

Porous nanomaterials associated with the adsorption of VOCs: A Review

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ABSTRACT: Volatile organic compounds (VOCs) have attracted worldwide attention due to their serious risks to the environment and human health. Industrial processes such as the combustion of fossil fuels, petrochemicals, dyes, coatings, pesticides, and plastics all contribute to the emission of VOCs. Degradation methods (catalytic oxidation and biofiltration) and recovery methods (adsorption, compaction, and membrane separation) have been developed to remove VOCs. Adsorption has been established as one of the most promising strategies to reduce VOCs due to its cost-effective properties, simplicity, and low energy consumption. Prominent advances in the adsorption of VOCs by various types of porous materials (such as carbon-based materials, oxygen-containing materials, organic polymers, and composites) have been carefully summarized in this study. This study aims to provide a comprehensive understanding of the mechanisms of VOCs adsorption for different porous materials.

Keywords: Absorption mechanism; Porous material; VOCs refinement.

INTRODUCTION

VOCs refer to a group of organic substances characterized by their low boiling point [1]. The various definitions of VOCs are conducted by main international organizations. It can be defined as any compound of carbon, excluding carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate, which participates in atmospheric photochemical reactions, proposed by US Environmental Protection Agency (US EPA) [2]. The World Health Organization (WHO) regards VOCs as organic compounds with saturated vapor pressure over 133.322 Pa and boiling point ranging from 50 to 260 °C at atmospheric pressure [3]. The common VOCs can be

classified into several groups on the basis of their different properties. Based on the boiling point, the VOCs can be divided into very volatile organic compounds (VVOCs), VOCs, semivolatile organic compounds (SVOCs) and particulate organic matters (POMs) by WHO. For the molecular structure, the VOCs include alkanes, alkenes, aromatic hydrocarbons, alcohols, aldehydes, ketones etc. Moreover, the polar and nonpolar VOCs are distinguished according to the degree of molecular polarity [4-6]. The emission of biogenic VOCs consisting of isoprene and monoterpenes accounts for almost 90% of total global emissions [7]. With the accelerated urbanization and industrialization, the emission amount of VOCs from anthropogenic sources in

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China are predicted to be persistently increased above 5.9% annually (from 19.4 Tg in 2005 to 25.9 Tg in 2020) [8]. As shown in Fig. 2, the anthropogenic emission sources of VOCs are primarily derived from industrial process (43%), vehicle exhausts (28%), daily life (15%) and agriculture (14%). Industrial VOCs emissions are extensively involved in petroleum refinement, solvent production, use of fossil fuels, coal combustion, etc. [9-11]. Among them, the VOCs emission amount from coal combustion accounts for a large proportion of 37% in the industrial sources [12]. Benzene, toluene, ethylbenzene, and xylene (BTEX) are known as major VOCs species emitted from coal combustion, which have been all identified as hazardous air pollutants (HAPs) by the US EPA [13]. VOCs as the important precursors of ozone, photochemical pollutants and secondary organic aerosols (SOAs) pose serious harms to both the ecological environment and human health. The condensation and nucleation of OVOCS (oxygenated volatile organic compounds), SOAs and SNAs (secondary nitric aerosols) can contribute to PM 2.5 formation [14]. VOCs are also responsible for the greenhouse effect, especially methane, which is more than 20 times more potent than CO₂. Most VOCs in particular aromatic compounds and polycyclic aromatic hydrocarbons are malodorous, toxic and carcinogenic to human health even at low concentration (above 0.2 mg m⁻³), leading to respiratory inhalation and skin mucosa and damage of nervous and blood systems. In addition, some VOCs such as ethylene and propylene are widely used in the petrochemical industry. Although these compounds are less toxic and less harmful to the human body, they are flammable and explosive as the solubility reaches a certain amount [15].

Stringent regulations have been proposed to control VOCs by developed countries (US legislation calls for a 90% reduction in emissions of 189 pollutants over the next few years where VOCs occupies about 70% of those pollutants) [16]. A large number of post-processing technologies have been developed for VOCs abatement, which can be categorized into destruction technology and recovery technology. The destruction technology can decompose VOCs into CO₂, H₂O and non-toxic or less toxic compounds through different chemical or biological methods, such as thermal/

catalytic oxidation, photocatalytic oxidation, and bio-filtration and plasma catalysis. The recovery technology, termed as absorption, adsorption, condensation and membrane separation, can separate VOCs via changing the conditions of temperature and pressure in the process. Adsorption is regarded as one of the most promising VOCs treatment technologies owing to its characteristics of cost-effectiveness, flexible operation, and low energy consumption. A number of porous materials (such as carbon-based materials, oxygen-containing materials, organic polymers, composites etc.) were investigated for improving the adsorption of VOCs in terms of capacity, hydrophobic property, thermal stability and regenerability. It needs to be noted that activated carbon, zeolite and organic polymer are considered as three of the most popular adsorbents for VOCs treatment estimated by the US EPA [17-19]. The work is trying to provide a comprehensive understanding of VOCs adsorption and guidance for future research directions in this area and Porous materials for VOCs adsorption.

Carbon-based materials

Activated carbon

Activated carbon (AC) is considered as a versatile adsorbent owing to its large specific surface area (600~1400 m² g⁻¹), well-developed pore structure (0.5~1.4 cm³ g⁻¹) and high VOCs adsorption capability (10~600 mg g⁻¹). Industrial AC commonly uses carbonaceous material as the precursor such as coal, wood, coconut shell, peat and lignite, cost of which production is ranging from \$1000 to 1500 t⁻¹ [20]. It can be manufactured in form of pellet, granule, powder or sphere after the process of carbonization and activation [21]. Environmental applications of AC have been widely studied such as wastewater treatment, soil remediation and air purification, especially for VOCs disposal.

Yang et al. (2018) investigated adsorption behaviors of ACs on toluene at 25 °C, 200 ppm and N₂ atmosphere, which derived from different raw materials including wood, coal and coconut shell. The specific surface area and total pore volume ranged from 570 to 1284 m² g⁻¹, 0.25 to 0.83 cm³ g⁻¹, respectively. The adsorption capacity of these ACs ranged from 62.5 to 184.0 mg, g⁻¹ [22]. The wood-based AC with the

largest surface area and total pore volume had the maximal adsorption capacity of 184 mg, g⁻¹. Under the similar adsorption conditions except inlet toluene concentration of 2000 ppm, Li et al. (2012) found that the adsorption capacity of commercial AC were over 260 mg, g⁻¹, the surface area and total pore volume of which were 932 m² g⁻¹ and 0.432 cm³ g⁻¹ [23]. Yu et al. (2018) explored the impact of functional groups on adsorption of coconut shell based ACs at 27 °C, 500 ppm and N₂ atmosphere. The nitric acid modified AC exhibited high adsorption capacity of 433.9 mg, g⁻¹ on acetone, which might be attributed to the interaction between carboxylic groups and acetone [24]. Similarly, Zhou et al. (2018) observed magnesium oxide modified AC also presented high equilibrium amount of acetone (432.7 mg, g⁻¹) at 25 °C, 85.21 g m⁻³. It indicated that introduced oxygen functional groups offered active sites, which had strong adsorption affinity on polar acetone [25].

In summary, the adsorption performance of AC on VOCs is influenced by adsorption conditions, physicochemical properties of adsorbent. It seems that AC is suitable to be used to adsorb VOCs at room temperature, low/medium concentration and N₂ atmosphere [26]. The large surface area and pore volume of AC have positive effects on the adsorption capacity for VOCs. Specially, the chemical functional groups on the AC surface are also the key factor for certain VOCs adsorption. However, challenges of further large-scale applications of AC to industrial level still exist. Firstly, Jahandar Lashaki et al. (2016) implied that heel formation during the incomplete desorption process, affect the lifetime and regeneration cost of AC. This could be attributed to the irreversible adsorption including chemisorption, adsorbate coupling or decomposition [27]. Secondly, Jafari et al. (2018), Wang et al. (2016) and Wang et al. (2014) pointed that the flammability of AC may cause fire risk especially in the exothermic adsorption process. Moreover, high transmission resistance, pore blocking and hygroscopicity of AC also restrict its widespread application in VOCs abatement [28, 29].

Biochar

Biochar is a representative member of the carbon family, which is regarded as a potential alternative of com-

mercial AC due to its abundant feedstocks, and efficient-low cost. Compared to AC, biochar is produced in the milder pyrolysis condition under an inert atmosphere (slow pyrolysis and relatively low temperature of <700 °C) (Fig. 1) [30, 31]. Abundant carbon-rich materials, such as wood materials, agricultural and forestry residues, fruit byproducts, etc., can be used for biochar production [32]. Its production cost of \$20 t⁻¹ is much cheaper than that of AC (\$1000 to 1500 t⁻¹) [33]. The features of biochar highly depend on raw materials and production conditions. Generally, the raw materials with high lignin and mineral content tend to produce high yield of biochar, and the mineral content may decrease as the increasing pyrolysis temperature and time [34]. The biochar produced from carbonization is a disordered elementary graphitic crystallite with a rudimentary pore structure [35]. Zhang et al. (2017) evaluated 15 biochars carbonized from 5 common feedstocks for acetone, cyclohexane, and toluene adsorption at room temperature and 50 ml min⁻¹ VOCs. The specific surface area of these biochars ranged from 0.1 to 388 m² g⁻¹, and the adsorption capacity were all less than 90 mg g⁻¹. The adsorption performance of untreated biochars were supposed to have great potential to improve [36]. Physical or chemical activation are often used for biochars to develop large specific surface area and microporous structure [37]. Physical activation is conducted at high temperatures (around 700 °C) in the atmosphere of oxidizing gases such as steam, CO₂, air, or a mixture of them. The carbonization and chemical activation can be operated in a single step, where virgin biochar impregnated with activating agents are heated at temperature of 300-800 °C. The reagents frequently used are acid, alkali and metal salt [38-40]. Khan et al. (2019) developed biowaste-derived biochars with KOH activation for 2 h, which showed high adsorption capacity of 144 mg g⁻¹ on benzene. Compared with the virgin biochar, the specific surface area and total pore volume were increased from 228 to 1397 m².g⁻¹, and 0.02 to 0.51 cm³ g⁻¹ [41]. Hsi et al. (2011) prepared a series of biochars from biotreated agricultural residues for toluene adsorption via adequate 50% H₂O (g)/50% N₂ steam activation. The water molecule could react with the carbon surface to generate carbonyl and carboxyl groups during the activation process. The treated biochar was

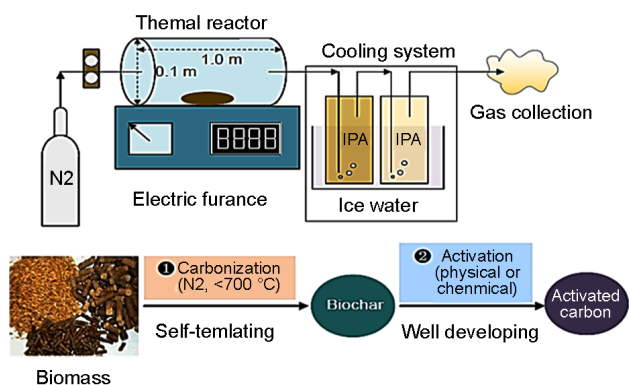


Fig. 1. The conventional carbonization and activation process of biochar [37].

with large surface area of $950\text{ m}^2\text{ g}^{-1}$ and high adsorption capacity of 227 mg g^{-1} , which was comparable to commercial AC [35]. Tham et al. (2011) studied the adsorption performance of phosphoric acid activated biochar on toluene at room temperature, 460 ppm. The results showed that high removal efficiency of 93% was obtained by impregnating acid concentration of 30%. Apart from the increased surface area ($1404\text{ m}^2\text{ g}^{-1}$), the oxygen functional groups introduced by acid also enhanced the chemical adsorption [42].

Briefly, the pore structure of untreated biochar is undeveloped, confining its VOCs adsorption capacity. The physicochemical properties of biochar can be improved a lot by physical or chemical modification. Biochar is regarded as a potential alternative to com-

mercial AC due to its abundant raw materials, effective-low cost and low energy consumption. Similar to AC, there are drawbacks of biochar include the flammability, pore blocking and hygroscopicity. Moreover, the production of biochar may cause the release of VOCs which are harmful for the environment. The in-depth research on the complicated interaction between surface groups of biochar and VOCs need to be taken far more effort.

Activated carbon fiber

Activated carbon fiber (ACF) developed in the 1960s' is in form of arranged microfilaments. It can be made into yarn, thread, fabric, felt/carpet, paper cloth and other shapes to optimize process designs for engineering use. The raw materials used in preparation of ACF are usually poor renewable such as viscose, polyacrylonitrile fibers and pitch fibers [43]. Fig. 2 illustrates a schematic of electrospinning system and characteristics of ACFs. The homogenous spinning solution contained spinnable functional material and polymer is prepared by magnetic stirring and ultrasonication, and then the nanofibers are collected on the roller under the action of the electrostatic field [44, 45]. ACF is a pure carbonaceous solid with surface area and micro-pore volume of $810\text{--}1400\text{ m}^2\text{ g}^{-1}$, and $0.36\text{--}0.92\text{ m}^3\text{ g}^{-1}$ [46]. Its pore width is usually concentrated between 0.5 and 1 nm. The commercially available ACF is very

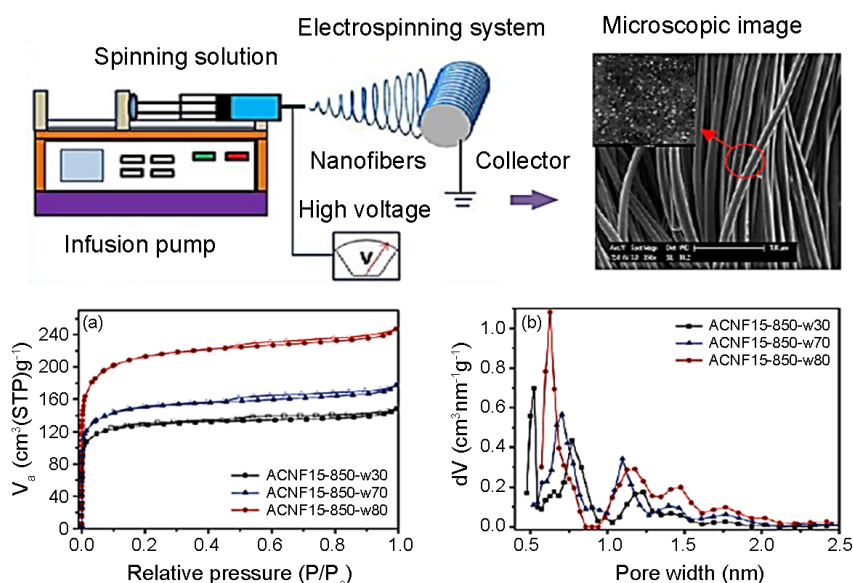


Fig. 2. Schematic of electrospun fibrous membrane and characteristics of ACFs [44, 45].

expensive due to high-cost raw materials, fiber spinning and weaving, subsequent thermal processes, as well as huge weight losses during activation. Unlike AC, ACF exhibits faster adsorption kinetics, higher mass transfer rate due to its thin-fiber shape with short and straight micropore. Furthermore, the fibrous structure can overcome the difficulty of high pressure drop, suppressed mass transfer limitations in the adsorption bed. So far, ACF is widely utilized in the fields of chemical and biochemical adsorption and separation, air and water purification, catalysts or catalyst supports, masks, medical care purposes, etc. [46].

ACF with The high micropore volume of 0.435-0.715 cm³ g⁻¹ and large surface area of 1000–2000 m² g⁻¹ was prepared by Yue et al. (2017). It had strong adsorption affinity for chloroform vapor, which adsorption capacity was 1004 mg g⁻¹ at 22 °C [47]. Similarly, Liu et al. (2019) found the activated-carbon fiber-cloth (ACFC) had the unique micropore structure with pore width ranging from 0.61 to 0.69 nm, which was superior to that of commercial AC. It exhibited high capture efficiency of isobutane (> 99%) under relative humidity of 5-80% [46]. Lin et al. (2012) reported that ACFs had less surface oxygen groups (<900 μmol g⁻¹) than AC (1000-4500 μmol g⁻¹) [48]. Besides, it was found that virgin ACFs tended to adsorb nonpolar VOCs (benzene, toluene) rather than polar VOCs (acetaldehyde, acetone) [49]. To break this limitation, Yi et al. (2008) investigated the adsorption performance of CuSO₄ modified ACF on ethanol at low concentration and 20 °C. Compared with original ACF, the adsorption capacity of modified ACF was increased from 480 to 560 mg g⁻¹[50]. In addition, Baur et al. (2015) used ACFs modified by La₂O₃, CaO, MgO, ZnO, Fe₃O₄ and Al₂O₃ to adsorb acetaldehyde at 25 °C, helium atmosphere and 1300 ppm. The adsorption capacity of La₂O₃/ACF increased from 3.2 to 20 wt. % compared to original ACF. It indicated that the modification by metal oxides improved the affinity between ACF and polar VOCs due to the introduced surface oxygen groups.

As a result, the micropore structure of ACF is superior to that of AC for VOCs adsorption, while there are few amounts of chemical functional groups on the ACF's surface. This results in the hydrophobic nature of ACF, which is beneficial for adsorbing nonpolar or

weak polar VOCs [43]. The modification technologies enable to introduce oxygen functional groups to enhance the affinity between the ACF's surface and polar VOCs [51]. It is worth to note that the limited application of ACF in practical industry ascribed to the high cost of fiber precursors and their associated processing costs [52, 53].

Graphene

Graphene with the two-dimensional (2D) structure is consist of a sheet with hexagonally arrayed carbon atoms that share sp² hybridized orbitals of one carbon with three neighbors [54]. The common preparation methods include exfoliation, hydrothermal self-assembly, chemical vapor deposition and nanotube slicing [55]. Graphene has excellent electrical conductivity ranging from 3000~5000 W mK⁻¹ (far beyond that of the copper), ultrahigh theoretical specific surface area along with great mechanical strength [56, 57]. Therefore, graphene has been applied in electronics, sensors, photonics, energy storage, biomedicine, and environment treatment owing to its outstanding physicochemical characteristics [58]. Graphene oxide (GO) and reduced graphene oxide (rGO) are the typical derivatives of graphene. The former is the product of graphene oxidation with different oxygen-containing groups such as carboxylic, hydroxyl, and epoxide groups. The latter is produced by eliminating the functional groups of GO by chemical treatment or thermal annealing [59]. Yu et al. (2018) compared the performance of benzene and toluene adsorption on GO and rGO at room temperature, 50 ppm and N₂ atmosphere. The surface areas of GO and rGO were 236.4 and 292.6 m² g⁻¹, respectively. The rGO showed higher adsorption capacities on benzene and toluene (276.4 and 304.4 mg g⁻¹) than that of GO (216.2 and 240.6 mg g⁻¹). It might be due to more hydrophobic nature, lower oxygen content and more defect sites of rGO [57]. In addition, Sun et al. (2014) synthesized MIL-101(Cr)/ GO composite, which was with large surface area of 3502 m² g⁻¹ and pore volume of 1.75 cm³ g⁻¹. Results showed that the composite had the n-hexane uptake of 1042.1 mg g⁻¹ at 25 °C, which was much higher than that of AC. The great improvement could be attributed to not only increasing specific surface area, but also stronger surface dispersive forces

of the MIL-101@GO by the introduction of the GO with dense arrays of atoms [60]. Lakshmi et al. (2018) reported magnetic graphene oxide (MGO) nanoparticles had high surface areas, nano size, high sorption performance, robust structures, magnetic nature at wide-ranging pH, and excellent chemical and thermal stabilities. MGO based materials were applied for the remediation of pollutants like metal ions, radionuclides, dyes, pesticides and opioids, which were expected to be employed for VOCs treatment.

It can be concluded that rGO exhibits strong hydrophobicity due to the removal of plentiful oxygen groups, which favor adsorbing nonpolar or weak polar VOCs [61]. Specially, MOF/GO composite seems to be a potential candidate as an efficient adsorbent for VOCs adsorption. However, the relative complicated synthesis and severe aggregation of graphene remain great challenges for its industrial applications. Therefore, replacement or removal of certain chemicals is required to be further discovered and studied to shorten the fabrication period and result in a better fabrication method [62, 63].

Carbon nanotube

Carbon nanotube (CNT) is comprised of a graphene sheet, which is rolled up in form of a cylindrical structure with sp² hybridized carbon atoms [64]. It can be divided into single-walled carbon nanotube (SWCNT) and multi-walled carbon nanotube (MWCNT) based on their arrangement of graphene cylinders [65]. It is commonly synthesized by the methods of arc discharge, laser ablation, and chemical vapor deposition (CVD). The arc discharge and laser ablation methods prefer a higher yield compared to CVD method [66]. CNT is a novel nanomaterial with unique characteristics like electrical conductivity, optical activity, and mechanical strength. In addition, the larger surface area, natural hydrophobicity as well as strong thermal stability make CNT superior to remove trace contaminants from liquid and gas phase [67, 68]. However, the utilization of CNTs for VOC abatement is quite rare. Yang et al. (2008) evaluated the adsorption behavior of CNT for indoor formaldehyde at low concentration (1.50 mg m⁻³), which adsorption capacity was 62.49 mg g⁻¹. It demonstrated that the surface of CNT exhibited good hydrophobicity and consistency, which can

maintain a strong interaction with organic compounds as promising adsorbent [69]. In order to promote the application to treat polar VOCs, Hsu et al. (2012) used the functional SWCNT oxidized by NaOCl to adsorb isopropyl alcohol vapor in air stream, and the adsorption capacity was 82 mg g⁻¹ [70]. Furthermore, modified MWCNT through covalent functionalization prepared to adsorb polar VOCs by Hussain et al. (2009). The polar functionality on the MWCNT surface dramatically altered their sorption characteristics, which prolonged the breakthrough time from 12 to 35 min for ethanol.

Although CNT is an optional VOCs adsorbents, it is worth to note that aggregation of CNT is still a challenge for its wide application. To overcome the CNT aggregation, surface oxidation and coating with surfactants is effective solutions for dispersion CNT in liquid phase [71].

Oxygen-contained materials

Zeolite

Zeolite is with crystalline aluminosilicate framework and consists of infinite three-dimensional (3D) arrangement of TO₄ tetrahedron. The two tetrahedrons share the oxygen atoms, resulting in crosslinking in space that generates channels and regular dimensions of cavities which can accommodate organic small molecules [72]. The structural formula of zeolite is A_(x/q)[(AlO₂)_x(SiO₂)_y]n(H₂O) (A: Ca, Na, K, Ba, Sr and other cations), which is containing 16-21 wt. % of water. Zeolite is widely used as chemical sieve, adsorbent and catalyst due to its excellent properties such as hydrophobicity, large surface area (250-800 m² g⁻¹), tunable porosities, and no flammability. Spe-

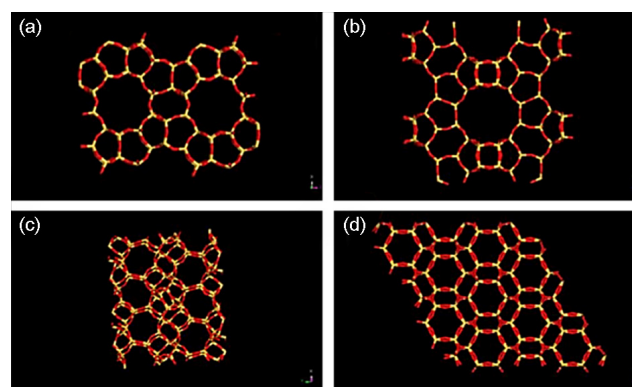


Fig. 3. Structures of the MFI-type zeosil (a), *BEA-type zeosil (b), STT-type zeosil (c) and CHA-type zeosil (d) [75].

cially, the textural properties of zeolite can be tailored by varying the Si/Al ratio [73]. Carbon-based materials often suffer from the drawbacks of flammability and regeneration difficulty. The superior hydrothermal and chemical stability of zeolite enable to overcome these problems. Besides, the temperature of complete desorption for zeolite is as low as 150 °C, while that for carbon-based materials is over 300 °C [74]. As shown in Fig. 3, zeolites including silicalite-1 (MFI-structure type), beta (*BEA-structure type), SSZ-23 (STT-structure type), and chabazite (CHA-structure type), have considerable potential as adsorbents for VOCs adsorption [75].

MFI zeolites (ZSM-5) and FAU zeolites (NaX and NaY) with different Si/Al ratios were synthesized by Kang et al. (2018). Their adsorptive removals of dichloromethane vapor at 30 °C, 5000 ppm were assessed. It showed that ZSM-5 (200) with the highest Si/Al ratio 204.5 showed the best adsorption capacity (179.2 mg g⁻¹) and was barely affected under the relative humidity of 10-90%. Zhu et al. (2017) also obtained innovative all-silica beta zeolite with excellent hydrophobicity, which surface area and total pore volume were 638 m² g⁻¹, 0.31 cm³ g⁻¹, respectively. It exhibited the adsorption capacity of 206.8 mg g⁻¹ on n-hexane at 25 °C and hydrous condition, which was little different with that at anhydrous condition [76]. Lee et al. (2011) investigated adsorption and thermal desorption of acetone and toluene vapors in dealuminated Y-zeolite bed at 20 °C, 4500 ppm and N₂ atmosphere, which surface area and total pore volume were 704 m² g⁻¹, 0.47 cm³ g⁻¹, respectively. The results suggested that Y-zeolite could be reused without a significant decrease in uptake after several regeneration cycles [77]. Nigar et al. (2015) also studied the desorption study of n-hexane (500 ppm) on NaY under microwave heating power of 150 W, which surface area and total pore volume were 750 m² g⁻¹, 0.34 cm³ g⁻¹, respectively. It found that the adsorption capacity of used NaY remained 98% of that of fresh NaY after two regeneration cycles. Consequently, the adsorption capacity of zeolite on VOCs is comparable to that of AC due to its tunable specific surface and pore structure. The Si content of zeolite is associated with its water resistance, which can be tailored in the synthesis process [78]. Zeolite is regarded one of the

conventional adsorbents for VOCs adsorption thanks to its high adsorption capacity, good thermal stability and easy reproducibility. However, the synthesis process of zeolite is complex and time-consuming. In addition, the source materials, such as tetraethyl orthosilicate and cetyltrimethyl ammonium bromide are relatively expensive compared to AC. These shortcomings would be detrimental for the widespread application of synthetic zeolites [79, 80].

Metal organic framework

Metal organic framework (MOF) firstly discovered by Hoskins et al (1989), is a novel class of crystalline hybrid porous materials [81]. Different kinds of porous MOFs are presents in Fig. 4, they are constructed from metal ions or clusters coordinated with organic ligands in ordered one, two, or three dimensional frameworks [82]. Evaporation solvent method, diffusion method, hydrothermal or solvent-thermal method, ultrasonic and microwave method can be used for the synthesis of MOF [83]. Notably, the structure of MOF can be flexibly controlled through selecting matching organic ligands. MOF has been attracted worldwide interest over the last two decades for its distinguished properties, such as ultra-high and surface area (up to 3000 m² g⁻¹), excellent thermal stability (>400°C), tailorable pore structure, and facile functionalization [84]. The great potential applications of MOFs in gas storage, separations, heterogeneous catalysis along with sensing have been widely explored [85]. The open metal sites on the pore surfaces of MOFs are available for enhancing diverse VOCs adsorption. Unlike conventional adsorbents, MOF enables to remain their permanent structure and crystalline order after regeneration [32].

Diverse types of MOFs including MIL series, UiO series along with ZIF series, have been synthesized to treat VOCs containments. Vellingiri et al. (2017) compared the different types of MOFs for toluene adsorption under ambient conditions. The equilibrated adsorption capacities of all MOFs were measured in the order of ZIF-67 (224 mg g⁻¹) > UiO-66 (166 mg.g⁻¹) > MOF-199 (159 mg.g⁻¹) > MIL-101(98.3 mg.g⁻¹). The maximum adsorption capacity of ZIF-67 might be attributed to the largest surface area of 1401 m².g⁻¹ [86]. Xian et al. (2015) also found that adsorption ca-

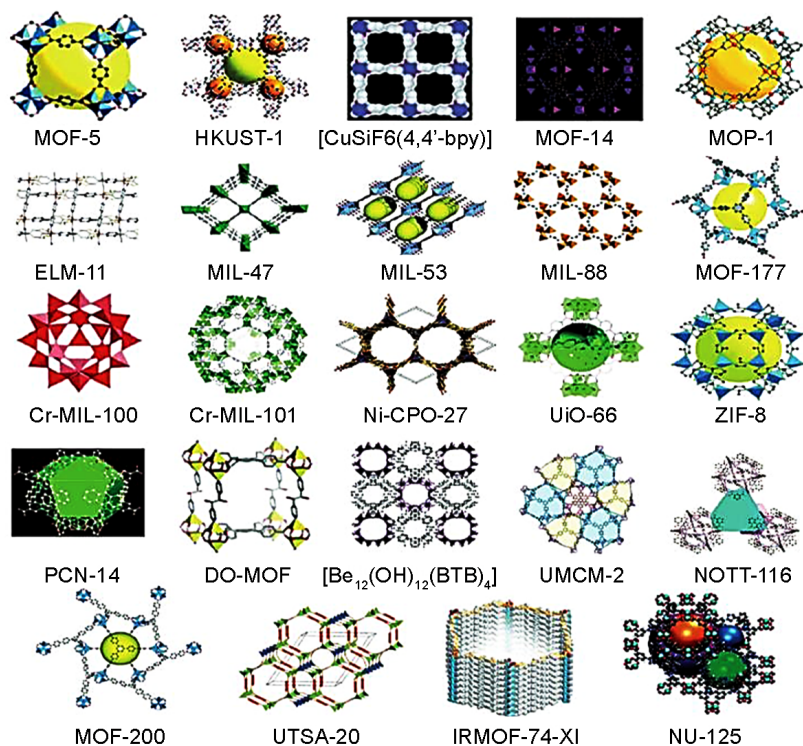


Fig. 4. Schematic diagram of different kinds of porous MOFs [82].

capacities of MIL-101 for 1,2-dichloroethane, ethyl acetate and benzene were 960.9, 510.2 and 293.7 $\text{mg}\cdot\text{g}^{-1}$, which were much higher than those of conventional adsorbents. However, the adsorption capacities of MIL-101 were significantly decreased under the humid condition due to the competitive adsorption between water molecule and VOCs. A novel enhanced hydrophobic MIL(Cr)-Z1 using naphthalene dicarboxylic acid as ligand was synthesized by Zhu et al. (2017), the surface area and total pore volume of which were 2080 $\text{m}^2\cdot\text{g}^{-1}$, 1.23 $\text{cm}^3\cdot\text{g}^{-1}$. The adsorption capacity of MIL(Cr)-Z1 on benzene at 20 °C and the relative humidity of 5, 40 and 60% were 261.7, 229.6, 205.4 $\text{mg}\cdot\text{g}^{-1}$, respectively [87]. Shafiei et al. (2018) synthesized a new modified MIL-101(Cr) by a new linker to cluster molar ratio (2:1 instead of 1:1) and different modulators (HF and HNO₃), the surface area and pore volume of which were 4293 $\text{m}^2\cdot\text{g}^{-1}$, 2.43 $\text{cm}^3\cdot\text{g}^{-1}$. The regeneration efficiency of modified MIL-101(Cr) (99.7 %) was higher than that of commercial AC (87.2 %) [88]. Kim et al. (2018) investigated the adsorption performance of amine-functionalized MOF (MIL-125-NH₂) for VOCs. The results showed that the adsorption capacities tend to follow the order of polarity

among the VOCs (p-xylene < toluene < benzene < acetone < isopropanol) due to strong interaction between amine groups and polar VOCs [89]. In short, MOF is the most promising adsorbent for VOCs adsorption due to its tunable pore structure and extraordinary physicochemical properties. Generally, the adsorption capacity of MOF on VOCs is superior to conventional adsorbents (AC and zeolite). The modification technologies can be flexibly applied for MOF to enhance the hydrophobic property and adsorptive selectivity. Nonetheless, some drawbacks are also existing to hinder its industrial application such as weak dispersive forces owing to their large amount of void space as well as the insufficient open metal sites beneficial for coordination and catalysis [90]. Furthermore, the utilization of MOF for VOCs adsorption is still an unaffordable option on account of its high preparation cost [91].

Clay

Clay is a class of water-bearing aluminosilicate minerals with layered structures, and it is the composition of rock and soil [92]. Kaolinite, montmorillonite, halloysite are three common representatives of clay minerals.

They have been pervasively applied to be adsorbents, catalysts, carriers, and templates due to their strong heat resistance and abundant raw materials. As reported, estimated deposits of Ca-bentonite alone consist of 2.5 billion tons of material in the global world. The cost (\$ 40 t⁻¹) of nature clays is much cheaper than that of AC [93, 94]. The large surface area, unique combined micro- and mesoporosity and fast mass transfer rates make clay become a potential adsorbent for VOCs abatement. Deng et al. (2017) evaluated the adsorption performance of porous clay minerals for benzene at 30 °C and N₂ atmosphere. Compared to conventional adsorbents, pristine clays had unitary micropore structure and the pore size ranged from 0.4 to 0.8 nm, the surface area and total pore volume of which ranged from 17.9 to 107.7 m² g⁻¹, 0.050 to 0.270 cm³ g⁻¹. The adsorption capacity on benzene ranged from 56.7 to 141.2 mg g⁻¹ [80]. In order to improve the textural property and adsorption capacity of clays, Wang et al. (2016) explored the effects of hydrochloric acid modification on clay minerals for toluene adsorption at 25 °C, 2000 ppm and N₂ atmosphere. The results demonstrated that the adsorption capacity of acid-activated clay on toluene increased from 44.6 to 90.4 mg g⁻¹ as the increased surface area from 228 to 329 m² g⁻¹ and introduced surface functional groups [29]. It was reported by Kimura et al. (1998) that the surface silanol groups on the surface of clay had strong hydrophilicity, which is easy to absorb water in the air [29]. Organosilanes with different head groups and chemical properties were used by Mu et al. (2018) to modify the surface properties of diatomite for methane. The surface silylation altered the surface of diatomite from hydrophilicity to hydrophobicity, which promoted the adsorption selectivity of methane under the humid condition. It can be concluded that raw clay has been proposed as an alternative adsorbent for VOCs adsorption due to low cost and desirable thermal stability. The adsorption affinity between clay and VOCs is limited due to the presence of silanol groups (Si-OH) on the clay's surface and the undeveloped pore structure. Modified methods such as acid and organic modifications are regarded as the affirmative measures to overcome the above difficulty and improve the adsorption capacity or hydrophobicity of clay [95].

Silica gel

Silica gel (SG) is an amorphous inorganic material with a 3D tetrahedral structure and silanol groups on the surface. Its molecular formula is mSiO₂·nH₂O [96]. The synthesis of SG is typically using tetramethoxysilane as the primary precursor [97]. SG has excellent thermal, mechanical, and chemical stability, low density, high microporous surface area, and plenty functional groups (such as silanols and siloxanes). As a novel porous adsorbent, the research on SG for VOCs adsorption is seldom reported. SG was employed for toluene adsorption at 25 °C and 12000 ppm by Sui et al. (2017), the surface area and total pore volume of which were 765.6 m² g⁻¹ and 0.444 cm³ g⁻¹. The results implied that SG was a suitable adsorbent for toluene adsorption with high concentration due to its rapid adsorption, high adsorption capacity (437.4 mg g⁻¹) and longer lifetime [98]. Sigot et al. (2015) compared three adsorbents of AC, zeolite and SG for the adsorption of VOSiC at 25 °C. It was found that SG was the most efficient adsorbent for VOSiC vapor, the adsorption capacity of which was 250 mg g⁻¹. However, SG often presents poor adsorption performance under humid environment due to hydrophilic silicon hydroxyl on the silica surface [99]. The modification method of coating tri-methyl-chlorosilane (TMCS) assisted with microwave irradiation certified by Huang et al., 2017 enabled to improve the hydrophobicity of SG surface effectively. Furthermore, the sufficient and in-depth researches on adsorption mechanism of SG for VOCs are imperative [100].

Organic polymer

Organic polymer is composed of light, non-metallic elements such as C, H, O, N, and B with extremely lower density than other known porous materials [101]. It can be divided into two categories: hypercrosslinked polymer (HCP) and macroporous polymer. HCP represents a novel class of predominantly microporous organic material, which is low-cost and mostly synthesized by the Friedel-Crafts alkylation reaction Fig. 5 [102]. The permanent porosity (0.5~2 nm) in HCP is attributed to extensive crosslinking reactions, which hinders the polymer chains from collapsing into a dense, nonporous state. Such highly crosslinked nature of HCP confers them high inner

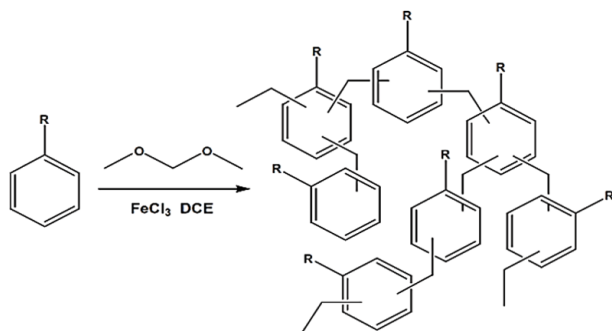


Fig. 5. Friedel-Crafts polymerization using formaldehyde dimethyl acetal [102].

specific surface area (up to 1000–1500 m²/g). While macroporous polymer is mainly with mesopore and macropore (2–50 nm) [103, 104]. Organic polymer especially HCP has garnered an increasing amount of interest for VOCs removal, which presents tailorable porosity, lightweight, strong thermal stability, and flexible regenerability. Moreover, HCP exhibits hydrophobic nature under the humid condition due to the absence of surface chemical functional groups.

Long et al. (2012) prepared a novel HCP with high surface area (1244.2 m² g⁻¹) and specific bimodal pore size distribution in the regions of micropore (0.5–2.0 nm) and meso-macropore (30–70 nm). It showed that the adsorption capacity (0.553 ml g⁻¹) on benzene at 30 °C and N₂ atmosphere were higher than that of AC (0.411 ml g⁻¹) [105]. Similarly, Zhang et al. (2012) and Wang et al. (2014) obtained HCPs with well-developed microporous and mesoporous structures for enhancing adsorption of n-hexane, dichloromethane and 2-butanone at 25 °C, the adsorption capacities of which were 0.955, 1.343 and 1.130 ml g⁻¹ [106, 107]. Wang et al. (2016) developed a novel HCP with surface area of 1345 m² g⁻¹ via one-step Friedel–Crafts reaction. The synthesized polymer presented superhydrophobic nature and excellent adsorption capacity on benzene at 25 °C and 800 ppm. The adsorption capacity of HCP on benzene was 124.2 mg g⁻¹ at 30 % relative humidity, which kept about 90% of that (137.4 mg g⁻¹) at dry condition [108]. Therefore, HCP would be a potential adsorbent for air purification and environmental protection under both dry and humid conditions due to its large surface area and superhydrophobic nature. However, complex synthesis processes may hinder its development and popularization in large-scale and real

application [108, 109].

Composite materials

The adsorbent with a single material is hard to satisfy practical needs in the complicated industrial application such as multi-component and high humidity. Composite material with hierarchically porous structure exhibits remarkably improved adsorption performance [110]. It has been applied in the fields of photoelectronics, gas storage, adsorption/separation, heterogeneous catalysis, chemical sensing, and drug delivery [110]. The preparation of hierarchical nanocomposites has drawn rise attention, and numerous studies revealed that MOF-based and zeolite-based composites are the potential adsorbents for VOCs abatement under different conditions. As mentioned in section of 2.2.2, MOF had ultrahigh surface area, tunable porosity and stable thermal property, which exhibits outstanding adsorption capacity for VOCs. However, its large amount of void space, insufficient unsaturated metallic centers as well as open framework fail to provide strong dispersive forces to capture light weighted VOCs vapors. In order to overcome these difficulties, a surface coating of a dense arrangement of atoms and a porous network is imperative. Various MOF-based composites such as MOF/carbon (MOF-C), MOF/metal oxide, MOF/silica and MOF/organic polymer have been developed as the efficient adsorbents for air purification. Notably, the addition of carbon-based materials such as GO or CNT into MOF have shown increasing surface area, dispersion force and active sites on the crystal surface. Zheng et al. (2018) reported that the adsorption capacity of MIL-101/GO on carbon tetrachloride was up to 2368.1 mg g⁻¹ at 30 °C, which increased 16% in comparison with that of pure MIL-101 (2044.4 mg g⁻¹). MIL-101/GrO composites were well above those of conventional adsorbents, such as AC (600 mg g⁻¹) and zeolite (430 mg g⁻¹) [111]. As shown in Fig. 6, Cu-BTC/GO obtained by Li et al. (2016) exhibited the maximum toluene uptake of 709.5 mg g⁻¹ at 25 °C, which had an increase of 47% in comparison with Cu-BTC [112]. More interestingly, the water-stability of Cu-BTC/GO had greatly been enhanced. After soaked in water for 10 h, it still remained original structure and porosity and its BET surface area remained 1205 m² g⁻¹. It may be as-

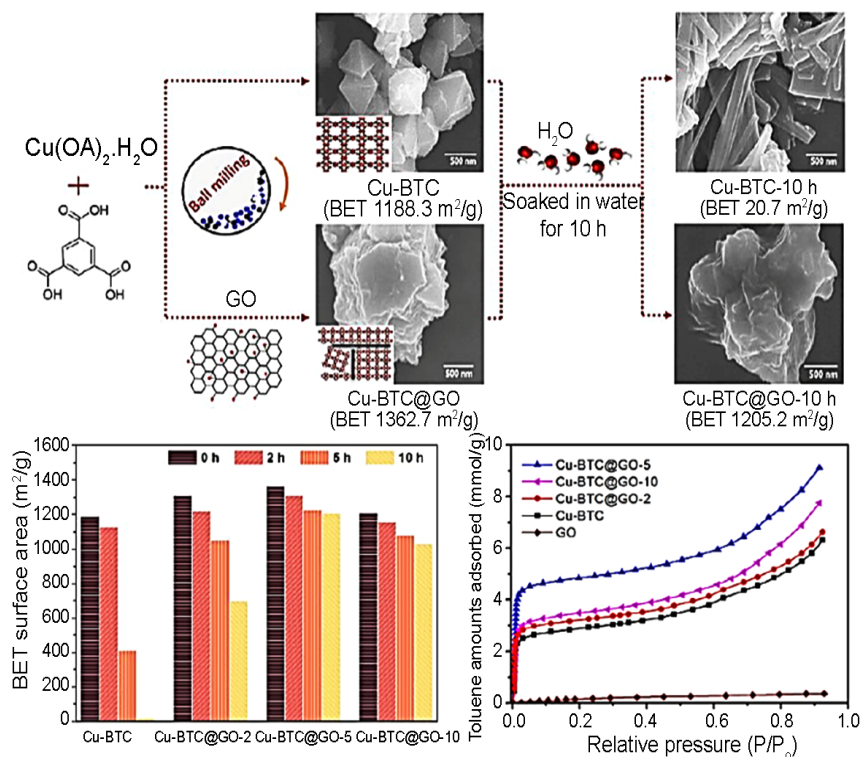


Fig. 6. Mechanochemical synthesis of Cu-BTC/GO composites performance [112].

cribed to the coordination between the oxygen groups in the GO and Cu^{2+} metal center in Cu-BTC. Furthermore, the presence of carbonaceous materials enabled to prevent these unoccupied Lewis metal sites inside MOFs from being poisoned by blocking the channels, which enhanced adsorption interactions with small VOCs molecules [112].

Zeolite is considered as the conventional adsorbent for VOCs capture due to its hydrophobicity, large surface area, controllable porosities and nonflammability. However, the drawbacks of nanoparticle agglomeration and monomodal microporosity (<1 nm) are still existing, which hinder the diffusion and mass transfer of certain macromolecules such as mesitylene, m-xylene and o-xylene [113]. Therefore, coating zeolite crystals at the surface of macroporous supports to fabricate hybrid composites is an effective strategy to overcome these difficulties. Various materials including clay minerals, MOFs, ordered mesoporous silicates, etc. have been applied as permanent supports. Among them, diatomite (Dt) is an attractive support with low cost, well-developed porosity as well as predominately mesoporous structure (50–800 nm). Prepared hierarchically porous Dt/MFI-type zeolite

composites with higher benzene adsorption capacity of 62.5 mg g^{-1} at 25°C in comparison with Dt. The Dt-coated zeolites enabled to integrate the advantages of both zeolites and supports. The resultant hierarchically porous structure (meso-/micropores or macro-/micropores) was beneficial for improving the efficiency of diffusion performance and mass transport, also synthesized Dt/silicalite composite for benzene uptake from industrial processes by a facile pre-modification in situ synthesis route.

The results showed that the composite exhibited considerably high benzene adsorption capacity (246.0 mg g^{-1}) at 25°C , which was much higher than that of Dt or silicalite. It might be due to the improved dispersity and reduced mass transfer resistance. Additionally, the pore structure could be regulated through varying the zeolite content, and the pore volume ranged from 0.051 to $0.720 \text{ cm}^3 \text{ g}^{-1}$.

Some other composite materials such as GAC/ACF, GO/CNT, SiC, etc. also have potential to be the effective adsorbents for VOCs capture. It is worth noting that the pre-synthesis or pretreatment process of the hierarchically porous structured composites may cause the extra expense [114]. Thus, the development

of simple and flexible synthesis method for composite adsorbents is still needed to be placed emphasis in future research work. The physiochemical properties and performance of different porous materials for VOCs adsorption are summarized in Table 1.

Other porous materials

Fly ash (FA) as a kind of industrial waste is mainly derived from coal-fired power plants. It contains valuable oxide components, such as SiO₂, Al₂O₃, CaO, MgO, Na₂O, and TiO₂, and essential elements, including P, K, Mg, Zn, Fe, Mn, and others. Based on its unique characteristics including honeycomb structure, func-

tional groups and unburned carbon content in the ash. Many researches have reported that FA and its derivatives (zeolite) are optional adsorbent for heavy metals removal from aqueous solutions. Ge et al. (2019) pointed that FA in the form of electrospun nanofibrous membranes had high adsorption capacity for trapping of BTX aromatic hydrocarbons and heavy metal ions [115]. Bandura et al. (2016) obtained the similar results by synthetic zeolites from fly ash for an effective trapping of xylenes, toluene and benzene. In brief, FA is a cheap, durable, easy-to-use, promising adsorbent for adsorbing some harmful substances and the research of its application on VOCs adsorption is still

Table 1. Summary of physiochemical properties and performance of different porous materials for VOC adsorption.

Adsorbent	Specific surface areas	Total pore volume	Adsorbate	Adsorption capacity mg.g ⁻¹	Adsorption Condition	References
ACs						
Coal-base AC	838	0.436	Toluene	137.3	N ₂ , 200 ppm	[22]
Coconut shell-base AC	952	0.458	Acetone	147.5	N ₂ , 25 °C, 200 ppm	[4]
Coal-base	868	0.5	Benzene	336	N ₂ , 30 °C, 400 mL/min	[120]
Coal-base	893	0.418	Chlorobenzene	51.26	5% O ₂ , 200 ppm, 300 mL/min	[121]
Biochars						
Cotton stalks	1256	0.88	Toluene	319	5% O ₂ , 25 °C, 300 ppm, 30 mL/min	[122]
Date Palm Pits	1100	-	Benzene	93.7	N ₂ , 23 ppmv, 1100 mL.min ⁻¹	[123]
ACFs						
Polyacrylonitril	1662	0.108	Toluene	538.8	Air, 30 °C, 86.5 ppm	[124]
Lignin	-	-	Acetone	106.71	N ₂ , 25 °C, 3000 mg.m ³ , 150 mL.min ⁻¹	[6]
Graphenes						
Graphite powder	292.6	-	Toluene	304.4	N ₂ , 10 °C, 50 mg.m ³ , 40 mL.min ⁻¹	[24]
Zeolites						
MCM-41	1081	1	Toluene	184	10% humidity, 100 ppm	[73]
HZSM-5	334	0.13	Acetone	1.72	N ₂ , 10 °C, 50 ppm	[125]
MOFs						
ZIF-67	1401	1.22	Toluene	224	N ₂ , 25 °C, 0.0379 bar	[86]
MIL(Cr)-101	2086	1.23	Acetone	227.3	5% humidity, 25 °C, 2 ppm	[76]
Clays						
Palygorskite	329	0.554	Toluene	90.4	N ₂ , 60 °C, 2000 ppm, 50 mL.min ⁻¹	[76]
Montmorillonite	69.5	0.119	Acetone	141.2	N ₂ , 25 °C, 3 mg.min ⁻¹	[80]
Silica gel						
Commercial SG	765.6	0.444	Toluene	437	N ₂ , 25 °C, 1200 ppm, 30 mL.min ⁻¹	[98]
Organic polymers						
Polydopamine	3291.03	1.78	Toluene	1254.95	N ₂ , 25 °C	[90]
Benzyl chloride	1345	1.75	Acetone	1394.3	N ₂ , 550 ppm, 50 mL.min ⁻¹	[108]
Styrene-divinylbenzene	1020.7	-	Chlorobenzene	389.8	30 °C	[126]
Composites						
Zeolite/GO	1112	-	Toluene	116	Air, 30 °C, 15 mL.min ⁻¹	[127]
Diatomite/silicalite-1	348.7	0.365	Acetone	246	N ₂ , 25 °C, 3 mL.min ⁻¹	[114]

lacking [116]. Sewage sludge as a potential precursor of adsorbent for wastewater treatment constitutes a paradigmatic application of the “zero-residue” concept in an anthropogenic activity. To solve its shortcoming of limited specific surface area, Anfruns et al. (2011) prepared adsorbents from pyrolysed sewage-sludge following two different methodologies, namely acid washing and activation with alkaline hydroxides for toluene, methyl ethyl ketone and limonene abatement. The adsorption performance of sludge-based adsorbents (with specific surface area up to 1000 m² g⁻¹) was comparable to commercial AC [117].

Many other porous materials such as titanate nanotube [118], carbon cryogels microsphere [79], ordered mesoporous carbon etc. [119] are envisioned to make more efforts to improve the efficiency of VOCs adsorption in the practical application.

CONCLUSIONS

The VOCs adsorption performance of various porous materials was overviewed in this work, involving the interaction mechanism and the modification methods. MOF-based composites in particularly coating the inexpensive microporous materials (biochars, clays or zeolites) would be the potential alternative to conventional single adsorbents (ACs or zeolites). The VOCs adsorption is dominated by textural properties and surface chemistry of porous adsorbents. Large specific surface area and high pore volume (in particular micropore volume) play a positive effect on physical adsorption. The chemical adsorption is associated with the surface functional groups of adsorbents as well as the polarity, boiling point, molecular size and weight of adsorbates. The relationship between the textural characteristics, chemical functional groups and VOC adsorption capacity is rarely to be quantitatively estimated. Besides, a balance between these factors needs to be taken into account to improve high adsorption capacity in some cases. The physical modification using CO₂ or steam enables to develop new micropores and regulate the pore structure of adsorbents. With regard to the above progress, the limitations and challenges in this area are carefully addressed as follows:

- 1) Low-cost and good-stability adsorbents as MOF-C

composites can be developed,

- 2) New modification method to enhance VOCs adsorption capacity and selectivity,
- 3) The hydrophobic property of porous materials can be substantially improved under high humidity condition,
- 4) How to establish the acceptable evaluation system of VOCs adsorption on porous materials,
- 5) The correct disposal of the used adsorbents and avoidance of their secondary pollution to the environment and human health;
- 6) Environmental evaluation of the VOCs adsorption process by means of life cycle assessment (LCA).

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