

Review on design strategies and applications of metal-organic framework-cellulose composites

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ABSTRACT

Metal-organic frameworks (MOFs) are among the most attractive functional porous materials. However, their processability and handling remains a substantial challenge because MOFs generally occur in powder form due to their crystalline nature. Combining MOFs and cellulose substrates to fabricate engineered materials offers an ideal solution to broaden their utilization as functional materials. MOF/cellulose composites further provide remarkable mechanical properties, tunable porosity, and accessible active sites of MOFs. In this review, we summarize current state-of-the-art fabrication routes for MOF/cellulose composites, with a specific focus on the unique potential of utilizing three-dimensional bio-based cellulosic scaffolds. We highlight their utilization as adsorbents in the gas and liquid phase, for antibacterial and protein immobilization, chemical sensors, electrical energy storage, and other emerging applications. In addition, we discuss current limitations and potential future research directions in the field of MOF/cellulose composites for advanced functional materials.

1. Introduction

Metal-organic frameworks (MOFs) are a class of crystallized porous functional materials formed by the coordination of metal ions/clusters and organic bridging ligands (Fu et al., 2019; Li & Huo, 2015). Their structures can be specifically designed according to the targeted functionality by choosing the organic linkers' geometries and coordination modes of the inorganic metal ions (Zhu & Xu, 2014). Their key structural features are an ultrahigh porosity (up to 90% free volume) and a large internal surface area, possibly extending even beyond a Langmuir surface area of 10,000 m² g⁻¹ (Farha et al., 2012; Furukawa et al., 2010).

These properties are crucial for applications, such as gas storage and separation (Kang et al., 2017; Ren et al., 2015), water treatment (Yao et al., 2021), sensing (Kreno et al., 2012), catalysis (Chen et al., 2018; Nguyen et al., 2018), water harvesting (Kim et al., 2017), energy production (Gomes Silva et al., 2010; Khandelwal et al., 2020), and drug delivery (Falcão et al., 2014; Wu & Yang, 2017). However, there occurs one major drawback when it comes to the application of MOFs. Their powder morphology impedes processability and handling (Ashour et al., 2020). Hence, the integration and combination of MOFs with polymeric substrates is the method of choice to obtain durable advanced functional

MOF materials.

MOFs have been deposited or grown on/within various polymeric substrates resulting in MOF/polymer composites with hierarchical porosity and complex multi-layered networks (Peterson et al., 2021). These have been successfully utilized for water purification, nanofiltration, gas adsorption and separation applications (Zhang et al., 2016).

Nevertheless, the primarily use of non-renewable synthetic polymers derived from petrochemical products drove the need for more sustainable substrate systems. Alternatives comprise, biopolymers such as cellulose, cyclodextrin, chitin, chitosan, alginate, agarose, and heparin (El Hankari et al., 2019; Musarurwa & Tavengwa, 2022). In particular, cellulose is highly attractive as it represents one of the most abundant biopolymers on Earth, which is easily obtainable from plant cell walls (Liu, Ahmed, et al., 2021). Owing to its biodegradability, biocompatibility, renewability, and low-cost, cellulose provides a sustainable material platform for preparing functional materials, which has been widely demonstrated in materials for water treatment (Chen, Zhu, et al., 2020), sensing (Chen & Hu, 2021), antibacterial applications (Tavakolian et al., 2020), and energy storage (Wang, He, et al., 2021; Zhao et al., 2020). Cellulose possesses excellent mechanical properties and tunable

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chemistry, attracting a great deal of attention for the fabrication of MOF composites (Abdelhamid & Mathew, 2022; Kim, Otal, et al., 2019; Liu, Xiao, et al., 2021; Zhang, Wang, Ding, et al., 2021).

The cellulosic materials for MOF/cellulose composites can be of different morphology and chemistry and the cellulosic materials morphology (fibrillated cellulose or cellulose scaffolds), determines the corresponding fabrication routes for the structured MOF/cellulose composites. The first approach comprises the preparation of MOF/cellulose fibers by depositing MOFs on fibrillated cellulose *via ex-situ* methods or *in-situ* methods (Lu, Liu, et al., 2022). Then the MOF/cellulose fibers can be further processed into membranes or more advanced material structures with tunable pore size using various bottom-up fabrication strategies such as spinning, casting, filtration, freeze-drying, and three-dimensional (3D) printing. A novel approach is the direct utilization of 3D cellulose/bio-based scaffolds. Here, one profits from the hierarchical structure of biomaterials and avoids extra processing steps (Abdelhamid & Mathew, 2022).

In this review, we summarize current routes for preparing MOF/cellulose composites, including raw materials selection, cellulose pretreatments, MOF/cellulose composites preparation (Fig. 1). We put a specific focus on recent works and potential of utilizing 3D cellulose/bio-based scaffolds as starting materials for MOF/cellulose composites. In addition, we highlight applications of MOF/cellulose composites in gas adsorption and separation, wastewater treatment, antibacterial applications, protein immobilization, chemical sensors, and electrical

energy storage. At the end, we address current major limitations, challenges, and future prospects.

2. Preparation of MOF/cellulose composites

The preparation of MOF/cellulose composites is a multistep process that necessitates careful tuning of each individual step. It comprises three main steps: raw materials selection, cellulose pretreatment, and MOF/cellulose composites preparation.

2.1. Raw materials selection

The chosen raw components determine the composites preparation process. For example, the starting cellulose materials morphology, fibrillated cellulose, or cellulose scaffolds, decides whether a processing step (Section 2.4) is needed or not. For the MOF synthesis, one has the option to either directly utilize pre-synthesized MOF crystals (*ex-situ*) or to use MOF precursors for *in-situ* synthesis methods. In the following we provide a brief overview on the various sorts of raw materials that can be employed.

2.1.1. Cellulose

The starting cellulose material for MOF/cellulose composites can be either fibrillated cellulose or 3D cellulose scaffolds (Fig. 2) and is derived from sources such as trees, plants, and bacteria. Fibrillated

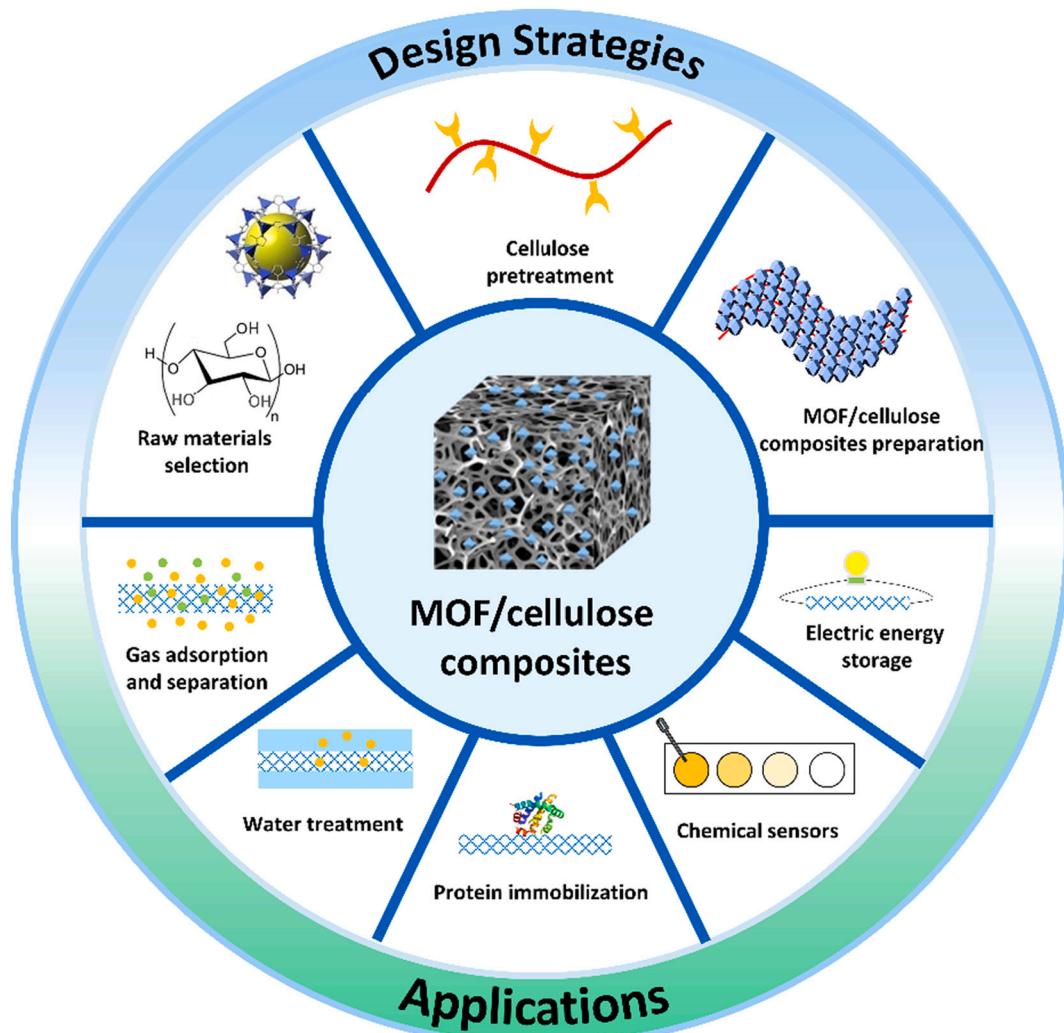


Fig. 1. Schematic of main design strategies and applications of cellulose/MOF composites.

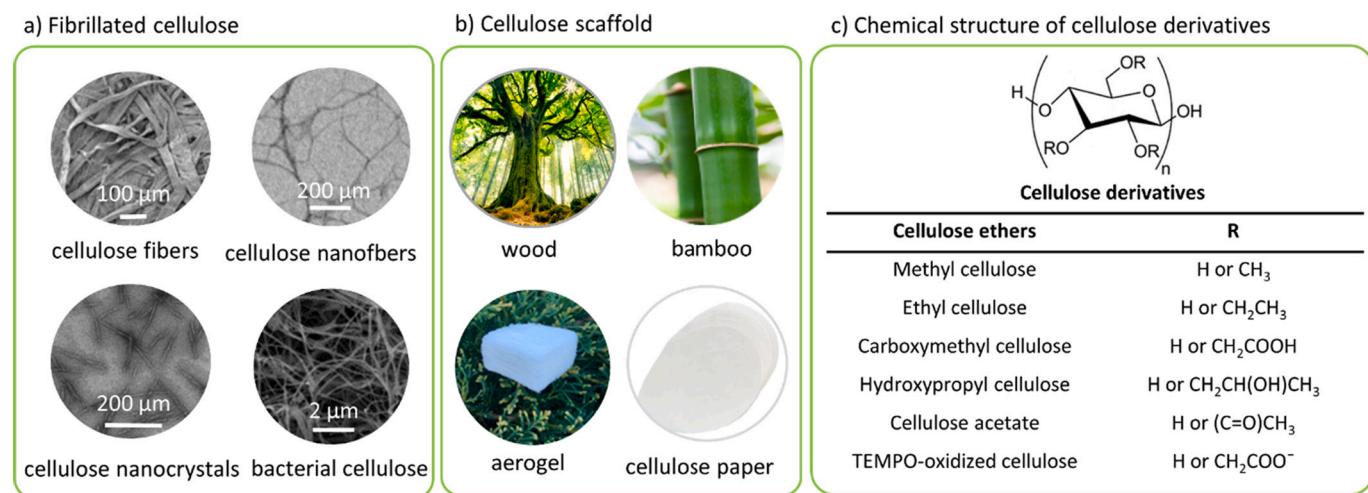


Fig. 2. Examples of a) fibrillated cellulose including cellulose fibers, cellulose nanofibers (CNFs), cellulose nanocrystals, and bacterial cellulose (Chen et al., 2011; Moon et al., 2011; Torres et al., 2012) and b) cellulose scaffolds including natural materials (wood and bamboo) and processed materials (aerogel and cellulose paper) used for preparing MOF/cellulose composites (Song et al., 2018). c) Chemical structure of cellulose derivatives.

cellulosic materials include fibers, microfibrils/nanofibrils, nanocrystals, and bacterial cellulose and they primarily differ in their morphology (e.g. dimensions and shape) (Fig. 2a) (Huang, Cai, et al., 2021; Moon et al., 2011; Seddiqi et al., 2021).

3D cellulose scaffolds can be directly obtained from natural materials characterized by a sophisticated hierarchical cellular structure, pronounced anisotropy, and good mechanical performance, such as wood, bamboo, and corncob (Duan, Liu, et al., 2019; Su et al., 2019; Wang, He, et al., 2021). Alternatively, artificial 3D cellulose scaffolds based on fibrillated cellulose materials are fabricated using bottom-up methods, such as aerogels, cellulose paper, and fabrics (Fig. 2b) (Emam et al., 2018; Huang, Cai, et al., 2021; Matsumoto & Kitaoka, 2016; Shen et al., 2019; Xu et al., 2019). Natural cellulose scaffolds and artificial scaffolds differ in their specific surface area, pore structure, and mechanical properties. The avoidance of time-consuming and energy-intensive processing steps, as well as the superior mechanical performance makes natural 3D cellulose scaffolds an ideal support material for MOFs.

2.1.2. MOFs

Despite the substantial number of developed MOFs, applicable MOFs for MOF/cellulose composites are restricted to several main MOF families, considering the required water, thermal, chemical, and mechanical stability, facile synthesis condition, and low cost.

Possible MOFs are MOF-199 (named Cu-BTC or HKUST-1) (da Silva Pinto et al., 2012; Ren et al., 2022; Wang et al., 2015), the ZIF (Zeolitic imidazolate framework) family (e.g., ZIF-8, ZIF-9, ZIF-12, ZIF-62, ZIF-67, and ZIF-90) (Au-Duong & Lee, 2017; Matsumoto & Kitaoka, 2016; Mubashir et al., 2021; Ren et al., 2018; Wang, Cao, et al., 2018), the MIL (Materials Institute Lavoisier) series (e.g., MIL-53(Al), MIL-101(Fe), MIL-88B(Fe)) (He et al., 2020; Mubashir et al., 2020; Qiu et al., 2019), and Zr-based MOFs (e.g., UiO-66 and UiO-67) (Wang, Ba, et al., 2019).

Pre-prepared MOF crystals are directly applicable for MOF/cellulose composites via *ex-situ* synthesis methods. Especially, in the case of elevated MOF synthesis temperatures, using MOF crystals as starting materials is preferable to avoid degradation of the lignocellulosic materials (such as wood) constituents. The corresponding main decomposition temperatures are as following: cellulose around 315 °C, hemicellulose 220 °C, and lignin 160 °C (Yang et al., 2007).

On the other hand, using MOF precursors (metal ions and organic ligands) as starting materials allows to prepare MOF/cellulose composites via *in-situ* synthesis methods (Section 2.3.2), which can avoid MOF particle aggregation and ensures a uniform distribution of MOFs on the cellulose substrates.

2.2. Cellulose pretreatment

A high interfacial affinity between MOFs and cellulose is crucial to avoid leakage of MOFs from the cellulose substrates. Hence, better interaction between MOFs and cellulose, and improved compatibility between MOFs and cellulose substrates require chemical pretreatments of cellulose before the MOF/cellulose composites preparation. For that purpose, the cellulose is chemically modified, including adjusting the cellulose charging, increasing the accessibility of the hydroxyl group, and adding functional groups, as summarized in Table 1.

2.2.1. Adjustment of cellulose charging

Changing the natural cellulose charging from neutral/slightly negative into a negative charge enables pronounced interactions between the obtained negatively charged cellulose and the positively charged metal cations of MOFs and form the nucleation site for MOFs. The MOF loading typically rises with the number of crystallization nucleation sites (Jhinjer et al., 2021; Qian et al., 2018; Rodríguez et al., 2014). Common methods for adjusting the charging comprise 2,2,6,6-Tetramethylpiperidine-1-oxyl (TEMPO)-mediated oxidation, carboxymethylation, and the incorporation of anionic sulfonate moieties.

TEMPO oxidation converts the C6 hydroxymethyl group of cellulose into carboxylate groups and equips the cellulose surface with a high density of negative charges (Tavakolian et al., 2020). For example, Matsumoto et al. (2016) utilized TEMPO-oxidized CNFs (TOCNFs) to prepare cellulose/ZIF-90 film (Matsumoto & Kitaoka, 2016).

Alternatively, carboxymethylation is also capable of adjusting the cellulose surface charge. The common sodium chloroacetate treatment in the presence of sodium hydroxide converts the -OH groups to negatively charged $-\text{CH}_2\text{COO}^-$ groups (Pinto et al., 2012). A rather specific case arises when native wood is utilized, as in that case wood itself possesses inherent carboxyl groups, which can be converted to carboxylate groups by simple sodium hydroxide treatment (Lu, Fan, et al., 2022; Tu et al., 2020). The carboxylate groups on the cellulose surface assist the initial coordination of metal ions by ion-exchange and facilitate the growth of MOFs (Park & Oh, 2017). Meanwhile, the metal center atoms in MOFs can establish coordination bonds with the carboxylate groups of the anionic cellulose and form electrostatic interactions between cellulose and MOF, which then stabilize MOF crystals on the cellulose surface (Duan et al., 2018; Fu et al., 2019).

As third option, cellulose equipped with a large number of anionic sulfonate moieties, can uptake and exchange metal cations, and supply adequate anchoring sites for MOF growth. A possible synthesis route

Table 1

Summary of methods and chemicals used for cellulose pretreatment.

Pretreatment Method		Cellulose Materials	Chemicals	Ref.
Adjust charging	TEMPO-mediated oxidation	Cellulose (nano)fiber, spruce pulp, corncobs	TEMPO, NaClO and NaBr	(Duan, Liu, et al., 2019; Valencia & Abdehamid, 2019; Wang, Wang, et al., 2021; Zhao et al., 2019; Zhou, Strømme, & Xu, 2019; Zhu et al., 2018)
	Hydrogen peroxide oxidation	Cotton	H ₂ O ₂ , sodium silicate and sodium hydroxide	(Abdelhameed et al., 2016)
	Carboxymethylation	Filter paper, cellulose fabrics, cotton fibers, lignocellulosic fibers, cellulose acetate	Sodium chloroacetate and NaOH	(da Silva Pinto et al., 2012; Duan, Liu, et al., 2019; Fu et al., 2019; Jhinjer et al., 2021; Laurila et al., 2015; Li, Hori, & Takemura, 2020; Park & Oh, 2017; Pinto et al., 2012; Rodríguez et al., 2014)
	Introducing anionic sulfonate	Wood	NaOH	(Tu et al., 2020)
	Dissolve cellulose	Cotton linter	Sodium p-styrene sulfonate and ammonium persulfate	(Ren et al., 2018; Song et al., 2020)
	Adding filler	Cellulose pulp	NaOH/urea/H ₂ O Precipitated calcium carbonate (PPC)	(Yang et al., 2017)
Increasing the accessibility of the hydroxyl groups	Catechol group	Cotton, bacterial cellulose	Dopamine hydrochloride, and tris (hydroxymethyl)aminomethane	(Au-Duong & Lee, 2017; Mirkovic et al., 2019; Zhou, Yuan, et al., 2019)
	Epoxy group	Cotton fabric, and linen, CNFs	3-Glycidyloxypropyltrimethoxysilane	(Abdelhameed, Emam, et al., 2017; Shen et al., 2019)

comprise the free-radical polymerization of sodium *p*-styrene sulfonate in the presence of acryloyl group-modified cellulose (Ko et al., 2018; Yang et al., 2020).

Commercially available cellulose derivatives with negative charges, such as carboxymethyl cellulose (V. Javanbakht & Rafiee, 2022), cellulose acetate (Abdelhameed et al., 2021; Chen, He, et al., 2021; Emam et al., 2021), and TOCNFs (Lin et al., 2021) are also widely used for the preparation of MOF/cellulose composites (Fig. 2c).

2.2.2. Increasing the accessibility of the cellulose hydroxyl groups

Strong inter- and intramolecular hydrogen bonds and van der Waals forces limit the accessibility of cellulose hydroxyl groups and therewith the fibers' reactivity. It can be enhanced by simple dissolving cellulose in alkaline/urea solutions or by adding inorganic fillers (Bui et al., 2008; Luo & Zhang, 2013). For example, cotton linter pulps were pretreated by dissolving in NaOH/urea (Ren et al., 2018), or NaOH/thiourea aqueous solutions (Zhang, He, et al., 2020) at low temperature before growth of MOF (Ren et al., 2018; Xiong et al., 2014). Yang et al. (2017) added precipitated calcium carbonate (PPC) to cellulose pulp as a filler to decrease the inter-fiber hydrogen bonds, making more hydroxyl groups available for the formation of ester functions with the MOF organic ligand (Yang et al., 2017). Due to the increase of active sites, the corresponding MOF loading and surface area of the MOF/paper composites increased accordingly.

2.2.3. Adding functional groups

By reacting with cellulose hydroxyl groups, functional groups such as amine groups, catechol groups, and epoxy groups can be introduced. These functional groups can form either hydrogen bonds with the metal moieties of MOFs or covalent bonds with the organic part of MOFs, facilitating the MOF nucleation, deposition and growth. For example, catechol moieties of a polydopamine (PDA) modified cellulose surface can bind strongly to a variety of metal ions, facilitating the formation of MOF crystals (Au-Duong & Lee, 2017; Cui et al., 2020; Zhou et al., 2015). The modification of cellulose with 3-glycidyloxypropyltrimethoxysilane (GPTMS) incorporates epoxy groups that can form covalent bonds with the amino moieties of specific MOF organic linkers (Abdelhameed, Kamel, et al., 2017; Shen et al., 2019).

2.3. Synthesis of MOF/cellulose composites

After the initial chemical modification of the cellulose material, MOF crystals can be deposited via *ex-situ* growth approaches (direct mixing and immersion coating) or *in-situ* growth approaches (one-pot synthesis, stepwise *in situ* growth, and layer-by-layer growth) which we explain in

more detail in the following.

2.3.1. Ex-situ growth methods

Ex-situ growth methods imply that MOF crystals are synthesized before fabrication of the MOF/cellulose composites and include two main approaches. In the case of fibrillated cellulose direct mixing is the method of choice whereas for cellulose scaffolds immersion coating is typical (Fig. 3).

2.3.1.1. Direct mixing. Direct mixing uses pre-prepared MOF powders or suspensions as starting materials and mixes them with fibrillated cellulose (Fig. 3a). It enables a high and controllable MOF loading. To avoid aggregation and precipitation of cellulose and MOFs, especially at high MOF loadings, ultrasonic and stirring processes are important during mixing. Cellulose pretreatments before the mixing stages help to increase the affinity and compatibility of cellulose and MOF powders. The obtained MOF/cellulose blends are easily processable into engineered constructs in subsequent steps. Based on this method, Zhu et al. prepared hybrid MOF/cellulose aerogels by mixing MOF nanoparticles, such as ZIF-8, UiO-66, and MIL-101(Fe) with aldehyde and hydrazide modified cellulose nanocrystals and assembled them into covalently cross-linked clusters (Fig. 3b and c) (Zhu et al., 2016). The prepared aerogels possessed a hierarchical porous structure with controllable MOF loadings of up to 50 wt%.

2.3.1.2. Immersion coating. Immersion coating involves submerging a cellulose scaffold in a solution containing MOF nanoparticles for a set time. (Fig. 3d). For example, Cu-BTC MOF/cotton composites were prepared by soaking hydrogen peroxide oxidized cotton fabric in a Cu-BTC MOF solution (Fig. 3e and f) (Abdelhameed et al., 2016). The Cu-BTC loading of the composites ranged from 5% to 10%, controlled by the concentration of the Cu-BTC MOF solution. Increasing the immersion coating cycles resulted in a higher MOF loading.

2.3.2. In-situ growth methods

In-situ growth methods are applicable for both fibrillated cellulose and cellulose scaffolds, and one distinguishes between one-pot synthesis, stepwise *in-situ* growth and layer-by-layer method (Fig. 4).

2.3.2.1. One-pot synthesis. One-pot synthesis is based on the reaction of MOF precursors and cellulose substrates in one reactor (Li & Huo, 2015) (Fig. 4a). Abdelhameed et al. compared Cu-BTC/fabric composites using one-pot synthesis and immersion coating method. The study revealed that the achieved MOF loadings by one-pot synthesis (102.1–110.0 mg

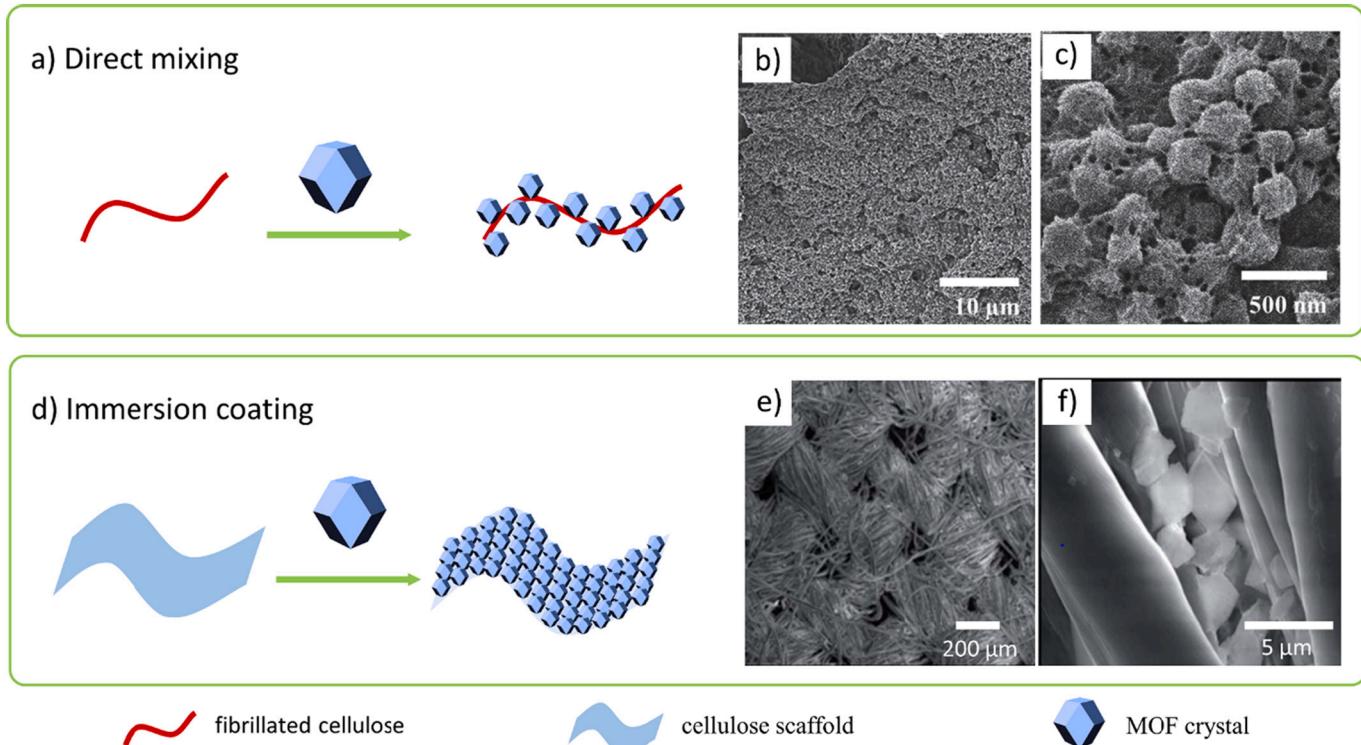


Fig. 3. *Ex situ* growth strategies for the preparation of cellulose/MOF composites. a) Direct mixing of MOF particles with fibrillated cellulose to obtain MOF/cellulose mixtures prior to processing. b-c) SEM images of UiO-66-containing cellulose aerogels prepared using direct mixing method (with 50 wt% UiO-66) (Zhu et al., 2016). d) Immersing cellulose scaffolds in MOF dispersion to prepare MOF/cellulose composites. e-f) SEM images of Cu-BTC@cotton prepared using immersion coating method (Abdelhameed et al., 2016).

g^{-1}) are two times higher than with the immersion coating method (Abdelhameed, Emam, et al., 2017). Mirkovic et al. also used one-pot synthesis to grow MOF-5 crystals on PDA modified cotton. The MOF-5 crystals were firmly attached to the cotton fibers and exhibited a stable “necklace” morphology (Fig. 4b and c) (Mirkovic et al., 2019).

2.3.2.2. Stepwise *in-situ* growth. Stepwise *in-situ* growth relates to the stepwise exposure of the cellulose substrate to the MOF precursors (metal ions and organic linkers), as shown in Fig. 4d. The metal precursors are first added and form the initial coordination between metal ions and the anionic groups of the pretreated cellulose, followed by adding the organic linkers for the MOF nucleation and growth.

Pinto et al. utilized a stepwise *in-situ* growth protocol to grow MOF-199 on carboxymethylated cotton (Pinto et al., 2012). The initial addition of copper acetate caused an ion exchange reaction between metal ions and carboxylate groups of the anionic cellulosic substrates, resulting in a dense copper ions layer on the cellulose surface that acted as nucleation center and anchor points for the formation of MOF-199 crystals. As the ion exchange provides the nucleation site for the subsequent growth of the MOF structure, the stepwise *in-situ* growth avoids the aggregation of MOF particles. They also compared the obtained results with the one-pot synthesis method and found that stepwise *in-situ* growth exhibited a higher yield of MOF-199 on cellulose.

2.3.2.3. Layer-by-layer. For the layer-by-layer synthesis method, the cellulose substrate is immersed in a solution of the metal ions, followed by submersion in the organic ligand solution (or vice versa), with possible rinsing steps in between (Summerfield et al., 2015). This method enables a homogeneous morphology and controllable coating thickness of MOFs via adjusting the number of growth cycles (Fig. 4g). Laurila et al. reported the *in-situ* crystal growth of HKUST-1 on carboxymethylated electrospun CNFs (Laurila et al., 2015). The MOF

loading increased from 11.8% after 8 synthesis cycles to 38.7% at 32 synthesis cycles (Fig. 4h and i).

2.3.3. Advantages and disadvantages of *ex-situ* and *in-situ* growth methods

The main advantage of *ex-situ* growth methods compared to *in-situ* growth methods is the controllable MOF loading, which is also beneficial for achieving high MOF loadings and therewith enhancing their performance, such as high adsorption capacity and catalytic activity. In addition, using pre-prepared MOF crystals avoids possible harsh MOF synthesis conditions during composites formation, for example synthesis temperatures being higher than the degradation temperature of the lignocellulosic constituents. This approach, however, suffers from MOF particle aggregation and uneven distribution within composites. On the contrary, *in-situ* growth methods ensure a strong interaction between MOFs and cellulose fibers, as well as the uniform MOF particles distribution, but at the expense of a high and controllable MOF loading, which can be increased by layer-by-layer methods.

2.4. Processing of cellulose/MOF composites

For applications, MOF modified cellulose materials based on fibrillated cellulose need to be processed into 2D membranes or 3D porous structures using bottom-up approaches such as spinning, casting, filtration, freeze-drying, or 3D printing (Section 2.4.1). In contrast, the direct utilization of natural or artificial 3D cellulose scaffolds as support for MOFs avoids these additional processing steps (Section 2.4.2).

2.4.1. Bottom-up approaches

2.4.1.1. Spinning process. As a common technique to fabricate cellulose fibers, electrospinning has also been employed for the synthesis of MOF/cellulose composites (Fig. 5a). First the MOF/cellulose suspension is transferred into a syringe for electrospinning, and then processed with a

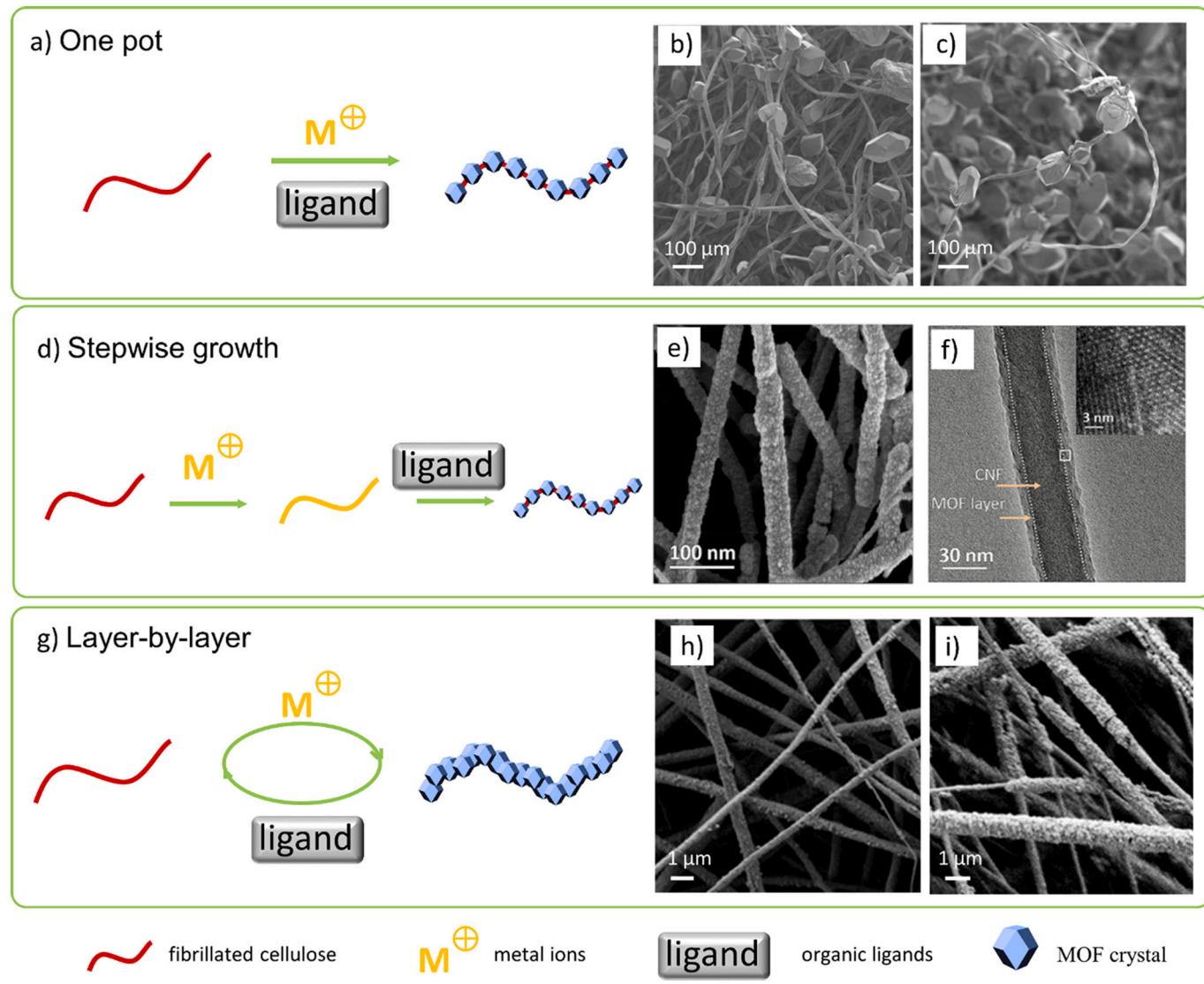


Fig. 4. *In-situ* growth strategies for the preparation of cellulose/MOF composites. a) Schematic of one-pot synthesis to prepare MOF/cellulose composites by mixing MOF precursors with fibrillated cellulose in one reactor prior to processing. b-c) SEM images of MOF-5 grown on cotton fibers using one-pot method (Mirkovic et al., 2019). d) Schematic of stepwise *in-situ* growth method to prepare MOF/cellulose composites by stepwise exposure of the cellulose substrate to the MOF precursors e) SEM image and f) TEM image of MOF/CNFs prepared using stepwise *in-situ* growth method (Zhou, Kong, et al., 2019). g) Schematic of using layer-by-layer method to prepare MOF/cellulose composites and controlling the MOF coating thickness and loading by adjusting the number of growth cycles. h-i) SEM images of HKUST-1/CNFs prepared layer-by-layer method with 8 cycles (h) and 32 cycles (i) respectively (Laurila et al., 2015).

constant rate under a high voltage. The electrospun nanofibers are collected on a rotary drum or a grounded collector (Zhang, Li, et al., 2020) and the fiber mats result after vaporization of the solvent (Hou et al., 2018; Mubashir et al., 2019). The technique allows for a simple fabrication of fiber composites for a wide variety of MOF/cellulose combinations with a wide range of possible sizes and morphologies. (Miranda et al., 2022) Moreover, the fiber diameters and the MOF loadings are tunable by varying the MOF/cellulose concentration and cellulose to MOF ratio in the initial spinning mixtures (Li & Huo, 2015; Mubashir et al., 2019).

2.4.1.2. Casting. Casting represents a straightforward way to build-up MOF/cellulose mixed matrix membranes (MMMs) (Fig. 5b) (Abdel-hameed et al., 2020). First, a MOF/cellulose suspension is poured onto a flat glass substrate or PTFE plate. After degassing, solvent evaporation, and drying, the MOF/cellulose membrane is peeled off from the flat substrate (Lee et al., 2019; Wang, Song, et al., 2019; Yang et al., 2019). The cellulose fraction in the MMMs serves as the matrix and the MOFs

function as filler. The composites prepared by casting show good permeability in gas separation, water purification and catalytic applications in continuous flow processes (Hou et al., 2017; Wang, Yao, et al., 2019; Yang et al., 2018; Yang et al., 2019).

2.4.1.3. Filtration. MOF/cellulose membranes' fabrication by filtration has gained tremendous interest in recent years (Fig. 5c). During filtration, the solvent of the MOF/cellulose suspension permeate the filtration membrane with the aid of a vacuum and after a drying process MOF/cellulose membranes with symmetrical structures are obtained (Zhu et al., 2022). For example, Xu et al. developed hierarchical porous and conductive nanosheets composed of ZIF-67/cellulose/CNTs for flexible, foldable electronic energy storage devices based on filtration (Xu et al., 2018). The suspension of cellulose, CNTs and ZIF-67 was first collected on a PVDF membrane filter and the membrane was received after drying. The thickness is adjustable by the amount of ZIF-67, cellulose, and CNTs in the initial mixture. In general, membranes obtained by filtration possess good flexibility, permeability, and hierarchical porosity (Voisin

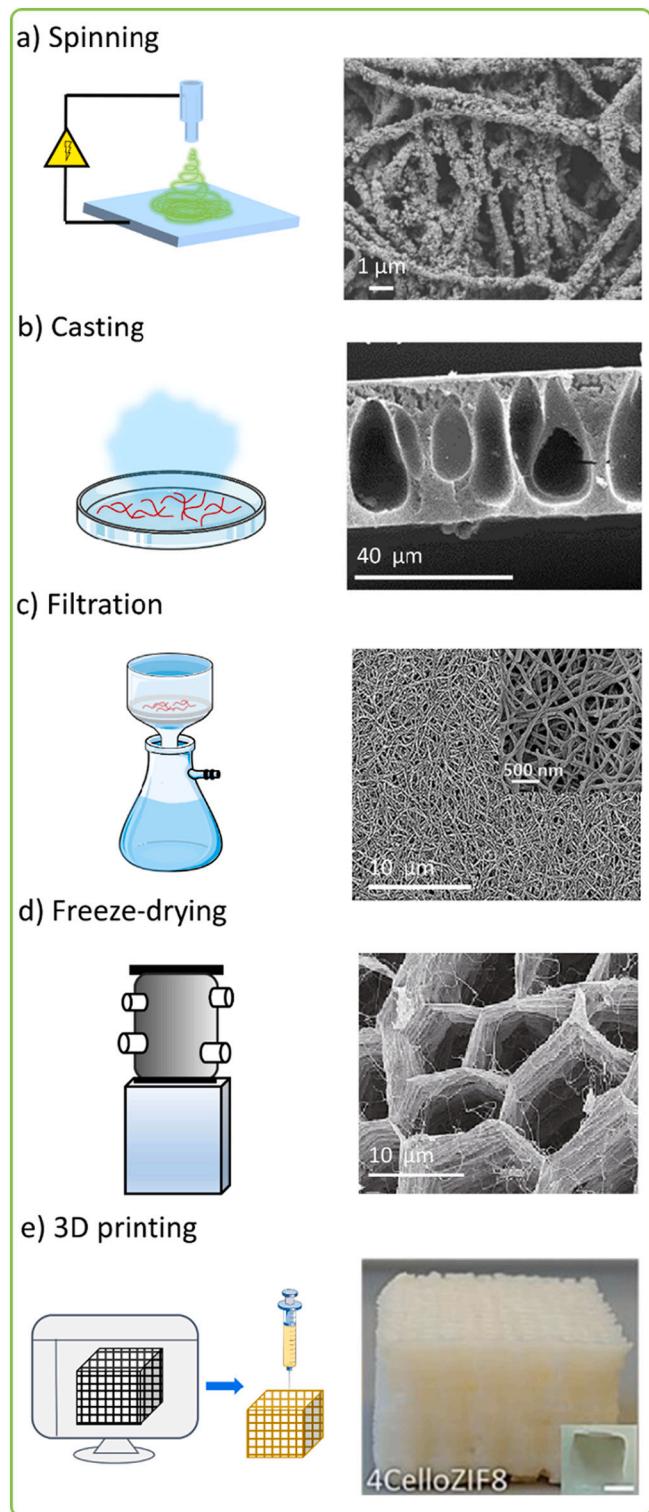


Fig. 5. Bottom- up processing strategies for preparation of cellulose/MOF composites using fibrillated cellulose as starting material: a) spinning (Laurila et al., 2015), b) casting (Kim, Choi, et al., 2019), c) filtration (Zhou, Kong, et al., 2019), d) freeze-drying (Zhou et al., 2020), and e) 3D printing (Sultan et al., 2019).

et al., 2017).

2.4.1.4. Freeze-drying. Freeze-drying is the most common way to build-up engineered porous MOF/cellulose composites (Fig. 5d). After

freezing the MOF/cellulose gel at a temperature below the freezing point of the liquid medium (usually water), the MOF/cellulose aerogel results by sublimating the frozen small molecular solvent using liquid nitrogen freeze-drying techniques (Long et al., 2018; Sharma et al., 2019). The pore morphology and pore distribution within the aerogel are tunable by altering the freezing speed and temperature. The obtained cellulose/ MOF aerogel form a 3D porous network structure, which is cross-linked by physical interactions such as van der Waals forces, hydrogen bonds, electronic associations, and chain entanglements (Zhu et al., 2018). Various key factors, such as MOF/cellulose suspension concentration, cellulose to MOF ratio, and the freezing rate determine the porous structure and mechanical properties of the composites. In MOF/cellulose aerogel, MOFs particularly retain their crystallinity, porosity, and accessibility, making them suitable absorbents for water purification and other separation applications (Zhu et al., 2016).

2.4.1.5. 3D printing. 3D Printing has gained popularity in MOF/cellulose composites processing, which includes multiple steps including MOF/cellulose ink preparation, objects structure design, and precise fabrication based on a computer-driven digital model in a layer by layer manner (Fig. 5e) (Hu et al., 2020).

It offers valuable advantages, such as high reproducibility, fabrication of complex geometries, controlled pore structures, tailored directionality, low cost, time effectiveness, and up-scalability (Rossi et al., 2018; Wang, Sun, et al., 2018). The MOF/cellulose ink used for 3D printing can be prepared using the methods mentioned in Sections 2.1 and 2.2, with nanocellulose as the carrier phase for MOFs. For example, a 3D porous MOF/cellulose composite was obtained by 3D printing of a ZIF-8/TOCNF hybrid hydrogel ink, that was synthesized by *in-situ* growth of ZIF-8 on TOCNF using the one-pot synthesis method. (Sultan et al., 2019) Shear thinning properties of the hybrid hydrogel inks enable the 3D printing of porous scaffolds with high shape fidelity.

2.4.2. Cellulose scaffolds support

The use of 3D natural or artificial cellulose scaffolds as a support for MOF particles eliminates the need for additional processing after MOF growth on the cellulose materials. The MOF/cellulose composites obtained using bottom-up methods possess relatively weak mechanical properties compared to the MOF/cellulose composite based on natural 3D cellulose scaffolds, such as wood, bamboo, and corn cob (Duan, Meng, et al., 2019; Guo et al., 2019; Huang, Huang, et al., 2021; Su et al., 2019; Tu et al., 2020; Wang, Shaghaleh, et al., 2021; Zhang, Wang, Song, et al., 2021; Zhu et al., 2021). Hence, using these robust natural cellulose scaffolds as MOFs supports is a promising method. Tu et al. prepared ZIF-8/wood composites by *in-situ* growth of ZIF-8 on wood supports (Sun et al., 2021; Tu et al., 2020). The obtained composites demonstrated excellent mechanical properties with 100 MPa of compressive strength and 74 MPa of ultimate tensile stress, respectively, which surpass those obtained with state-of-the-art polymer-based MOF composites. Wood aerogels prepared by delignification of wood materials are also used as substrate to support MOFs, showing an enhanced MOF loading and large surface area (Chen, Yu, et al., 2021; Wang, Lee, et al., 2021; Wu et al., 2021). However, delignification weakens the mechanical properties of wood, resulting in mechanical properties similar to the composites prepared by artificial 3D cellulose scaffolds (Chen, Zhang, et al., 2021; Wang, Lee, et al., 2021; Wang, Yao, et al., 2019; Wu et al., 2021; Xu et al., 2019). Those artificial 3D cellulose scaffolds such as aerogels and paper are fabricated using bottom-up processing methods prior to the MOF deposition process (Li, Tian, et al., 2020; Song et al., 2020; Thunberg et al., 2021).

3. Applications of MOF/cellulose composites

Combining MOFs and cellulose substrates to produce engineered materials significantly expands the application potential. Due to their

hierarchical porosity, high permeability, low MOF powder leakage, and simple reusability, 3D porous MOF/cellulose composites have been widely investigated as adsorbents in gas and liquid phases, which are discussed in detail in Section 3.1.

In addition, flexibility, foldability, and high mechanical stability makes MOF/cellulose composites promising for other applications, such as biological applications, chemical sensing, and electric energy storage, which we present in detail in Section 3.2.

3.1. Adsorbents

3.1.1. Gas adsorption and separation

With rapid urbanization and industrialization, air pollution, such as accumulation of harmful gases or CO₂, has a severe impact on environment, climate, and human health. Therefore, developing gas separation and adsorption systems with high absorption capability and selectivity has gained remarkable attention. In particular, membrane-based flow-through devices possess great potential due to their advantages of high mass transfer efficiency, simple operation condition, low energy demand and ease of scaling (Fig. 6a) (Zhou, Kong, et al., 2019). The incorporated membranes, as the key element of the flow-through devices, are commonly made from non-sustainable petroleum-based materials and cause secondary pollution after disposal. Therefore, developing environmental-friendly gas-separation membranes is a

cutting-edge topic in both material and environmental sciences.

In this regard, cellulose-based materials have drawn considerable interest due to abundant sources, degradability, good mechanical strength and processability. In synergy with MOFs, composites with excellent absorption capacity and selectivity were developed by various techniques (Arjmandi et al., 2018; Chen et al., 2016; Dhainaut et al., 2020; Ho & Leo, 2021; Li, Li, et al., 2020; Ma, Zhang, et al., 2019a, b; Ma et al., 2018; Matsumoto & Kitaoka, 2016; Mubashir et al., 2019; Mubashir et al., 2018; Pimentel et al., 2017; Su et al., 2018; Tu et al., 2020; Valencia & Abdehamid, 2019; Yang et al., 2018; Yang et al., 2017; Zhang, Dou, et al., 2018; Zhang, Feng, et al., 2018; Zhou, Yuan, et al., 2019; Zhuang et al., 2013). Overall, cellulose derivatives such as cellulose acetate, carboxymethyl cellulose and TEMPO oxidized cellulose are most common as matrixes because of their strong affinities for metal cations. (Matsumoto & Kitaoka, 2016; Zhuang et al., 2013) Various MOFs have been incorporated into cellulose matrix systems for gas separation applications. Among them, MOF-90/TEMPO oxidized cellulose has shown the best gas separation performance for CO₂/CH₄ mixtures (Matsumoto & Kitaoka, 2016b). ZIF-90 has an aperture of diameter 0.35 nm between the kinetic diameter of CO₂ (0.33 nm) and CH₄ (0.38 nm). In addition, ZIF-90 possesses a carbonyl group which could interact non-covalently with CO₂ (Fig. 6b). Furthermore, TEMPO oxidized cellulose has a high density of carboxy groups. The carboxylates cause a strong affinity for metal cations such as transition-metal ions and form

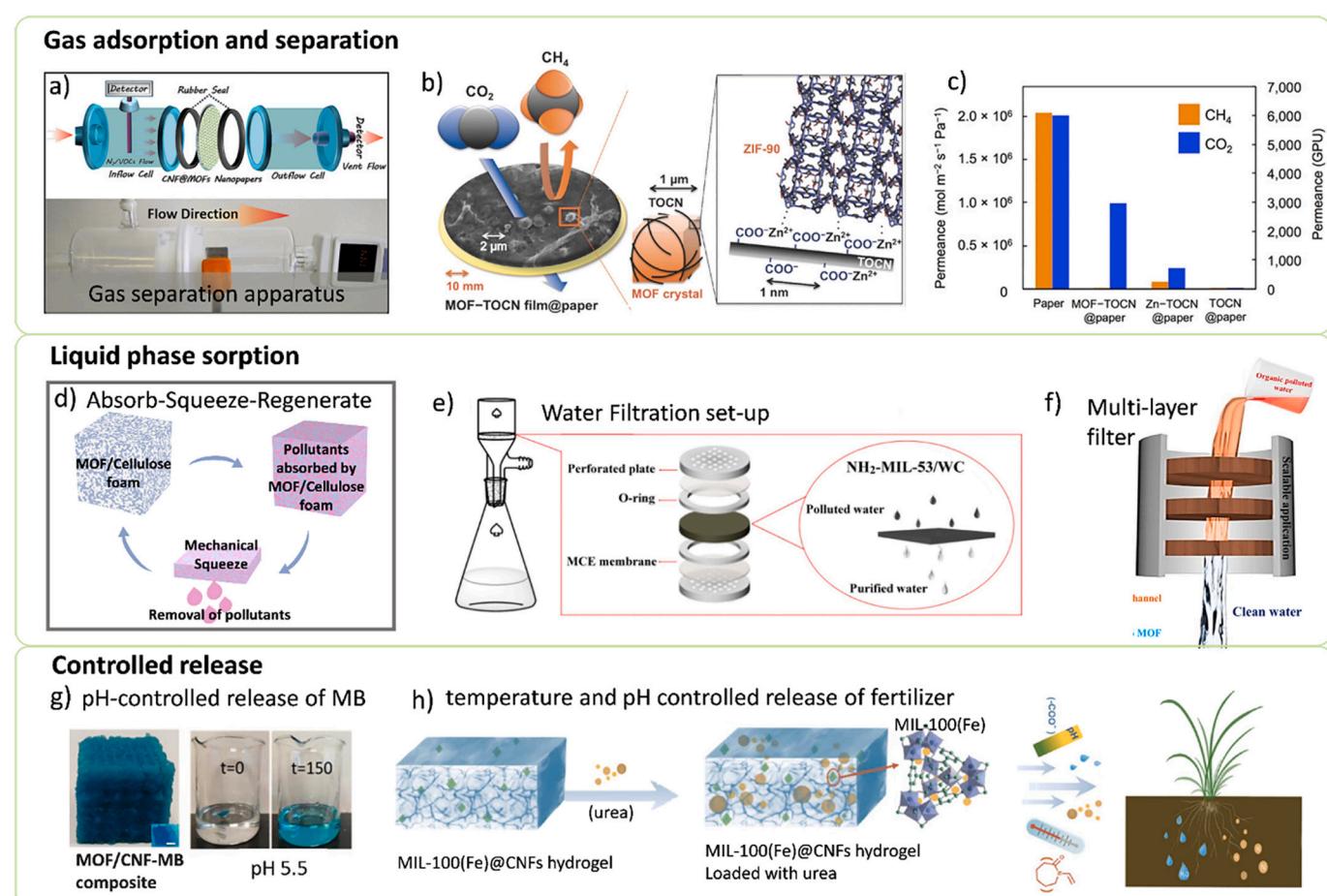


Fig. 6. Applications of MOF/cellulose composites as adsorbents a) Illustration and photograph of an apparatus for gas separation tests (Zhou, Kong, et al., 2019). b) Schematic diagram of MOF – TOCN film@paper and hierarchical structure of MOF – TOCN film@paper for CO₂ pass-through and CH₄ cut-off.c) Gas permeabilities of MOF – TOCN film@paper and CO₂/CH₄ selectivity (Matsumoto & Kitaoka, 2016). d) Illustration of the adsorption - squeeze - regeneration process of MOF/ cellulose foams for pollutant removal from water. e, f) Common waste water treatment set-up using MOF/cellulose composite membranes as filter (Gu et al., 2020; Guo et al., 2019). g) Photos of MOF/cellulose-methylene blue composite showing the release of methylene blue at different pH values (Sultan et al., 2019). h) A scheme of MIL-100(Fe)@CNFs hydrogel loading urea followed by controlled release via pH- and temperature stimuli (Lin et al., 2021). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

metal-carboxylate complexes. Therefore, the MOF-90/TEMPO oxidized cellulose showed high selectivity for separation of CO₂ from CO₂/CH₄ mixture, and exhibited great potential for designing of high-performance gas separation materials (Fig. 6c). Table 2 provides a detailed summary of MOF/cellulose composites gas separation systems and their related performance.

3.1.2. Liquid phase sorption

Water contamination has drawn serious global concerns and is a threat to both human health and environment (Schwarzenbach et al., 2010). Multiple methods have been explored for the treatment of contaminated water, including oxidation-reduction (Marinho et al., 2019; Vilar et al., 2021), precipitation (Sharma & Bhattacharya, 2017), photocatalytic degradation (Som et al., 2020; Velempini et al., 2021), adsorption (Cheng et al., 2021) etc. Among these methods, adsorption approaches possess advantages, like low cost, high removal rate, easy accessibility, as well as low secondary pollution (Ali & Gupta, 2006). Up to now, applied adsorbents, include carbon nanomaterials (Thines et al., 2017), zeolites (Wang & Peng, 2010), clays (Han et al., 2019; Srinivasan, 2011) etc. However, the practical application of these absorbents has been limited by their weak mechanical strength, low processability and non-sustainability.

In this regard, cellulose/MOF composites show immense potential for water treatment applications. The cellulose substrates equip the composites with good mechanical performance and processability. For example, by introducing MOF into cellulose foams, respective composites could achieve the absorption of pollutants and release of clean water by a simple immersing/squeezing process (Fig. 6b) (Ma, Zhang, et al., 2019a, b). Moreover, MOF/cellulose composites can also be utilized in membrane-based water filtration systems (Fig. 6e) (Gu et al., 2020). In this application scenario, the meso-pores of MOFs enable the absorption of pollutants, while the micro-pores of the cellulose substrates allow the water flux.

Recently wood/MOF composites, were developed for liquid transport applications, profiting from the unique characteristics of natural wood, a bio-based, renewable and mechanically robust material, composed of well-connected hollow fibers (Berglund & Burgert, 2018; Chen, Kuang, et al., 2020). MOF/wood composites can be directly utilized as freestanding filter in a continuous flow reactor. Guo et al. fabricated UiO-66/wood membranes by *in-situ* growth of mesoporous UiO-66 MOFs in the 3D low-tortuosity wood lumina (Guo et al., 2019). This unique structural combination improves the mass transfer of organic pollutants and increases the contact probability of organic contaminants with UiO-66 MOFs as the water flows through the

membrane, thereby improving the removal efficiency. An all-in-one filter device was designed by assembling three pieces of the UiO-66/wood membranes for large-scale organic pollutant removal. Importantly, the UiO-66/wood membrane can be readily regenerated by washing with methanol and can be up-scaled by adjusting the number of membranes (Fig. 6f).

Irrespective of the cellulosic scaffold type, the adjustable pore sizes and porosities of MOFs allow the treatment of different types of adsorbates, including dyes, heavy metal ions and organic solvents. Table 3 provides a performance overview of MOF/cellulose composites in wastewater treatment.

Besides the removal of pollutants from liquids, the controlled release of molecules could also be of the interest and there are recent studies focusing on the molecule release behavior of MOF/cellulose composites (Javanbakht et al., 2018; Sarkar et al., 2019). In a work by Sultan et al., a ZIF-8/TOCNF composite has been synthesized for a drug molecules release system. It achieved methylene blue and curcumin release in a well-controlled manner at acidic pH (5.5) (Fig. 6g) (Sultan et al., 2019). Another work by Lin et al. reported a cellulose nanofibers/MOFs hydrogel for the slow release of fertilizers, controlled by temperature and pH stimuli. (Fig. 6h) The hydrogels were composed of temperature-responsive monomer (N-vinyl caprolactam) and pH responsive CNFs. After introducing the porous MIL-100(Fe) into hydrogels, the loading and release speed of fertilizers were optimized. The MIL-100(Fe)@CNFs hydrogel significantly improved the growth of wheat due to the achieved slow release of fertilizer (Guo et al., 2021; Lin et al., 2021; Wang, Lee, et al., 2021).

3.2. Biological applications

Cellulose/MOF composites possess great potential in biological fields benefiting from their biocompatibility and biodegradability. One of the most important application in this field is their utilization as antibacterial material (Duan et al., 2018; Duan, Liu, et al., 2019; Kim, Choi, et al., 2019; Lu et al., 2018; Ma, Zhang, et al., 2019a, b; Qian et al., 2018; Rickhoff et al., 2019; Rodríguez et al., 2014; Rubin et al., 2018; Su et al., 2019; Wang et al., 2015; Yang et al., 2019). Antibacterial effects in composites mainly origin from heavy metal nanoparticles, for example Zn, Ag, and Cu nanoparticles. The antibacterial effect of MOF/cellulose composites can be a result of the MOF metal ions and/or the loading with metal nanoparticles within the MOFs.

ZIF-8, HKUST-1, and Ag-based MOFs were commonly used to prepare antibacterial composites (Duan, Meng, et al., 2019; Ma, Zhang, et al., 2019a, b; Rickhoff et al., 2019; Rodriguez et al., 2014; Rubin et al.,

Table 2
Gas separation performance of MOF/cellulose composites.

Composing materials			Permeability	Selectivity	Test conditions	Ref.	
Cellulose	MOFs	Other materials					
	Types	loading					
Cellulose acetate	NH2-MIL-53(Al)	10–20 wt%	/	CO ₂ 34.8 N ₂ 1.2 CH ₄ 1.1	CO ₂ /N ₂ 15.8 CO ₂ /CH ₄ 18.4	25 °C, 3 bar	(Mubashir et al., 2018)
Cellulose acetate	NH2-MIL-53(Al)	15 wt%	/	CO ₂ 5.15 N ₂ 0.18 CH ₄ 0.19	CO ₂ /N ₂ 12.1 CO ₂ /CH ₄ 14.7	25 °C, 3 bar	(Mubashir et al., 2019)
Cellulose acetate	MOF-5	6%–12%	/	H ₂ 6.08 CO ₂ 3.19	H ₂ /CO ₂ 1.91	25 °C, 6 bars	(Arjmandi et al., 2018)
TOCNFs	ZIF-90	44.2 wt%	/	CH ₄ 28.3 CO ₂ 290	CO ₂ /CH ₄ 123	25 °C, 2 bar	(Matsumoto & Kitaoka, 2016)
TOCNFs	UiO-66-NH ₂	24.4 wt%	/	CO ₂ 139	CO ₂ /N ₂ 46	25 °C, 2 bar	(Zhang, Feng, et al., 2018)
Carboxymethyl cellulose	ZIF-L nanosheets	30 wt%	/	H ₂ 11.8 CO ₂ 1.1 N ₂ 0.55	H ₂ /CO ₂ 10.62 H ₂ /N ₂ 21.54 CO ₂ /CH ₄ 17.87	25 °C, 1 bar	(Zhang, Feng, et al., 2018)
Pulp from Norwegian spruce	ZIF-L nanosheets	21–50 wt%	/	CH ₄ 0.062	N ₂ /CH ₄ 8.93		
Ethyl cellulose	ZIF-8	5–20 wt%	GO	no	CO ₂ /N ₂ 10.88	25 °C	(Valencia & Abdehamid, 2019)
				CO ₂ 203.3	CO ₂ /N ₂ 33.4,	25 °C, 2 bar	(Yang et al., 2018)

Table 3

Liquid-phase absorption performance of MOF/cellulose composites.

Composing materials			Adsorbate	Application and solution	Test conditions	Maximum adsorption capacity/ mg g ⁻¹	Ref.	
Cellulose	MOFs	Other materials						
	Types	loading						
Cellulose aerogel (cellulose nanocrystals + carboxymethyl cellulose)	UiO-66/MIL-100 (Fe)/ZIF-8	50 wt%	/	Cr(VI) (10 mg L ⁻¹) potassium dichromate/ benzotriazole (200 mg L ⁻¹)/ Rhodamine B (RhB) (0.355 mg L ⁻¹)	Dye capture, Remove ions and Organic pollutant absorb	25 °C, 24 h	39.06 mg g ⁻¹	(Zhu et al., 2016)
Nanocellulose	UiO-66	50 wt%	/	Methyl orange (MO)/ methylene blue (MB)	Dye capture	/	71.7 mg g ⁻¹	(Wang, Ba, et al., 2019)
Cellulose nanofiber	ZIF-8/ZIF-67/ HKUST-1	11 wt% – 81 wt%	/	Rh B (10 mg L ⁻¹ aqueous solution)	Dye capture	/	81 mg g ⁻¹	(Zhu et al., 2018)
TEMPO oxidized corncobs	HKUST-1/ZIF-8	0.09 wt%	/	MO (50 mg L ⁻¹)	Dye capture	25 °C, pH = 5	1.05 mg g ⁻¹	(Duan, Liu, et al., 2019)
Filter paper	ZIF-8/ZIF-67	13 wt% (ZIF-8), 9 wt% (ZIF-67)	/	Organic dye (An aqueous solution of the Organic dye (MO ⁻ , MB ⁺ , indigo carmine, rhodamine 6G, 5 mg L ⁻¹)	Dye capture	/	~99%	(Park & Oh, 2017)
Cellulose fibers	UiO-66-NH ₂	~5.8 wt%	/	Cr(VI)/ MO (2.5–25 mg L ⁻¹ aqueous solution)	Dye capture and Remove ions	/	78.2% for Cr(VI) and 84.5% for MO removal	(Hashem et al., 2019)
Cellulose fibers/ cellulose foams	ZIF-8	15.8–55.2 wt%	/	RhB (667 mg L ⁻¹)/ Cr (VI) (4667 mg L ⁻¹)/ DMF (1000 mg L ⁻¹)	Dye capture, Remove ions and Organic pollutant absorb	/	24.6 mg g ⁻¹ for rhodamine B, 35.6 mg g ⁻¹ for Cr (VI) and 45,200 mg g ⁻¹ for DMF	(Ma, Zhang, et al., 2019a, b)
Cotton	ZIF-67	40.7 wt%	/	MO 125 mg L ⁻¹	Dye capture	pH 4 to 10	617.4 mg g ⁻¹	(Song et al., 2020)
Cellulose microfibril	HKUST-1	/	Fe ₃ O ₄	MB (10 mg L ⁻¹)	Dye capture	/	98%	(Lu, Zhang, Wang, et al., 2019)
Regenerated cellulose	UiO-66	/	/	MB (0.0355 mg L ⁻¹)/ PEG (0.5 mg L ⁻¹)	Dye capture and Organic pollutant absorb	/	3.5 mg g ⁻¹	(Trinh et al., 2017)
Carboxymethyl cellulose	Cu-MOF	/	ibuprofen	/	Molecular release	pH 1.2–7.4	70% of release at 480 min	(Javanbakht et al., 2018)
Carboxymethyl cellulose	ZIF-8	17 wt%	Dexamethasone Hydroxyapatite	/	Molecular release	pH 7.4, 37 °C	75% release at day 28	(Sarkar et al., 2019)
Cotton	Cu ₃ (NH ₂ BTC) ₂	/	/	/	Molecular release	37 °C	90% Cu ²⁺ release at 24 h	(Rubin et al., 2018)
TOCNPs	ZIF-8	30.8%–70.1%	Curcumin	MB	Molecular release	pH 5.5–10	~40 mg g ⁻¹ curcumin release at 30 h	(Sultan et al., 2019)
Cotton fabric	Zinc-glutamate-MOF (ZnGlu)	13.96 wt%	Nitric oxide and 5-fluorouracil	/	Molecular release	pH 7.4, 37 °C	~1% NO release at 30 h, ~1% 5FU release at 75 h	(Noorian et al., 2019)
Carboxymethyl cellulose	MOF-5 (Zn-BDC)	/	/	Pb(II) (10–1000 mg L ⁻¹)	Remove ions	pH 2–6	322.58 mg g ⁻¹	(Jin et al., 2019)
Bacterial cellulose	ZIF-8	70 wt%	Polydopamine	I ₂ /KI (aqueous solution 1–10.4 × 10 ³ mg L ⁻¹)	Remove ions	pH 7, 0.24 h	1310 mg g ⁻¹	(Au-Duong & Lee, 2017)
Cellulose aerogel	ZIF-8	30 wt%	/	Cr(IV) (1–100 mg L ⁻¹)	Remove ions	120 min	41.8 mg g ⁻¹	(Bo et al., 2018)
Bacteria cellulose	ZIF-8	5 wt%	/	Pb ²⁺ / Cd ²⁺ (100 mg L ⁻¹)	Remove ions	pH 5.5, 25 °C, 50 h	390 mg g ⁻¹ for Pb ²⁺ , 220 mg g ⁻¹ for Cd ²⁺	(Ma, Zhang, et al., 2019a, b)
Cellulose acetate	ZIF-67	/	2-Methylimidazole	Cu(II) (78.9 mg L ⁻¹), Cr(VI) (88.3 mg L ⁻¹)	Remove ions	pH 6.5, 25 °C, 24 h	18.9 mg g ⁻¹ for Cu(II) and 14.5 mg g ⁻¹ Cr(VI)	(Hou et al., 2018)
Bacterial cellulose	ZIF-67	46.1 wt%	Chitosan	Cu ²⁺ and Cr ⁶⁺ (1000 mg L ⁻¹)	Remove ions	pH 6.	200.6 mg g ⁻¹ for Cu ²⁺ and 152.1 mg g ⁻¹ for Cr ⁶⁺	(Li, Tian, et al., 2020)

(continued on next page)

Table 3 (continued)

Composing materials				Adsorbate	Application and solution	Test conditions	Maximum adsorption capacity/ mg g ⁻¹	Ref.
Cellulose	MOFs	Other materials						
	Types	loading						
Cotton	UiO-66-NH ₂	/	/	Pb ²⁺ and Cu ²⁺ (100 mg L ⁻¹)	Remove ions	/	89.40 mg g ⁻¹ for Pb ²⁺ 51.33 mg g ⁻¹ for Cu ²⁺	(Lei et al., 2019)
Balsa wood Cellulose nanocrystal	NH ₂ -MIL-53	52.2 wt%	/	Pb ²⁺ (10 mg L ⁻¹)	Remove ions	pH 6	223.405 mg g ⁻¹ for Pb ²⁺	(Gu et al., 2020)
	Zn-BTC	/	Fe ₃ O ₄	Pb ²⁺ (200 mg L ⁻¹)	Remove ions	pH 2–6, 25 °C	558.66 mg g ⁻¹	(Wang et al., 2017)
Cellulose acetate	Aluminum fumarate (AlFu)	2 to 10 wt%	/	Fluoride (10 to 800 mg L ⁻¹)	Organic pollutant absorb	pH 7	179 mg g ⁻¹	(Karmakar et al., 2017)
Viscose fabrics	Cu-BTC	1.56–11 wt%	/	Phenol (2500 mg L ⁻¹ in noctane)	Organic pollutant absorb	30 °C	333 mg g ⁻¹	(Abdelhameed, Emam, et al., 2017)
Basswood	UiO-66	2.22 wt%.	/	Organic Pollutants (propranolol, 1-NA, BPA, and BPS in Water). Rh6G-polluted water (10 mg·L ⁻¹)	Organic pollutant absorb	/	690 mg·g ⁻¹ for Rh6G (based on the content of MOFs)	(Guo et al., 2019)
Basswood	MIL-101(Fe)	0.79 wt%	/	Rh6G/ propranolol/ diclofenac	Organic pollutant absorb	/	~190 mg g ⁻¹ for Rh6G, ~ 220.4 mg g ⁻¹ for propranolol, and ~178.6 mg g ⁻¹ for diclofenac	(He et al., 2020)
11 Carboxymethyl cellulose sodium	MOF-199 (HKUST-1)	70 wt%	/	Dipeptide EE/RR	Organic pollutant absorb	/	544.38 mg g ⁻¹ for EE dipeptide	(Cui et al., 2019)
	cotton fabric	UiO-66-(COOH) ₂	15.3 wt%	Creatinine (200 mg·L ⁻¹ in Tyrode buffer solution)	Organic pollutant absorb	37 °C	212.8 mg g ⁻¹	(Abdelhameed et al., 2018)
Cotton	Cu-BTC	10 wt%	/	Organophosphate pesticide (100 ppm)	Organic pollutant absorb	/	182 mg g ⁻¹	(Abdelhameed et al., 2016)
Cotton fiber	MOF-199 (HKUST-1)	/	Polyoxometalate	organophosphate pesticide (methyl parathion 20 mg L ⁻¹)	Organic pollutant absorb	25 °C	89 mg g ⁻¹ based on MOF	(Lange & Obendorf, 2015)
Carboxymethylated cellulose fibers	MOF-199	32.53 wt%	Ag NPs	4-nitrophenol (4-NP) (0.1 mM) NaBH ₄ (10 mM))	Degrad organic pollutant	/	~100% 4-NP reduction	(Duan, Liu, et al., 2019)
Viscose fabrics	Ln (Eu ³⁺ , Tb ³⁺) MOF	/	/	RhB	Degrad dye	Xe-Hg lamp	~100% RhB reduction	(Emam et al., 2018)
Cellulose aerogel	ZIF-9/ZIF-12	30 wt%	Peroxymonosulfate	p-nitrophenol (PNP) (20 mg L ⁻¹ solution)	Degrad organic pollutant	25 °C, pH 6	Remove PNP about 90% in one hour	(Ren et al., 2018)
Cellulose microfibril	HKUST-1	/	CuO/Fe ₃ O ₄	4-NP (35.5 mg L ⁻¹ 4-NP, 355 mg L ⁻¹ NaBH ₄ solution)	Degrad organic pollutant	/	~100% 4-NP reduction	(Lu, Zhang, Ma, et al., 2019)
Carboxymethyl cellulose	HKUST-1	/	Dopamine, 2-methylimidazole, and melamine	4-NP (7.1 mg L ⁻¹ 4-NP, 1420 mg L ⁻¹ NaBH ₄ aqueous solution)	Degrad organic pollutant	25 °C	~100% 4-NP reduction at 60s	(Sun et al., 2019)
CNFs	UiO-66-NH ₂	34.5 wt%	γ-Glycidoxypolytrimethoxysilane	4-Nitrophenyl Phosphate (DMNP)	Degrad organic pollutant	25 °C	90% at 20 min	(Shen et al., 2019)
CNFs	NH ₂ -MIL-88B (Fe) (NM88)	/	PAN nanofibers g-C ₃ N ₄	Sulfamethoxazole (20 mg L ⁻¹)	Degrad organic pollutant	Xenon lamp	100% at 200 min	(Qiu et al., 2019)

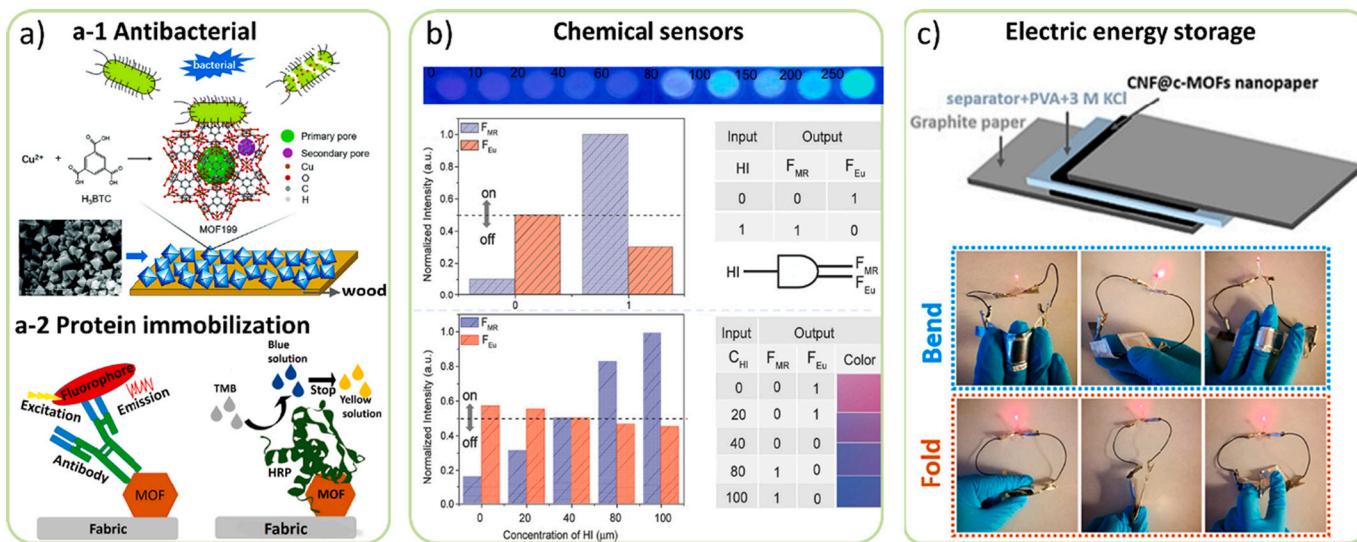


Fig. 7. a) Applications of MOF/Cellulose composite as antibacterial material and for protein immobilization. a-1) Schematics of the fabrication of MOF wood composite materials and their antibacterial mechanism (Su et al., 2019). a-2) Illustration of antibody or enzyme immobilized by MOF on fabric substrate. b) MOF/ cellulose hydrogel exhibited a color transition upon sensing histamine (HI) vapor and the truth table of the logic analytical device for HI monitoring (Xu et al., 2017). c) Photograph of the CNF@c-MOFs double layer supercapacitor device and the LED powered by devices in series under different deformations (Zhou, Kong, et al., 2019).

2018; Su et al., 2019; Wang et al., 2015). Previous works of HKUST-1/TEMPO oxidized cellulose composites reached an E. coli bacterial inactivation efficiency of up to 90% (Duan, Liu, et al., 2019). The disinfection mechanism is based on the Cu²⁺ ions of HKUST-1. The interaction of Cu²⁺ with the bacteria cell membrane via oxidation of membrane proteins and fatty acids or transmembrane potential alterations leads to rupture of membranes and cell lysis (Fig. 7a-1) (Ma, Zhang, et al., 2019a, b; Rodríguez et al., 2014; Rubin et al., 2018; Su et al., 2019).

Another intensively studied biological application is protein immobilization. The pore size and structure adjustability of MOFs make them a perfect candidate for immobilization of various proteins, while the cellulose mostly serves as the supporting substrate for the MOFs, ensuring the processability and biocompatibility of the composites. Fu et al. reported ZIF-8/fabric composites with excellent protein adsorption ability (Fu et al., 2019). The composite's porosity increased with a higher loading of ZIF-8, which favors the protein absorption. The cotton fabric provides favorable biocompatibility to maintain the bioactivity of enzyme proteins. Furthermore, results prove that carboxymethylation treatment of cellulose helps to enhance the protein immobilization ability of the cellulose/MOF composites by increasing the loading of MOF (Fig. 7a-2).

3.3. Chemical sensors

Chemical sensors are of particular interest in environmental and biological systems for the sensitive and selective detection of heavy metal ions in liquids, organic toxicants and hazardous gas analyses, which could monitor product quality, facilitate medical diagnostics and guarantee occupational safety (Giannakoudakis et al., 2017; Li et al., 2017; Li, Li, et al., 2020; Roales et al., 2017; Wang et al., 2020; Xu et al., 2017). Two kinds of chemical sensors based on photoluminescence and electrochemistry have been well developed. Both require first the adsorption of analyte molecules onto the surface of the sensors which then react to generate a signal for detection. Among the various sensing materials, MOFs, which contain electrochemically active metal sites for electrochemical sensors and photoluminescent components like inorganic clusters (especially lanthanides) or organic linkers (containing aromatic or conjugated π moieties) for photoluminescent sensors, are

promising candidates to construct these devices.

Besides, their high porosity makes them good hosts for reversible adsorption and release of guest molecules, and the tunable pore dimensions and the functional sites, including open metal sites or Lewis basic/acidic sites with different affinities towards guest molecules, also could contribute to an enhanced sensing sensitivity. Nevertheless, the relatively poor flexibility still hinders the application of single-phase MOFs as chemical sensors. Introducing MOFs particles to substrate materials is one of the solutions to this problem.

Cellulose can serve as a substrate material. The flexibility of cellulose could adapt to different using scenarios by adjusting cellulose sources and processing techniques. Depending on the cellulose source and form of substrate, the MOF/cellulose sensors could be divided into three main categories: hydrogel of the sodium salt of carboxy methyl cellulose (Xu et al., 2017), cotton textile (Xu & Yan, 2018), and cellulose paper (Li et al., 2017). For example, Xu et al. proposed an effective fluorescence sensor by dispersing methyl red@lanthanide metal-organic frameworks in water-phase sodium salt of carboxy methyl cellulose (CMC-Na). The hydrogels exhibit a color transition upon "smelling" histamine (HI) vapor. This transition and shift in the MR-based emission peak are closely related to the HI concentration. Using the HI concentration as the input signal and the two fluorescence emissions as output signals, an advanced analytical device based on a one-to-two logic gate was constructed (Fig. 7b) (Xu et al., 2017).

3.4. Electric energy storage

Because of their high electrical conductivity and large surface area, conductive metal-organic frameworks (c-MOFs) offer significant potential in electrochemical energy storage. However, because MOF crystals are brittle and insoluble, processing them into desirable nanomaterials with good flexibility and high areal capacitance is problematic. As a result, it is critical to develop effective solutions to these issues to increase the use of MOFs in electric energy storage. One intriguing answer to this problem is to embed MOFs into substrates. To date, only few studies report the fabrication of MOF on conductive substrates (e.g., graphene oxide, MoS₂ nanosheet, and polycarbonate). These approaches depend on the surface and structure of the substrates and are not suitable for the formation of all types of MOFs.

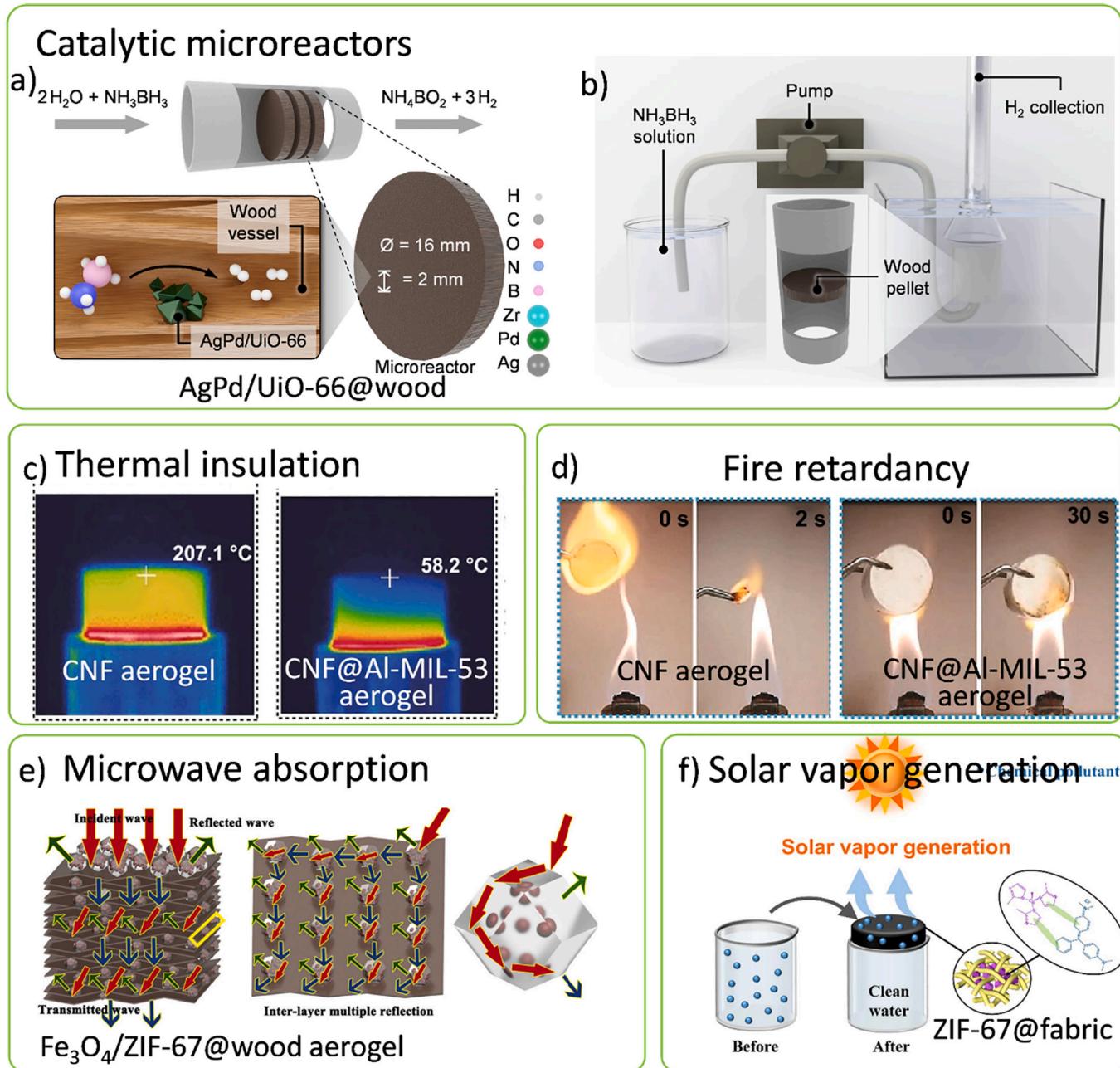


Fig. 8. a) Utilization of wood-based microreactors for hydrogen generation from ammonia borane in flow and b) the hydrogen generation setup (Tu et al., 2022). c) Infrared side-view images of the pure CNF aerogel, the pure Al-MIL-53 pellet, and the ACNF@1-MIL-53 aerogel with the temperature of the top surfaces. d) Burning tests of pure CNF aerogel and CNF@Al-MIL-53 aerogel in the flame of an alcohol lamp (Zhou et al., 2020). e) Scheme showing the reflected waves and transmitted waves rapidly decreased when the incident electromagnetic waves pass through the fabricated $\text{Fe}_3\text{O}_4/\text{ZIF-67}$ @wood aerogel (Xu et al., 2019). f) Illustration of solar vapor generation of ZIF-67@fabric composite (Li et al., 2022).

Meanwhile, MOFs' poor processability makes the fabrication of MOF-based flexible materials challenging. Cellulose is an excellent substrate for electrochemical energy storage benefiting from its chemical and physical properties. Because of its hydrophilic properties and capacity to swell in water, cellulose can also serve as an intrinsic electrolyte reservoir. The electrolyte and diffused ions in the electrolyte can be transferred to electrochemically active materials using the hierarchical porous structure generated by crosslinking cellulose fibers. Fabrication of hybrid materials based on MOFs and cellulose could combine the advantages of both materials and allow the construction of novel functional materials, given their diverse functions and flexibility (Wang, Sun, et al., 2018; Zhang et al., 2019; Zhou, Kong, et al., 2019; Zhou,

Yuan, et al., 2019; Ma, Lou, Chen, Shi, & Xu, 2019; Xu et al., 2018; Xu & Yan, 2018; Giannakoudakis et al., 2017; Ozer & Hinestroza, 2015).

Zhou et al. reported the fabrication of c-MOF nanolayers on CNFs (Zhou, Yuan, et al., 2019). The obtained hybrid nanofibers of CNF@c-MOF can be easily constructed into freestanding nanopapers with high electrical conductivity of up to 100 S cm^{-1} , hierarchical micromesoporosity, and outstanding mechanical properties. Given these advantages, the nanopapers were tested as electrodes in a flexible and foldable supercapacitor. The electrodes' high conductivity and hierarchical porous structure allow for fast charge transfer and efficient electrolyte transport. Furthermore, after 10,000 continuous charge-discharge cycles, the assembled supercapacitor demonstrates high

cycle stability with capacitance retentions of >99% (Fig. 7c). These promising results show that the potential of developing flexible energy storage devices based on sustainable cellulose and MOFs.

3.5. Other emerging applications

Beyond the above-mentioned major applications, there are also other applications gaining increasing attention, for example, catalytic micro-reactors, nanogenerator, thermal insulation, fire retardancy, microwave absorption and solar vapor generation. It demonstrates the high functionality and flexibility of cellulose/MOF composites to meet various needs and the following sections highlight these developments.

3.5.1. Catalytic microreactors

Catalytic microreactors are one of the emerging application fields of MOF/cellulose composites. They are obtained by combining the porous structure of the cellulose substrates and the catalytic properties of MOFs (Wu et al., 2021). A work by Tu et al. reported a novel MOF/wood composites microreactor for hydrogen generation (Tu et al., 2022). This native wood microreactor for continuous hydrogen generation was successfully prepared using MOF-functionalized natural wood supports to stabilize metallic palladium and silver nanoparticles (Fig. 8a, b). The inherent microchannels and mechanical strength of native wood was maintained and served as perfect substrate of the flow-through micro-reactor. The interfacial interaction between MOF and wood was improved by the amine-containing linker, which further ensured the performance and long-term stability of the MOF/wood microreactor.

3.5.2. Thermal insulation and fire retardancy

Even though MOFs are known for their high thermal stability, their usage as thermal insulation and fire retardant materials was limited by the difficulties in processing and handling. A work by Zhou et al. reported a mechanically strong and elastic CNF@Al-MIL-53 aerogel. The composite aerogel showed a relatively low thermal conductivity of ~40 mW m⁻¹ K⁻¹ and superior fire-retardancy (Fig. 8c, d). This work demonstrates that MOF/cellulose composites are promising to be used as thermal insulation and flame-retardant materials in the future (Zhou et al., 2020).

3.5.3. Microwave absorption

Materials with superior porosity are of great interest in the field of electromagnetic wave absorption. In a work by Xu et al., Fe₃O₄/ZIF-67@wood aerogel was reported to show a broad absorption bandwidth under an ultrahigh frequency. (Fig. 8e) The incident electromagnetic waves were effectively decreased because the Fe₃O₄/ZIF-67 microparticles loaded onto the three-dimensional wood aerogel cause multiple magnetic hysteresis losses. The prepared Fe₃O₄/ZIF-67@wood aerogel shows potentials for smart building and miniaturized devices applications (Xu et al., 2019).

3.5.4. Solar vapor generation

Solar vapor generation from seawater or contaminant water is a promising approach to alleviate the water crisis. A work by Li et al. reported a ZIF-67@fabric composite for simultaneous solar vapor generation and organic pollutant adsorption (Fig. 8f). The reported MOF/fabric composite achieved fast evaporation rate of 1.50 kg m⁻² h⁻¹ 1 under one sun and 99.9% contaminant removal efficiency (Li et al., 2022; Mansor et al., 2018; Wang, Yao, et al., 2019; Zhao et al., 2019).

4. Conclusion and outlook

In summary, this review highlights the remarkable advances in the development of both fabrication and application of MOF/cellulose composites. However, there are still limitations and challenges that should be addressed in the future.

First, MOF/cellulose composites still face challenges in reaching

sufficient mechanical performance at high porosity as well as high MOF loadings. The bottom-up processing methods used in preparing MOF/cellulose composites lead to sufficient porosity and controllable MOF loadings, but limited mechanical properties, which could be an obstacle for sorbents to achieve high mass transfer under big pressure. Using mechanically robust 3D cellulose scaffolds supports can partly solve this problem, still challenged by realizing high MOF loadings due to the surface area limitation of the cellulose scaffolds.

Second, it is complex to maintain high MOFs loading within cellulose scaffolds along with sufficient interaction between MOFs and cellulose, to avoid significant leaching of MOFs during usage.

Enhancing the interaction between MOFs and cellulose is critical for long-term stability of the composite. Except for the methods mentioned in this review (surface modification of cellulose), the functionalization of MOFs can also be explored. Meanwhile, it may be possible to have post-modifications using cross-linkers after preparation of MOF/cellulose composites to further enhance the interaction of MOFs with cellulose substrates. Figuring out the attachment forces by simulations and molecular modeling can indicate the inner structure of a composite and provide important design guidelines for high-performance composite synthesis in the future.

Third, each of the two main components of the composites must be further developed to improve the performance and fundamental theory of MOF/cellulose composites. From the perspective of MOFs, the MOFs used are restricted to a few well-known types which limits the application scope of the MOF/cellulose composites. More efforts should be put on developing controllable, simple, cheap, low toxic, and large-scalable MOF synthesis methods as well as improving the water, thermal, chemical stability of the MOF crystals. Various promising novel synthetic routes, *i.e.* electrochemical, mechanochemical, microwave, spray drying, and flow chemistry synthesis can be employed for MOF/cellulose composites preparation (Rubio-Martinez et al., 2017). In addition, biodegradation studies of the composites are still very limited and should be extended to achieve a closed-loop lifecycle of the MOF/cellulose composites. From the perspective of cellulose, we need to develop versatile processing methods to allow large-scale fabrication, and at the same time, maintain high porosity and robust mechanical properties.

Last but not least, new technologies and methods should be explored to characterize MOF/cellulose composites. Techniques like X-ray tomography might be useful as one can visualize macro pore arrangements as well as MOF crystals distribution within the cellulose substrates by 3D reconstructions with the aid of advanced image software. Detailed investigations with high resolution are possible with the help of synchrotron radiation facilities. In combination with simulation, the liquid and gas transport pathways within the composites can be predicted prior to applying it to real applications.

Overall, the development of MOF/cellulose composites is still in its infancy, but it shows huge potential and provides a springboard for further development.

CRediT authorship contribution statement

Kunkun Tu: Conceptualization, Writing – original draft, Writing – review & editing. **Yong Ding:** Conceptualization, Writing – original draft, Writing – review & editing. **Tobias Keplinger:** Supervision, Writing – review & editing.

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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