Recent Progress of Novel Porous Materials in Wastewater Treatment

Deqi Kong*, Hua Chen*, Zhen Xiang** and Bin Wang***†

*Sichuan Institute of Building Research CO., Ltd., Chengdu, 610084, China
**Sichuan Huashi Overseas Investment & Engineering CO., Ltd., Chengdu, 610081, China
***Faculty of Quality Management and Inspection & Quarantine, Yibin University, Yibin, China
†Corresponding author: Bin Wang; binwang17711@163.com

ABSTRACT

Unavoidably, the expansion of industry causes the release of numerous heavy metals, radionuclides, and organic pollutants into the environment. Due to these pollutants, the extremely toxic, highly carcinogenic chemicals provide a serious risk to people and aquatic life. Wastewater pollutants must be removed to safeguard the ecology. A huge specific surface area, multiple binding sites, a plethora of functional groups, variable pore size, and simplicity of surface modification are just a few advantages of porous materials. They are considered viable candidate materials for the efficient and selective removal of contaminants from aqueous solutions in a range of difficult circumstances due to their benefits. This work reviews the characteristics, methods of functionalization, and ways of modification of many novel porous materials in recent years. The use of these porous materials in the treatment of wastewater was examined. The development potential of porous materials is finally summed up.

INTRODUCTION

Urbanization and the rapid advancement of industrial technology have greatly improved people's quality of life, but they have also brought up new environmental issues, including widespread concern over water contamination (Mestre & Carvalho 2019, Osorio et al. 2016).

These encompass natural dyes, personal care products (PPCPs), endocrine disruptors, and some non-biodegradable Persistent organic pollutants (POPs) from the fabric industry (Michael et al. 2013). In particular, antibiotics in PPCPs, such as xanthamine antibiotics (SAs), are widely used to treat bacterial infections in humans, plants, and livestock, and most of them can't be totally metabolized (Doretto et al. 2014). More than 10 types of SA have been detected in rivers, sediments, groundwater, and soils in many countries, including the United States, the United Kingdom, China, and Australia (Verlicchi et al. 2012). Studies have proven that the concentrations of common sulfonamide Sulfamethoxazole (SMZ) in wastewater are in the vary of 100-2500 ng.L^{-1}, and these in floor water are in the range of 60-940 ng.L^{-1}, the concentration detected in drinking water used to be about 12 ng.L^{-1} (Padhye et al. 2014). Although SMZ is launched at low stages and does not have direct poisonous results on humans, it can affect microbial communities and ecosystems, posing a long-term chance of elevated drug resistance and the manufacturing of resistant genes (Rodriguez-Escales & Sanchez-Vila 2016). Therefore, it is imperative to undertake some environmentally friendly techniques to do away with organic pollution from water.

The common strategies of natural wastewater remedy are adsorption, chemical precipitation, biofilm method, electrochemical method, photocatalytic technique, and so on (Reza et al. 2017). Adsorption, biofilm, and other techniques can correctly treat natural pollutants in water, but these techniques are via using some substances to adsorb natural pollution to its surface. This manner belongs to the transfer of pollution process, easy to cause the use of substances on the environment again pollution. Therefore, it is common for post-use materials to be similarly processed, which will result in a bigger cost. Chemical precipitation and electrochemical methods are additionally frequent methods in the therapy of organic pollution in water. Still, in the method of reaction, it is handy to produce toxic and unstable metabolites, which will have negative outcomes on animal and human health. In recent years, photocatalytic technological know-how has received massive interest due to the fact it is a clean and efficient water pollution treatment method. Through the photocatalyst is excited by light, the light power is transformed into chemical energy, and the chemical response happens with the target so as to achieve the purpose of the cure of pollutants. Photocatalytic

science has been extensively used in wastewater sterilization, degradation of organic pollution in water, and photocatalytic cracking of water hydrogen production and different fields.

Porous substances have the advantages of a large, unique floor area, many binding sites, ample purposeful groups, controllable pore dimensions, and convenient floor modification. It is viewed as a manageable candidate material for efficient and selective removal of contaminants from aqueous solutions underneath complicated conditions. In this work, the properties, functionalization, and modification strategies of new porous materials such as porous natural polymer, metal-organic frameworks, MXene, and covalent organic frameworks are reviewed and discussed. As shown in Fig. 1, this work critiques the application of four porous materials in the therapy of heavy metallic ions, radionuclide contaminants, and natural pollutants from aqueous solution. It presents beneficial education for the lookup of wastewater therapy and material development.

SYNTHESIS OF NOVEL POROUS MATERIALS

Metal-organic Frameworks

The technical constraints of conventional porous materials and nano-based materials for environmental applications have been acknowledged, and metal-organic frameworks (MOFs) have been identified as a promising alternative. MOFs are uniformly organized porous materials made of positively charged metal ions and organic ligands that act as bridges. Because of the special chemical flexibility provided by their metallic centers and organic linkers, their shape and porosity can be rationally tailored. Different MOFs can be created through inventive synthetic design using a variety of metal clusters and organic linkers.

The utilization of MOFs in numerous fields, together with sensors, catalysis, electricity storage, separation, storage, drug transport systems, nonlinear optics, and many others. Their feasibility toward wastewater cure for a number of pollution (e.g., heavy metallic ions, pesticides, volatile organic pollutants (VOCs), and other hazardous chemicals) has been evaluated.

A magnetic zirconium-based metal-organic framework nanocomposite was created by Far et al. (2020) using a simple solvothermal process, and it was previously employed as an adsorbent to remove direct and acid dyes from aqueous solution. The examined direct and acid dyes were absorbed by the dendrimer-functionalized magnetic composite at rates of 173.7 and 122.5 mg.g^{-1}, respectively, which was previously higher than that of the available magnetic adsorbents. This research offers fresh perspectives on the creation and application of hybrid magnetic adsorbents, as
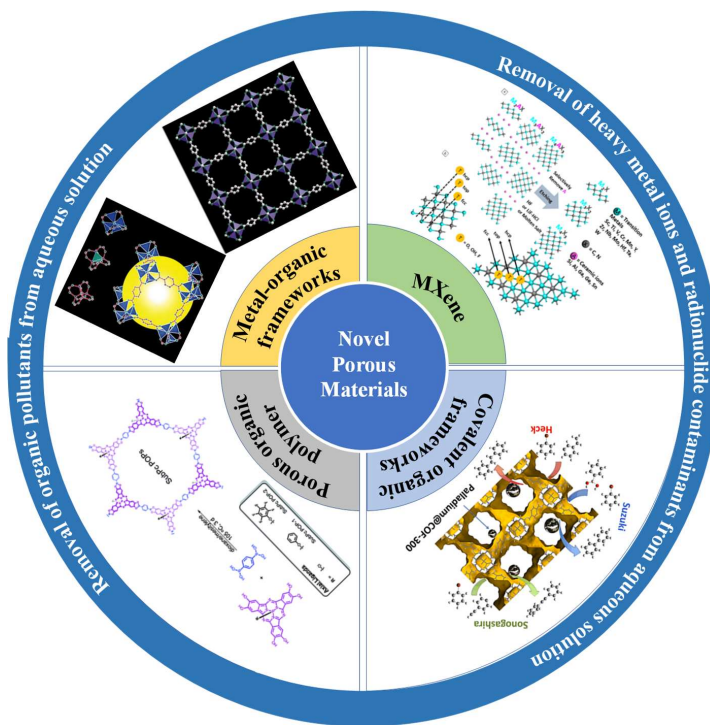

Fig. 1: Schematic illustration of the application of four porous substances in the treatment of heavy metallic ions, radionuclide contaminants, and natural pollution from aqueous solution (Gonçalves et al. 2016, Wu & Jiang 2023, Eder et al. 2017, Li et al. 1999).
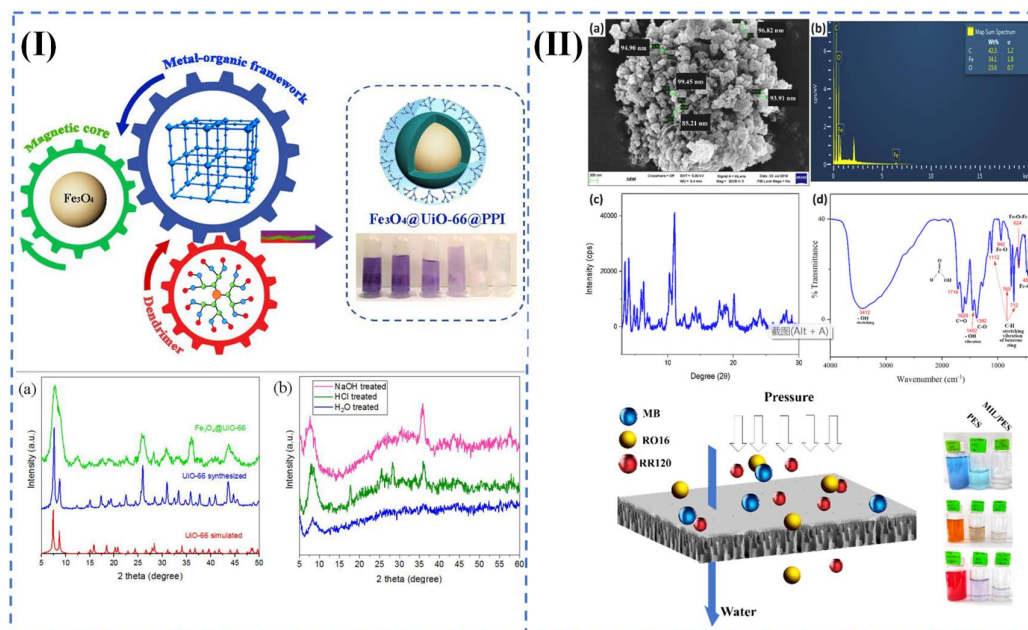
Fig. 2: (I) Fe_3O_4@UiO-66 nanocomposite's XRD patterns before and after a stability test, as well as the adsorption of the dye AB92, (II) FESEM picture, EDX, XRD, and FTIR of MIL-100(Fe), as well as the polymeric membrane modified with Fe-MOF nanoparticles rejecting both cationic and anionic dyes.

depicted in Fig. 2 (I), that combine the synergistic properties of nanoporous metal-organic frameworks and dendrimers with a wide range of functional corporations.

Johari et al. (2021) fabricated a hybrid flat sheet combined matrix membranes by blending polyethersulfone (PES) with an exclusive weight percentage of ferric-based metal-organic framework (Fe-MOF). The results showed that the Fe-MOF/PES membranes had first-rate rejections ($>98.5\%$) for cationic and anionic dyes, and the permeation flux was as high as 165.68 $L.m^{-2}h^{-1}$. Fig. 2 (II) depicts a schematic illustration of the preparation.

MXene

MXenes has a layered structure resembling graphene. It has garnered a lot of interest due to its distinct structure, exceptional physical chemistry characteristics, and great
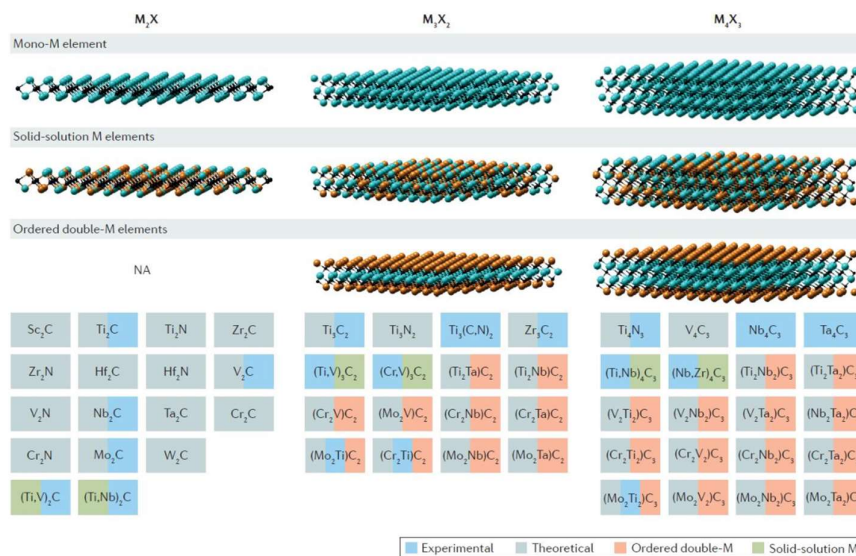

Fig. 3: Mxene model diagram with different structures (Naguib et al. 2011).

carrier mobility. MXenes' surface functional groups and elemental composition both have a significant impact on the properties of electron dispersion and carrier transport. It is possible to modify the surface functional groups and electrical characteristics, opening the door to the creation of composite catalysts based on MXenes and the subsequent production of more effective catalysts.

MXene is a substance made of transition metal carbides and nitrides with the chemical formula $M_{N+1}X_nT_x$ (n = 1-3) (Tariq et al. 2018, Folorunso et al. 2021), where M stands for transition metal elements belonging to the IIIA and Iva families, such as Ti, Sc, Cr, V, Zr, Nb, Mo, Hf, and Ta; X is either C or N; and T_x is the surface functional groups -O, -OH, and -F. As shown in Fig. 3, the solid solution structures of at least two metals are $(Ti, V)_2C$, $(Cr_2Ti)_3C_2$, and $(Nb, Zr)_4C_3$. MXene contains three molecular formulas, M_4X_3, M_3X_2, and M_2X_1, which are stacked alternatively in the form of $[M-X]_nM$. Bimetallic structure $(Mo_2Ta)C_2$, $(Cr_2V)C_2$, and $(Mo_2Ta_2)C_3$ that is ordered.

MXene has been extensively used in recent years in the domains of energy storage (Naguib & Gogotsi 2015), electromagnetic shielding (Shahzad et al. 2016), biosensors (Shi et al. 2019), water purification (Ihsanullah 2020), electrocatalysis and photocatalysis (Wang & Lee 2020) due to its properties of having a large specific surface area, metal conductivity, and tunable energy band. Particularly, it has been demonstrated that MXene is a highly effective co-catalyst whose conductivity is on par with noble metals.

Chen et al. (2019) organized the composite with excessive catalytic pastime via loading MoS_2 on MXene

lamellar through the hydrothermal method. Under visible light conditions, the hydrogen generation rate reached 9679 $\mu mol \cdot g^{-1} \cdot h^{-1}$, as shown in Fig. 4 (I). My Tran (et al. 2021) produced MXene by Al-selective etching of Ti_3AlC_2 MAX phase with HF. The mainly F-terminated $Ti_3C_2T_x$ MXene was shown to be an effective methylene blue (MB) adsorbent in wastewater. Surprisingly, the mainly F-terminated MXene absorbed 92% of MB in 20 μM of MB aqueous solution in 5 minutes. Furthermore, the MXene was largely recyclable, allowing for recurrent use. Fig. 4 (II) depicts a schematic illustration of the preparation.

Covalent Organic Frameworks

Covalent organic frameworks (COF) are a novel class of porous materials that have received a lot of research (Bukhari et al. 2023). COF is highly organized via reversible reactivity, in contrast to conventional short-range covalent polymers joined by irreversible condensation. By using pre-designed structures, COF may perform specialized tasks in contrast to typical porous crystalline materials like zeolite and metal-organic framework (MOF). They are architecturally diversified, low-density materials with the advantages of excellent thermal stability and permanent porosity (da Silva et al. 2023).

Cote et al. (2005) produced COF for the first time in 2005. Following it, there were numerous COFs. These characteristics of organic porous polymers make them suitable for use in optoelectronics, sensing (Martínez-Perinan et al. 2022), drug delivery, adsorption, separation (Dautzenberg et al. 2023), catalysis (Pachfule et al. 2018), energy storage, and drug delivery. COF, as a powerful metal-

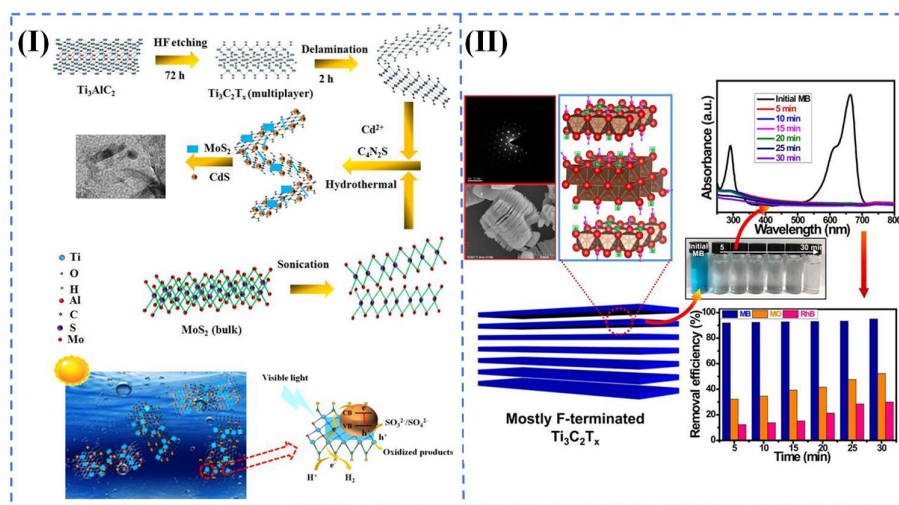Fig. 4: (I) The schematic illustration for the preparation of CdS-MoS_2-MXene by hydrothermal method and the mechanism of the CdS-MoS_2-Mxene catalyst under visible light, (II) the structures of $Ti_3C_2T_x$ MXene and the adsorption performance for MB.

free photocatalyst and as a contaminant-removal adsorbent, is very challenging.

Ruidas et al. (2023) prepared a C_6-TRZ-TPA COF by segregation of donor-acceptor moieties with the extended Schiffbase condensation between tris(4-formylphenyl)amine and 4, 4′, 4″ - (1, 3, 5-triazine-2, 4, 6-triyl)trianiline. The COF had a Brunauer-Emmett-Teller (BET) surface area of 1058 $m^2 \cdot g^{-1}$, and it had a pore volume of 0.73 cc.g^{-1}. The COF has been investigated as a potent metal-free photocatalyst for wastewater treatment and as an adsorbent for iodine extraction, and it could be used to harness solar energy for environmental cleanup. Fig. 5 (I) displays a schematic illustration of the preparation.

El-Mahdy et al. (2020) created the bifluorenylidene-based covalent organic frameworks (COFs) BFTB-PyTA, BFTB-BFTB, and BFTB-BCTA using one-pot polycondensations of BFTB-4CHO with PyTA-4NH$_2$, BFTB-4NH$_2$, and BCTA-4NH$_2$. These three COFs have outstanding crystallinities, high specific surface areas, and exceedingly high thermal stabilities. Rhodamine B (RhB), a tiny dye molecule, was exceptionally well-adsorbed by the COFs in water, outperforming all previously reported COFs, conjugated polymers, activated carbons, and other typical nanoporous adsorbents with maximum adsorption capacities of up to 2127 mg.g^{-1}, as shown in Fig. 5 (II).

Porous Organic Polymer

Porous materials have completed the transition from traditional inorganic hybrid materials to porous organic materials, realizing the cross-fertilization of inorganic and organic chemistry, showing the vigorous development of porous materials and broad application prospects. As an important branch of porous materials, porous organic polymer (Pops) materials have developed rapidly in recent years. According to the different structures and synthesis strategies, POPs can be classified into covalent organic building block materials, super-connected polymers, self-microporous polymers, conjugated microporous polymers, porous aromatic skeletal materials, and covalent triazine organic skeletal materials. Depending on the diversity of organic chemistry, there are many pathways for the synthesis of Pops, including Yamamoto coupling, Suzuki coupling, Sonogashira-Hagihara cross-coupling, oxidative polymerization, imine condensation, nitrile trimerization, and solvate-thermal radical polymerization reactions.

Comparison of organic-inorganic hybrid materials such as metal oxide inorganic porous materials and metal-organic skeletal compounds. The advantages of Pops are outstanding: the molecular chain consists of only light elements, and the skeleton density is low. The structural units are designed and synthesized by organic reactions and are easy to functionalize. Covalent bonds connect the structural units and have highly stable physicochemical properties for use in high-temperature, high-pressure, humid, or acidic environments. The structural units are mostly rigid aromatic structures that resist large surface tensions during drying and prevent pore collapse, resulting in permanent pore structures.
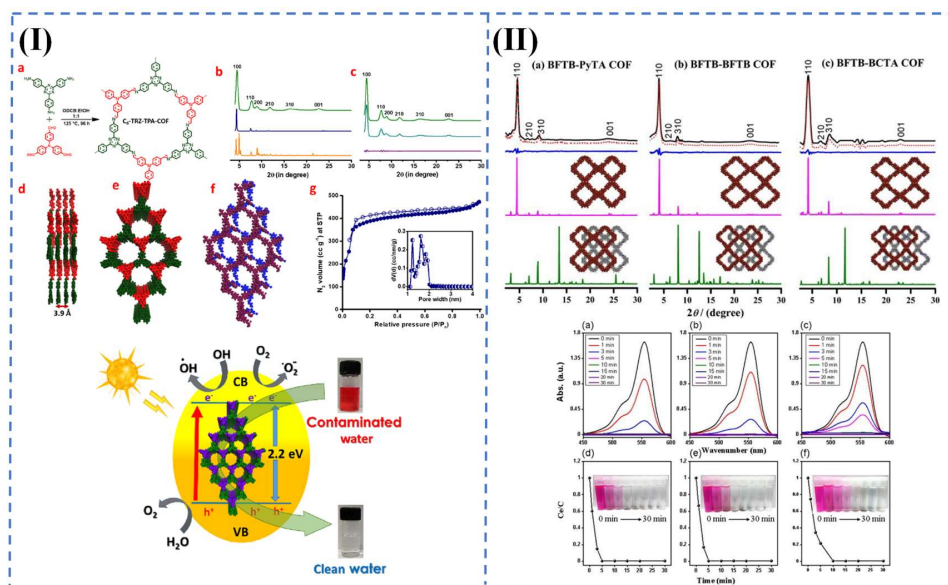

Fig. 5: (I) A schematic illustration of the preparation of C6-TRZ-TPA COF and their properties characterizations, (II) PXRD spectrum of the BFTB-PyTA, BFTB-BFTB, and BFTB-BCTA COFs and their adsorption results of RhB solution.
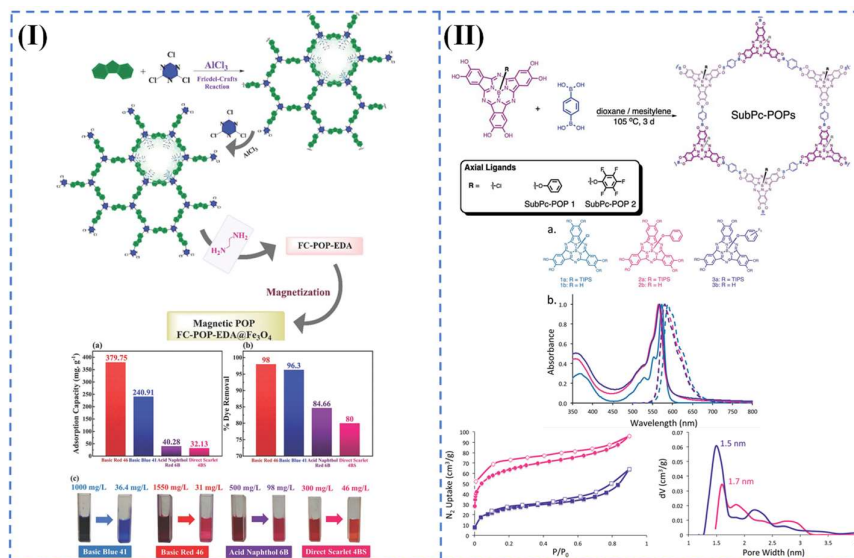
Fig. 6: (I) Preparation of FC-POP-EDA@ Fe_3O_4 and the dye removal effect, (II) preparation of the SubPc-POPs and the structures of monomers.

An accessible binding site-containing magnetic POP(FC-POP-EDA@Fe_3O_4) with amino functionality was created by Taheri and Dinari (2022). The mesoporous FC-POP-EDA@ Fe_3O_4 structure has a specific surface area of 402.11 $m^2.g^{-1}$ and well-defined crystallinity. Compared to anionic dyes, it showed better adhesion toward cationic dyes. For the synthesized POP, basic red 46, basic blue 41, direct scarlet 4BS, and acid naphthol 6B, respectively, had maximal adsorption capacities of 379.75, 240.9, 32.13, and 40.28 $mg.g^{-1}$. The findings show that FC-POP-EDA@Fe_3O_4 is an effective adsorbent for the selective filtration of cationic dyes from textile effluent. Fig. 6 (I) displays a schematic illustration of the preparation.

The creation and evaluation of two SubPc-based porous organic polymers (POPs) were described by Eder et al. (Eder 2017). The SubPc-POPs have reasonable surface areas, and in the solid state, their Q-bands are considerably redshifted. Fig. 6 (II) displays a schematic illustration of the preparation.

APPLICATIONS OF POROUS MATERIALS IN WASTEWATER TREATMENT

Removal of Heavy Metal Ions and Radionuclide Contaminants from Aqueous Solutions

Heavy metals are significant environmental pollutants that have a high chemical toxicity, are easily absorbed by humans and ecosystems, and constitute a major threat to both human health and the environment's ecological balance. By incorporating heavy metal ion adsorption sites into the pore structure of porous organic polymers by synthetic design, it is conceivable for porous organic polymers to function as efficient adsorbents for heavy metal ions.

Common heavy metal and radionuclide pollutants (such as Cd^{2+}, Cu^{2+}, Hg^{2+}, Pb^{2+}, U(VI), etc) in the water environment are difficult to degrade. As kinds of porous materials, they have been widely used in the removal of heavy metals in water environments in Table 1.

Nuclear energy can significantly lessen the strain of the energy problem in today's society because it has a high energy density and is a low-carbon energy source. Uranium, a crucial resource for the growth of nuclear energy, directly affects the long-term viability of the industry. On the other hand, uranium and its derivatives are highly polluted, radioactive, and poisonous.

A wide range of conventional materials, such as mineral clays, oxide aerogels, hydroxylapatite, metal sulfides, carbon-based materials, and hybrid materials based on graphene oxide (GO), have been created for the adsorption of toxic/radioactive metal ions. However, the following areas still have certain restrictions: Poor adsorption selectivity, slow adsorption kinetics, weak recyclability, difficulty in repeated treatment, poor environmental stability, complex material preparation process, difficulty in producing quickly on a large scale, and low maximum adsorption capacity of the adsorbent are all to be considered. To get over the constraints of present practical applications, it is critical to create novel materials for the manufacture of adsorbents.

New porous materials, such as covalent organic frameworks (COF) and metal-organic frameworks (MOF), have broad application prospects in the fields related to

Table 1: Removal of heavy metal ions and radionuclide contaminants from aqueous solution by porous materials.

Pollutant	Material	$S_{BET}/S_{Langmuir}$ [m^2.g^{-1}]	C_0 [mg.L^{-1}]	Q_{max} [mg.g^{-1}]	Removal rate [%]	Reference
Cr(VI)	ZIF-8	-	2.5	0.15	0.1	(Niknam Shahrak et al. 2017)
	Walnut shell biochar	-	110	-	93	(Kokab et al. 2021)
	MS-HMS-PL	414.5	-	257.67	-	(Soltani et al. 2020)
	Chitosan-g-PNVCL/ZIF-8 nanofibers	-	500	495.6	49.6	(Bahmani et al. 2019)
Hg(II)	FT-MOF	-	10	-	99.95	(Mon et al. 2017)
	Magnetic bentonite (M-B)	206.71	50	26.18	99.52	(Zou et al. 2018)
	Sulfurized magnetic biochar (SMBC)	337.5	-	8.93	-	(Hsu et al. 2021)
	Th-1	610	-	125.0	-	(Modak et al. 2019)
	Th-2	750	-	145.0	-	
	Th-3	480	-	128.0	-	
Pb(II)	UiO-66	1578	100	-	4	(Saleem et al. 2016)
	AT-MAC	-	-	310.9	99.5	(Waly et al. 2021)
	TMU-4	-	0.1	237	-	(Tahmasebi et al. 2015)
U(VI)	MIL-101-CMPO	2365	30	5.32	-	(De Decker et al. 2017)
	CMPO-functionalised polymers	-	-	26.7	-	(Annam et al. 2018)
	Polymer-coated polyacrylinitrile	20.9	50	33.0	-	(Aly et al. 2017)

environmental protection because of their excellent chemical stability, designability, and porosity. As kinds of porous materials, they have been widely used in the removal of heavy metal ions and radionuclide contaminants from aqueous solution in Table 1.

Removal of Organic Pollutants from Aqueous Solution

Textile, leather, plastic, paper, cosmetics, pharmaceuticals, printing, and other industries utilize organic dyes extensively. Dye waste that is inappropriately disposed of into the water cycle will unavoidably harm the aquatic ecosystem because dyes are non-degradable and carcinogenic. Adsorption is currently thought to be one of the most efficient ways to get rid of organic dyes. Still, conventional adsorbents like activated carbon, zeolite, and natural fiber have several drawbacks, including poor selectivity, poor reusability, and so on.

More and more people are becoming interested in porous polymers. They have the benefits of a high specific surface area, low mass, low density, high thermal stability, and high water stability when compared to other porous materials. There have been numerous types of organic porous polymers developed over the past ten years, and they can be classified into two groups based on their structural makeup: crystalline organic porous polymers and amorphous organic porous polymers. Covalent organic framework materials (COFs) are a common example of crystalline organic porous polymers. Hypercrosslinked polymers (HCPs), self-microporous polymers (PIMs), conjugated microporous polymers (CMPs), and other such materials are examples of amorphous organic porous polymers. As kinds of porous materials, they have been widely used in the removal of organic pollutants from aqueous solutions in Table 2.

Table 2: Removal of organic pollutants from aqueous solution by porous materials.

Pollutant	Material	$S_{BET}/S_{Langmuir}$ [m^2.g^{-1}]	C_0 [mg.L^{-1}]	Q_{max} [mg.g^{-1}]	Removal rate [%]	Reference
Organic dye	ZIF-67@Fe$_3$O$_4$@ESM	1263.9	10,15,20	208.33	-	(Mahmoodi et al. 2019)
	COFs	-	-	-	99	(Pan et al. 2019)
	TPOP–SO$_3$H	1002	-	97.1	-	(Li et al. 2019)
Phenolic compound	CNTs–PDMS	-	-	25 mg.g^{-1} for 4-NP	-	(Turco et al. 2018)
	DPP	513.9	-	254.2	78.9	(Liu et al. 2022)
	FPP	772.8	-	339.4	81.9	
	R-g-Ch	-	-	188.6 mg.g^{-1} for phenol	-	(Heydaripour et al. 2019)

CONCLUSIONS AND PERSPECTIVES

Porous materials are viewed as potential candidate materials for the effective and selective removal of contaminants from aqueous solutions under challenging circumstances because of their advantages of having a large specific surface area, numerous binding sites, abundant functional groups, controllable pore size, and convenient surface modification.

In this work, a sizable number of published literatures that are crucial to the adsorption process, such as pH value, temperature, and others, are reviewed. Another crucial element influencing the effectiveness of adsorption is the chemical and physical makeup of the adsorbents. Adsorbents having a lot of functional groups and a high specific surface area can improve this process. Future studies should focus on modification and composite materials, as well as ways to increase stability, decrease cost, evaluate toxicity, and increase adsorption capacity. There is still much to learn about how to regulate the synthesis conditions to create organic porous polymers with a greater specific surface area. The application of organic porous polymers in environmental remediation will be a significant area of research in the future.

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