined with hydroxy groups as reaction sites and the π -conjugation structure as the signal sensor (Yu et al., 2020). More than 95% Hg²⁺ can be efficiently removed within 2.5 h, proving that the performance of the interaction between Hg²⁺ and allyl cooperated with hydroxy groups is higher than that of the interaction between Hg²⁺ and S atoms. Cui et al. found that the interaction between hydroxyl and imine group-functionalized-COFs and Cu²⁺ could cause fluorescence quenching; fluorescence intensity decreases with increasing Cu²⁺ concentration due to photoinduced electron transfer (C. Cui et al., 2020). To hinder fluorescence quenching caused by photoinduced electron transfer, Cui et al. realized an enhanced fluorescence intensity by using the coordination between COFs nanosheets and Al³⁺ (Fig. 14a) (Cui et al., 2019a). A COF-based electrochemical sensor was applied to detect Pb²⁺ and showed an excellent linear range (0.0050–2.0 $\mu\text{mol/L}$) and a low detection limit (1.9 nmol/L) due to the many active sites and high surface area of the COFs (T. Zhang et al., 2018).

Sensors analysis is a convenient, fast, and effective method for detecting antibiotics in the environment. To achieve sensitive detection of tetracycline, a Zr-coordinated amide porphyrin-based COFs sensor was synthesized, exhibiting a suitable linear range of 5–60 pmol/L with a detection limit of 2.3 pmol/L (Ma et al., 2019). A COF@NH₂-CNT/GCE (NH₂-CNTs/Terephthalaldehyde/1,3,5-tris-(4-aminophenyl) benz-ene = 11:4:7, *m:m*) sensor was developed for the detection of furazolidone (Y. Sun et al., 2020). The detection range was from 0.2 $\mu\text{mol/L}$ to 100 $\mu\text{mol/L}$ and the limit of detection was $7.75 \times 10^{-8} \text{ mol/L}$. In addition, it also showed adequate recovery (87.8%–126.9%) for the detection of furazolidone in chicken and lamb samples.

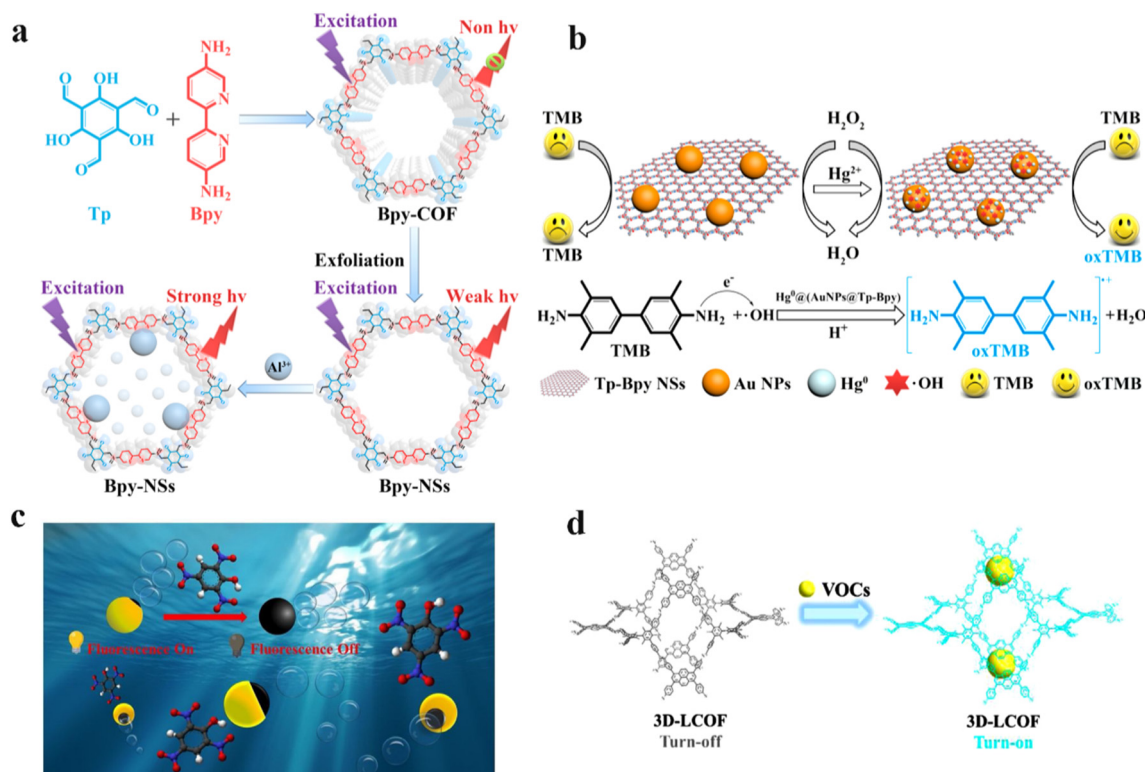


Fig. 14. (a) Schematic diagram for preparation of COF and detection of Al³⁺ (Cui et al., 2019a); (b) Mechanism of colorimetric detection of mercury ions using AuNPs@Tp-Bpy (Cui et al., 2019b); (c) Schematic illustration of fluorescence detection of TNP by COF-based micromotors (K. Wang et al., 2020); (d) Schematic diagram of the decoding of 3D COF (Y. Liu et al., 2020).

The rapid and simple detection of EDCs remains challenging. Recently, bisphenol A and bisphenol S were quickly detected by a COFs electrochemical sensor (Pang et al., 2020). The ranges for the detection of bisphenol A and bisphenol S were 0.1–50 mmol/L and 0.5–50 mmol/L with detection limits of 0.02 mmol/L and 0.09 mmol/L ($S/N = 3$), respectively, owing to its outstanding electrocatalytic activity and electrochemical responses. Furthermore, this sensor showed considerable repeatability, selectivity, and stability and was able to successfully detect bisphenol A and bisphenol S in actual environmental samples.

COFs also play a vital role in detecting explosives in the environment (K. Wang et al., 2020; M. Wang et al., 2020; Chakravarty et al., 2018). The fluorescent self-propelled COFs microsensor achieved effective and fast detection of 2,4,6-trinitrophenol (several minutes), along with MnO_2 microsphere catalysts and magnetic Fe_3O_4 nanoparticles (K. Wang et al., 2020). When hydrogen peroxide was broken down into oxygen bubbles, an autonomous movement guided by magnetic force occurred under the driving force of oxygen bubbles. The interaction between 2,4,6-trinitrophenol and the microsensor caused fluorescence quenching even when the concentration of 2,4,6-trinitrophenol was as low as 1 mg/L (Fig. 14c). Fluorescent sensors constructed using magnetic COFs, molecularly imprinted polymers, and carbon dots () also showed highly sensitive detection for 2,4,6-trinitrophenol with a detection limit of 100 pmol/L and a detection time of 1 min (M. Wang et al., 2020). Chakravarty et al. proved the practical value of azine-linked COFs in the selective sensing of nitroaromatics (Chakravarty et al., 2018).

COFs sensors for gas detection have also been continuously explored. CTFs containing a triazine ring were prepared and showed a fast response and excellent selectivity for NH_3 detection (Niu et al., 2020). Both theoretical calculations and experimental results showed that the sensing performance was attributed to the interaction of the NH_3 molecule and nitrogen atom in the triazine ring, which enhanced the adsorption of NH_3 and reduced the resistance of the sensor. Additionally, 3D COFs with high specific surface area ($875\text{ m}^2/\text{L}$) and microporous structure ($\sim 0.85\text{ nm}$) have exhibited outstanding recognition ability for more than 20 types of volatile organic compounds (VOCs) such as aromatic, alcohol-based, and other industrial common VOCs (Fig. 14d) (Y. Liu et al., 2020). Identification is also possible using 3D COFs when the concentration of nitro pesticides reaches the mg/L level.

Other kinds of matter, such as aflatoxin B1 and quinones, can also be detected via various COFs sensors. In addition, the detection of water and humidity is also a major application of COFs sensors. An iron-porphyrin-based COFs electrochemical sensor was synthesized (Xie et al., 2020). It not only could detect pH in the range of 3–9, but showed excellent detection range (6.85 nmol/L to 7 $\mu\text{mol/L}$) for H_2O_2 .

4.6. Energy storage

Another area of application of COFs is energy storage, mainly for batteries and supercapacitors. COFs with periodic ordering, well-defined porosities, and excellent chemical stability not only allow electron and chemical species access but do not interfere with cycle life, thereby facilitating COFs as an excellent candidate material for energy storage.

4.6.1. Battery

Rechargeable batteries such as Li-ion batteries, Li-S batteries, and Li-Se batteries, have attracted increasing attention for energy conservation and emission reduction due to their high theoretical energy and energy density. COFs have been identified as promising materials for battery applications due to excellent thermal stability, tunable structure.

For Li-ion batteries, the biggest challenge was their dissolution into electrolytes, which reduced the recycling life of batteries. To solve this problem, COFs with $C=O$ and $C=N$ groups were designed and applied

to the cathode material of Li-ion batteries (Wu et al., 2020). Then, a high reversible capacity (502.4 mA h/g) at 0.05C and a high rate performance (170.7 mA h/g), even at 7.73 A/g, was obtained. To avoid the capacity decay of the electrode, COFs as coating layers have also been used to modify Si nanoparticles to reduce electrolyte decomposition (Ai et al., 2020). As expected, the specific capacity (1864 mAh g^{-1}) and current density (2000 mA/g) were dramatically improved. The limited capacity of Li-ion batteries hinders them to meet the demand for increasing energy. COFs-based Li-S batteries and Li-Se batteries were developed to overcome the aforementioned shortcomings, greatly improving the long cycle and high rates performance of batteries. COFs as separators for Li-S (Se) batteries could effectively reduce the “shuttle” effect, thus achieving high-performance batteries. For example, an imine-based COF with 0.56 nm pores was used as the coating layer of the ceramic separator; the resulting Li-S batteries showed outstanding initial discharge capacity of 1415 mA h/g and suitable cycle performance (100 cycles at 0.5C) (J. Wang et al., 2017). A specific composite ceramic separator with COFs was applied to enhance the electrochemical performance of Li-Se batteries and exhibited stable charge/discharge capacity (700 cycles) and a high capacity of 126 mA h/g at 6C (Si et al., 2019).

4.6.2. Supercapacitors

The key to preparing high-performance supercapacitors was to choose materials with large surface areas and redox-active chemical groups. The supercapacitors were divided into two types: 1) based on electrical-double-layer-capacitance (EDLC) behavior supercapacitors and 2) based on pseudo-capacitance behavior supercapacitors. Unlike other inorganic materials (activated carbon), COFs possess distinctive designability, which allows their pores to be designed as microporous or mesoporous, leading to large surface areas and the integration of active sites into COFs, resulting in redox-active chemical groups (Halder et al., 2018; M. Wang et al., 2019). These features make COFs have promising potential in the field of supercapacitors.

A method for improving the performance of supercapacitors based on EDLC behavior is to accelerate the electrolyte ion mass transfer. Wang et al. prepared a hybrid material (COF/rGO) for supercapacitors (C. Wang et al., 2020). In this study, a high gravimetric specific capacitance (321 F/g) and volumetric specific capacitance (237 F/cm^3) by COF/rGO electrodes were obtained under given conditions, which surpassed the capacitance of the graphene electrodes. The excellent performance was attributed to the introduction of 2D COFs nanosheets, which enhanced the ion mass transfer and hindered the stacking of the rGO nanosheets. CTFs enriched with abundant nitrogen atoms could enhance the wettability of the electrolyte, introduce false capacity (aqueous electrolyte), and increase the effective surface area (Deng et al., 2016). For the first time, Hao et al. reported that an ionic-liquid-based supercapacitor prepared by the derivatives of CTFs showed desirable high surface area and conductivity (Hao et al., 2015). Furthermore, it was proven that the improvement of supercapacitor performance achieved the effect of nitrogen doping on the relative permittivity of the electrode materials. The size of the pores also affected the mass transport and ion diffusion of supercapacitors. For instance, hierarchical porous carbons with different pore sizes (2.5–8.0 nm) were prepared by pyrolysis of CTFs, and the experimental results showed that the larger the mesopores, the better the ion mass transport (Baumann et al., 2019). A high specific capacity value (155 F/g) was obtained under the given conditions. The addition of redox-active groups was considered to be an effective way to improve pseudo-capacitance behavior. Additionally, 2D COFs with redox-active pyridine units were used for the supercapacitor and showed a high specific capacitance of 102 F/g at 0.5 A/g and outstanding charge/discharge cycles (6000 times) (Khattak et al., 2016). Hybrid capacitors with EDLC and pseudo-capacitance behavior are an effective strategy to obtain high capacitance. The synergy of EDLC and pseudo-capacitance promoted charge

storage, thus achieving an excellent specific capacitance of 546 F/g at 500 mA/g and ~ 92 mF/cm² at 0.5 mA/cm² in an acidic solution and a solid-state device, respectively (Haldar et al., 2019).

5. Conclusion and perspectives

For the first time, the progress and hotspots of the COFs field have been described by the bibliometric method. Although many articles have reported on the design, synthesis, and application of COFs, these materials are still in their infancy in many aspects compared to traditional materials. This is because COFs, as an emerging material, has had a relatively short development history (2005–present). China, with 1373 articles, was the most active country, followed by the USA (507) in research on this material. The keywords of “photocatalysis” hold high frequency among all keywords, indicating that it is a current research hotspot. COFs with characteristics of a high specific surface area, a permanent porosity, orderly arrangement of porous channels, and an adjustable internal environment have become excellent candidate materials in the fields of adsorption, catalysis, membranes, sensors, and energy storage. In addition, the distinct thermal and chemical stabilities of COFs are also important indicators that determine the widespread use of COFs.

There are still many challenges that need to be solved, providing greater scope for improvement: 1) The preparation method still needs further improvement. At present, the preparation method has some unavoidable shortcomings such as stringent conditions, complicated synthesis steps, and expensive raw materials. Most of these methods are still in the laboratory preparation stage and have a long way to go before large-scale applications. It is imperative to find a preparation method with low cost, one that can be performed at room temperature, and one that requires mild conditions. For example, improvements to methods such as low-temperature synthesis, mechanochemical grinding, or microwave-assisted methods are expected to achieve industrial-scale production; 2) the relationship between the structure (e.g., crystallinity, conformation, porosity, etc.) and performance (e.g., adsorption, catalysis, sensor, etc.) of COFs is still unclear. For example, in the photocatalytic process, the band structure of COFs photocatalyst could have evolved to different valence bands or conduction bands and altered the band gaps driven by light sources while exhibiting desired or improved performances. Research on the structure–performance relationship is rare. Techniques such as in situ characterization and in situ reaction monitoring have been developed to solve these problems. 3) The functional diversification of COFs still needs to be further strengthened. COFs are used in many areas such as adsorption, catalysis, and sensors, but specific research on some applications is still lacking. For instance, there is little or no research on oxygen reduction based on COFs electrocatalysts. The photodegradation of organic pollutants by COFs is mostly concentrated in dyes or tetracyclines and little attention is paid to other types of pollutants. Moreover, there is a lack of a unified photocatalytic evaluation standard to evaluate the photocatalytic performance of COFs because photocatalytic activity can be affected by materials, temperature, light source, light intensity, and time. Compared with the research in the field of adsorption, there is less research in the fields of membranes and sensors. Research on water treatment applications of membranes still needs to be expanded and new sensors need to be developed, such as biological sensors, surface-enhanced Raman sensors, and surface plasmon resonance sensors. 4) In the future, the safety risk assessment research of COFs materials should be strengthened. When the characteristics of COFs are fully utilized, the safety of COFs materials must also be considered. Currently, regarding COFs entering the environment, the biological toxicity of COFs research on the ecosystem and human health is in its infancy. In summary, although COFs research still has many challenges, in the past few years, COFs have shown outstanding advantages. We believe that COFs materials will be considered among the best materials and will play a major role in practical applications.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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