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REVIEW ARTICLE

Recent advances in activated carbon driven PFAS removal: structure-adsorption relationship and new adsorption mechanisms

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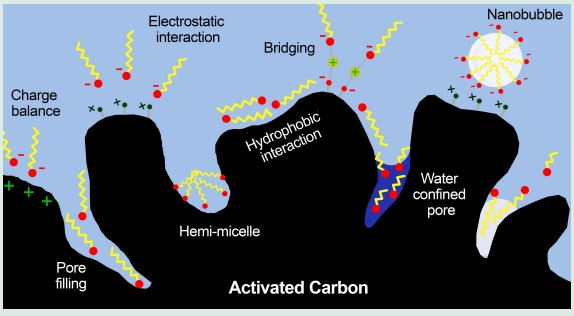
HIGHLIGHTS

- Air nanobubbles in water and AC cavities assist high PFAS removal.
- Water confining in AC's nanopores is efficient for short-chain PFAS removal.
- High adsorption force and multiple mechanisms improve adsorption selectivity.
- Amination, fluorination, or defunctionalization boosts adsorption driving forces.
- New AC modulation approaches and adsorption mechanisms are critically discussed.



ABSTRACT: Highly persistent per- and polyfluorinated alkyl substances (PFAS) have been extensively used worldwide for decades and are now ubiquitous in the ecosystem. To combat problems related to PFAS accumulation in the environment and their intrusion into the human body, PFAS adsorption and subsequent breakdown of carbon and fluorine chains are under intense research. Activated carbon (AC) is a widely used adsorbent for PFAS removal from water or wastewater. However, some of its shortcomings include inefficiency in short-chain PFAS removal, a lack of selectivity, overall low adsorption performance, and concerns regarding economic sustainability. Herein, we reviewed the recent innovative carbon-based technologies that aim to address these challenges. In particular, we focus on AC's topography engineering, defunctionalization (e.g., removing surface functional groups), hydrophobicity or surface charge modification, water-confining nanopores, and AC-nanobubbles synergy. The underlying mechanisms of these novel approaches and their effectiveness in PFAS adsorption are discussed, along with their advancements and limitations. Additionally, the PFAS adsorption and regeneration ability of high-performance ACs are presented and compared. Finally, we address current challenges and offer perspectives on advancing this technology.

KEYWORDS: PFAS, Activated carbon, Novel technologies, Adsorption mechanisms, Factors

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1 Introduction

PFAS are a group of anthropogenic chemicals containing at least one fully fluorinated methyl or methylene group without any halogen group and it excludes fully degradable subgroups (Ling, 2024). Since its invention, this material has gained significant interest from researchers, manufacturers, and consumers due to its water and oil repellency, low friction properties, high stability, and certain temperature resistance, which are attributed to its highly stable carbon-fluorine bonds (Liu et al., 2023; Evich et al., 2024). As a result, PFAS are used in a wide range of fields, including cookware, packaging, pesticides, and firefighting foam (Scheringer, 2023). The excellent performance and diverse applications of PFAS led to their mass production in the late 20th century. However, by the early 21st century, concerns began to arise regarding their safety, as these substances were frequently detected in both human bodies and the environment (Voulgaropoulos, 2022). Currently, approximately 45% of tap water in the USA contains at least one type of PFAS (Scheringer, 2023). The growing research evidences its carcinogenic, immunotoxic, genotoxic, and endocrine-disrupting to humans (Wee and Aris, 2023; Sonne et al., 2023). To mitigate the adverse effects of PFAS, there has been a push for stricter rules and regulations on the production and release of these substances. In the USA, the Environmental Protection Agency has reduced the legal limits for PFOA and PFOS in drinking water from 70 ng/L to 4 ng/L in 2024, aiming to eliminate these substances from drinking water (US EPA, 2024). Similar stringent regulations have also been implemented in many EU countries (Union E, 2020; Ateia and Scheringer, 2024). However, ensuring such a standard requires significant economic and technological efforts (Fang et al., 2024).

Currently, concentrating PFAS through adsorption followed by degradation is regarded as the most effective strategy to close the PFAS life cycle (Tan et al., 2023). The most commonly used adsorbents for removing PFAS from water include AC (Ilango and Liang, 2024), anion exchange resin (Boyer et al., 2021), nanofilter (Liu et al., 2022a), and metal-organic frameworks (MOFs). Among these, AC has been extensively studied because it is widely employed in conventional drinking water and wastewater treatment plants. As of now, over 571 articles in the Web of Science have been published on the topics of "PFAS" and "activated carbon", with approximately half of these articles being released in the last three years. AC is a highly porous carbon material produced through gas or chemical activation before, during, or after carbonization. Its extensive pore area serves as a hub for adsorbing various chemicals (Ganjoo et al., 2023; IUPAC 2025). For long-chain PFAS removal, AC is more advantageous than resins, nanofiltration, and MOFs (Liu et al., 2022b). Although resins and nanofilters are effective at removing high concentrations of PFAS, AC is more practical because of its low cost, recyclability, and ability to remove other micropollutants (Tisler et al., 2025). Jiang et al. (2024a) reported that AC is a feasible material for removing PFAS from large-scale electroplating wastewater in China.

To date, various ACs or modified carbon materials are being explored for PFAS remediation, including AC (Yao et al., 2014; Meng et al., 2019), modified AC (Deng et al., 2012b; 2013), biochar (Chen et al., 2011; Deng et al., 2015), and metal-carbon composites such as iron-based AC or biochar (Fe-AC/BC) (Xu et al., 2020). Besides, several reviews documented AC-based PFAS removal strategies. Earlier reviews address the fundamental adsorption mechanisms, influencing factors, limitations, and performances (Rahman et al., 2014; Pauletto and Bandosz, 2022). For instance, Gagliano et al. (2020) reviewed various adsorbents and their effectiveness in removing PFAS, providing insights into different adsorption processes. Sonmez Baghirzade et al. (2021) discussed the thermal regeneration processes of spent PFAS-laden AC, outlining their advantages and disadvantages. Lenka et al. (2021) demonstrated PFAS removal scenarios in actual wastewater treatment plants. Besides, some recent reviews explored the role of fluorine-fluorine interactions (Fu et al., 2024) and amine functional groups (Ateia et al., 2019). Despite extensive research on the carbon-based PFAS removal process, numerous challenges persist for practicability, including AC's low adsorption performance compared to other adsorbents, limitations in removing short-chain PFAS, difficulties with selective adsorption, and concerns about economic sustainability. Earlier reviews also provide no insights in these regards. However, recent breakthroughs have shown effectiveness in tackling these issues, which motivated us to write this review.

Here, we reviewed novel carbon-based technologies for PFAS removal, such as AC's topography engineering, surface functionalization or defunctionalization, water-confining into nanopores, amplifying hydrophobicity, and AC-nanobubbles synergy. We critically demonstrated how effectively these novel methods address the current challenges in the PFAS remediation process. In addition, we

underscored key adsorption influencing factors and considerations for efficient PFAS removal. Besides, an array of high-performance carbon materials is introduced, highlighting their PFAS adsorption and recycling capabilities. Finally, the review concludes by addressing the existing challenges and the outlook for advancing this technology.

2 Role of pristine AC properties on PFAS adsorption

2.1 Topography and particle size

Surface topography and particle size of AC play a crucial role in the diffusion and adsorption of pollutants. Micropores are considered the primary adsorption active sites, while mesopores facilitate the mass transfer process (Zeng et al., 2020). However, the suitability of these pores varies based on the size of the target pollutants. Generally, a pore size that is 2–3 times wider than the target chemical is ideal for effective adsorption (Sun et al., 2003; Ao et al., 2024). The minimum-maximum projectile diameter range of short-chain PFAS (e.g., PFBA, PFPeA, PFHxA, and PFBS) is 0.64–1.1 nm, and the medium-long chain PFAS (e.g., PFHpA, PFHxS, PFOA, and PFOS) is 0.68–1.41 nm (Cantoni et al., 2021; Shi et al., 2023). The length dimension varies depending on the chain length or PFAS head groups; for example, PFOA length is 1.20 nm, while PFOS length is 1.32 nm (Deng et al., 2015). The reported dimensions of PFAS are not standardized and may vary slightly depending on the measuring software used (Deng et al., 2015; Zhi and Liu, 2015). Considering the reported dimension of PFAS, mesopores are suitable for long-chain PFAS sliding and the formation of possible hemi-micelles into pores. Empirical studies confirmed that AC's pore size above 2 nm effectively adsorbed PFOA and PFOS (Deng et al., 2015; Xu et al., 2020). However, mesopores range from 2 to 50 nm, which is a relatively broad range. Identifying a more precise pore range for effective PFAS adsorption is challenging because chemical properties, such as adsorbent basicity, surface charge, and functional group, can overwhelm the effects of pore size. A study on ten commercial ACs reported no significant correlation between AC's topography and PFOS or PFOA adsorption (Zhi and Liu, 2015). Nevertheless, the adsorption performance significantly correlates with the adsorbents' chemical properties.

The particle size of AC influences the diffusion of PFAS into its pores. Powdered activated carbon (PAC) has shallower pores compared to granular activated carbon (GAC), allowing faster diffusion of PFAS and demonstrating faster adsorption kinetics. However, particle size does not appear to have a notable impact on overall adsorption capacity (Ateia et al., 2019). This is because the PFAS adsorption capacity on pristine AC primarily depends on the specific surface area and porosity. Although the surface area increases with smaller particle sizes, commercial AC has a fixed surface area and porosity, whereas many GACs have higher porosity and surface area than PACs. Park et al. (2020) used three different sizes of F400 and observed similar PFAS adsorption breakthrough profiles. A similar scenario is also reported in Sörengård et al. (2020) and Zhang et al. (2011) studies. The changes in particle size during AC synthesis and activation can alter the surface topography and the distribution of functional groups, which in turn can affect adsorption performance. In some instances, PAC exhibits significantly higher PFAS adsorption performance compared to granular forms, attributed to its increased surface area, porosity, and appropriate functional groups (Murray et al., 2019).

Tuning pores on AC surfaces is a straightforward process that involves activating carbon at a desired temperature. There are two primary activation methods: 1) chemical activation, which uses various chemical agents, and 2) physical activation, which employs steam or CO_2. Chemical activation creates a highly microporous surface due to the high concentration of activator used (Yao et al., 2024). However, it can be costly and less practical. In contrast, physical activation using cost-effective gases, such as steam, is more efficient and feasible for tuning micropores (Mian et al., 2023). In addition to activation methods, the precursor material also influences the porosity and surface area of AC because it is the pseudomorph of the parent material. Zhang et al. (2012) prepared AC from bituminous coal, coconut shell, and hawthorn and found that the AC from bituminous coal obtained more porosity and surface basicity. Similarly, Yeganeh et al. (2006) examined the carbon properties derived from different agricultural residues. The AC from the hard shell of apricot stones was better than other nut shells and rice hulls. Generally, commercial ACs are produced from high carbon-containing stable materials to achieve a strong end product with high yields and significant porosity. While softwood materials can provide substantial surface porosity, their yield and reliability can be inconsistent. As a result, AC derived from coal and coconut shells is among the most commonly manufactured types. Commercial ACs

typically contain 60%–80% micropores and 20%–35% mesopores (Selmi et al., 2024). This is because ACs were designed to remove various organic contaminants from wastewater and drinking water, rather than targeting a specific pollutant (e.g., PFAS).

Microporous AC endows slower PFAS adsorption and reaches saturation after 24 to 72 h. Enlarging a small fraction of micropores can amplify PFAS diffusion and remarkably increase the adsorption kinetics. Low-temperature air oxidation is a feasible method to improve mesoporosity without destructively altering the structures (Strong et al., 2023). A recent study developed mesoporous carbon from bamboo biomass through vacuum pyrolysis followed by a controlled post-steam activation (Ao et al., 2024). The resultant material doubles the initial PFAS adsorption rate while maintaining a similar adsorption capacity to commercial AC.

2.2 Surface hydrophobicity

Hydrophobicity is the most crucial characteristic influencing PFAS adsorption onto AC (Park et al., 2020; Sörengård et al., 2020). Since PFAS are amphiphilic compounds, containing a hydrophobic tail and hydrophilic head, they readily adsorb onto an apolar surface through hydrophobic interaction. Although most studies underscored the hydrophobic adsorption of PFAS onto the AC surface, the underlying mechanisms of the adsorption process were overlooked. Hydrophobic adsorption is the interaction between two non-polar molecules, or the non-polar moieties of amphiphiles, to form an anhydrous domain by assembling them in water matrices. Essentially, the destruction of surrounding water bonding via entropic effects of non-polar molecules (Xiao et al., 2020a). Molecular dynamics (MD) simulation, based on a computational technique incorporating motion forces and an explicit water environment, is a suitable means of understanding the hydrophobic interaction between PFAS and the apolar surface. In MD simulation, PFAS at a distance of 1.5 Å from an apolar surface did not spontaneously adsorb onto the surface (Jiang et al., 2021). It remained parallel upon the carbon surface or free-floating in the water matrices. Conversely, when PFOS approached the NH_2 moieties, it spontaneously bonded with the positively charged functional group, and for C-OH, it repealed (Fig. 1(a)). The dual phenomena during an apolar surface and PFOS situations suggest that the hydrophobic interaction may not occur readily. This is because the C-F chain of PFOS endows lower polarizability and surface energy than the C-H chain of the graphene surface. As a result, it not only repeals the water molecules but also most hydrocarbons. Nevertheless, hydrophobic adsorption of PFAS on AC surfaces is evident. Wang et al. (2023a) shed light on this aspect by performing the simulation several times. Initially, PFOA did not adsorb onto the apolar surface and remained free-floating, as previously noted. However, after several attempts, it ultimately adsorbed onto the surface. The water layer between PFAS and the nonpolar surface inhibits this adsorption process. Surprisingly, the water layer was disrupted by the hydrophilic head group of PFAS, not the hydrophobic tail. After breaking the water barrier, the hydrophilic head of PFAS is attached to the apolar surface while the tail ordination fluctuates. Eventually, the tail also breaks the water barrier to attach to the surface and remains affixed (Fig. 1(b)). Willemsen and Bourg (2021) also reported similar findings. The adsorption was driven by Van der Waals and hydrophobic interaction, precisely displacement of interfacial water.

The hydrophobicity on the AC surface can be rendered by pyrolyzing carbon at high temperatures, eliminating aliphatic functional groups, and increasing the degree of aromatization (Mian et al., 2023; Pala et al., 2023). Usually, the carbon activation takes place at a temperature above 900–1000 °C, at which the carbon is graphatized and forms a highly hydrophobic surface. However, the activating reagents or the gases used for pore-forming tune oxygen functional groups and endow carbon polarity (Mian et al., 2022). The oxygen functional groups do not electrostatically adsorb or form hydrogen bonds with PFAS in water; instead, they repel PFAS (Jiang et al., 2021). Zhi and Liu (2015) reported that these oxygen functional groups are the most unfavorable characteristics of AC for PFAS adsorption. Therefore, pristine AC primarily removes PFAS through hydrophobic interactions and pore-filling mechanisms. The method of amplifying hydrophobicity in AC is discussed in Section 3.1. A study examining the PFAS adsorption mechanisms of 44 carbonaceous materials concluded that hydrophobic interaction is the dominant mechanism for the removal of long-chain PFAS, while the short-chain PFAS were absorbed via electrostatic interaction (Sörengård et al., 2020). A relationship between PFAS hydrophobicity and adsorption capacity is displayed in Fig. 1(c). The pore types can also influence the hydrophobic interaction. The mesopores are more convenient for adsorbing more hydrophobic long-chain PFAS adsorption (Deng et al., 2015; Cantoni et al., 2021). This might be due to the avoidance of steric hindrance and the facilitation of multilayer adsorption and hemi-micelle formation (Liu et al., 2015; Ateia et al., 2019). Besides, long-chain

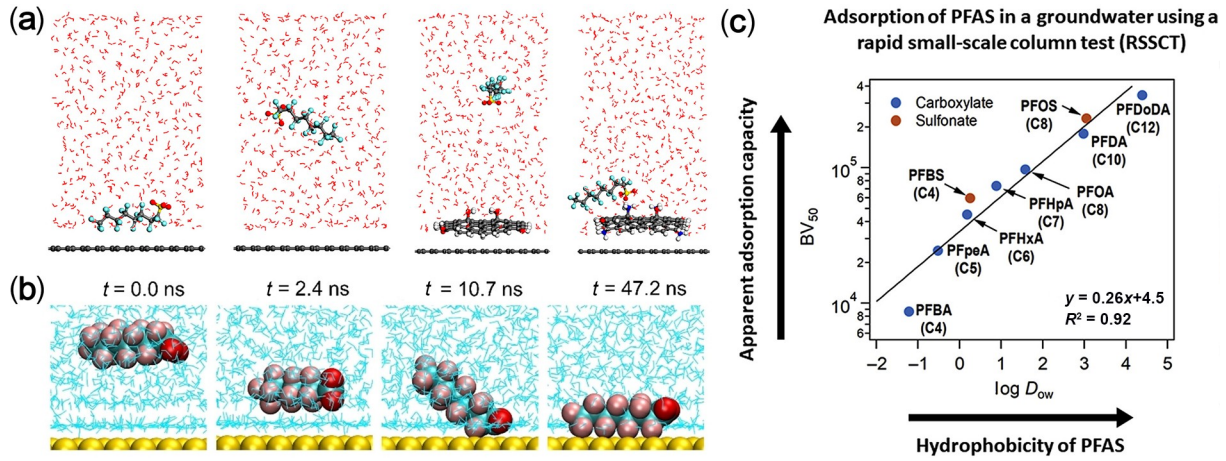

Fig. 1 MD simulation of (a) PFOS (Jiang et al., 2021); Copyright 2021, reprinted with permission from American Chemical Society, and (b) PFOA (Wang et al., 2023a); Copyright 2023, reprinted with permission from Wiley, adsorption onto apolar surface, and (c) Effects of hydrophobicity on PFAS adsorption (Park et al., 2020); Copyright 2020, reprinted with permission from Elsevier.

PFAS is more hydrophobic; their insertion and breaking of surrounding water energy barrier into narrow pores for hydrophobic adsorption might be affected.

3 AC engineering methods and new adsorption mechanism

In recent years, advances in the modification of AC and other low-cost carbonaceous materials and the incorporation of innovative technologies in AC-based PFAS removal processes have made significant progress in addressing key challenges of PFAS treatment, including improving the adsorption performance of conventional ACs, effectively adsorbing short-chain PFAS, and ensuring selectivity during PFAS adsorption in environmental water. In this section, we will critically summarize these technologies, discuss the effectiveness of adsorption, and highlight the limitations.

3.1 Surface properties modifications and adsorption selectivity

3.1.1 Nitrogen functionalized AC

Enhancing positive charges on the AC surface increases its capacity for anion exchange and electrostatic interactions. Common methods to alter the surface charge from negative or neutral to positive include introducing nitrogen functional groups or removing acidic groups (Ateia et al., 2019; Saeidi et al., 2021). For nitrogen functionalization, quaternary nitrogen-rich polymers, such as poly (dimethyl diallyl ammonium chloride) (PDADMAC) (Ramos et al., 2022; Wang et al., 2023b), cetrimonium bromide (CTAB) (Wan et al., 2022; Shaikh et al., 2023), and chitosan (Liu et al., 2022c), are integrated with AC surface through weak intermolecular aggregation (Fig. 2(a)). The long-chain polymer with enormous positive charge can extend beyond the electrical double layer of carbon surface and decorate with positive charges (Ramos et al., 2022). Quaternary ammonium in nitrogen-based polymers is considered a permanent cation and can potentially remove PFAS in a wide pH range, particularly at a strong basic pH (Shaikh et al., 2023). The N functional groups can also be grafted through pyrolyzing nitrogen precursor blended AC or biomass at the desired temperature. Usually, a high temperature is used in this process, which results in aromatic N species on the surface, such as pyridinic, pyrrolic, graphitic, or oxides nitrogen ascribed to heating temperature (Zhang et al., 2024b). The nitrogen species produced through heating method are not selective.

Nitrogen tuning on AC can change the PFAS adsorption mechanism from hydrophobic interactions to a more complex process involving anion exchange, electrostatic interactions, and hydrophobic adsorption (Fig. 2(b)). Unlike hydrophobic GACs or PACs, almost all N-rich materials, including AC or biochar, show Langmuir isotherm fitting, indicating monolayer chemosorption rather than physiosorption (Deng et al., 2013; Li et al., 2022; Ramos et al., 2022; Wan et al., 2022; Li et al., 2023; Shaikh et al., 2023). Furthermore, this approach also expands the adsorption preferences for long-chain and linear isomeric PFAS to a diverse

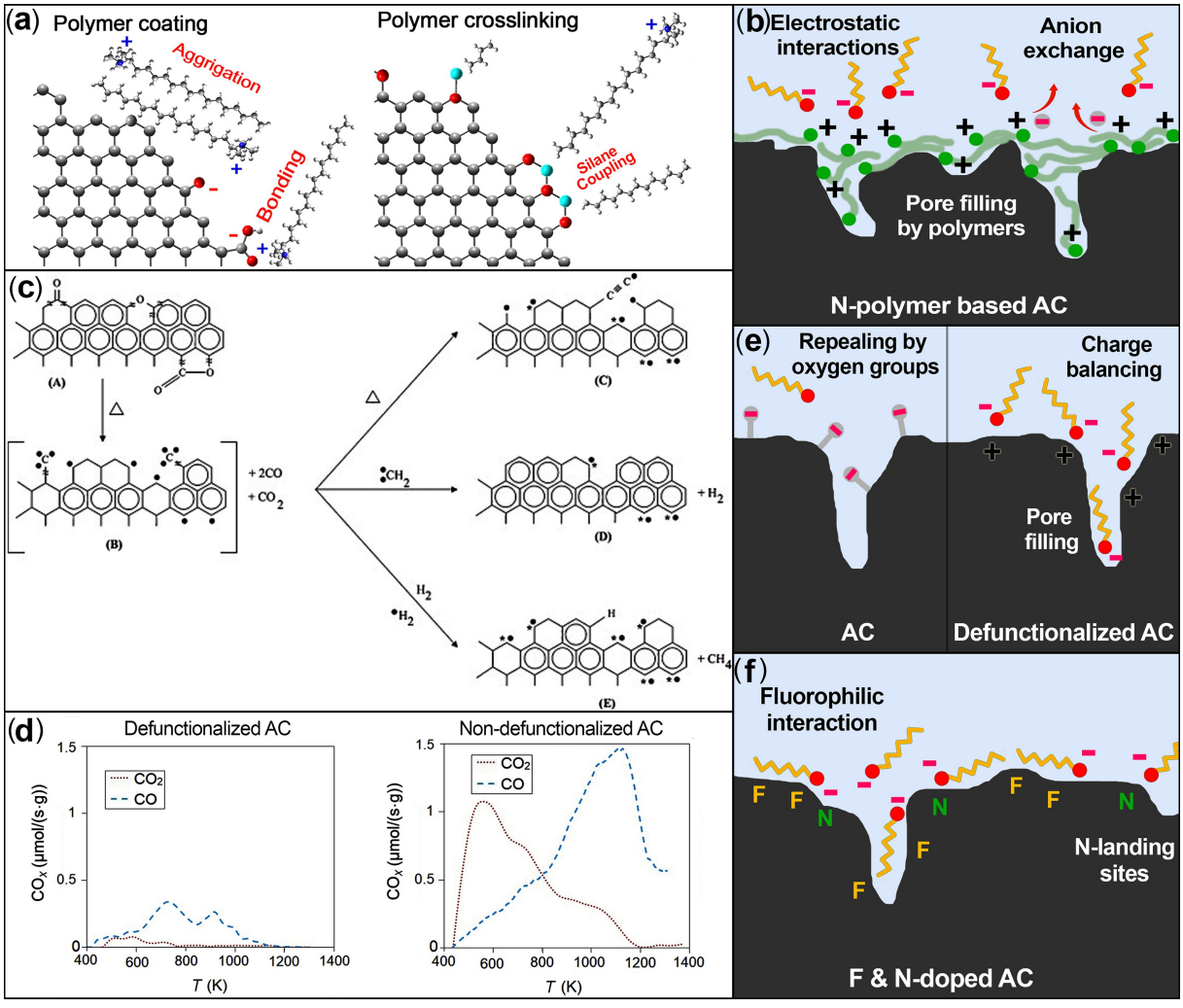

Fig. 2 (a) Nitrogen-based polymer doping process on AC surface, (b) mechanism of PFAS adsorption onto N-polymer grafted AC, (c) defunctionalization process through H_2 gas pyrolysis (Shafeeyan et al., 2010); Copyright 2010, reprinted with permission from Elsevier, (d) temperature-programmed decomposition of functional groups of defunctionalized and pristine AC (Saeidi et al., 2021); Copyright 2021, reprinted with permission from Elsevier, (e) mechanism of PFAS adsorption onto pristine and defunctionalized AC, and (f) N- and F-grafted AC.

range of PFAS, such as ether-grafted PFAS, branched PFAS, and some short-chain PFAS. The typical long-chain PFAS improved adsorption onto N-tuned AC because of the higher anion exchange capacity, while the sulfonic PFAS shows more affinity than carboxylic PFAS because of higher charge delocalization and weaker solvation property (Wang et al., 2023b; Sadia et al., 2024). The dispersion interaction favored sulfonic PFAS over carboxylic counterparts because the dispersion is related to polarizability. The atomic polarizability of SO_3 (5.31×10^{-24} cm^3) is higher than CO_2 (3.36×10^{-24} cm^3) (Wang et al., 2023b). Although N-tuned AC is efficient for long-chain linear PFAS removal, their performance in adsorbing linear short-chain PFAS removal is not remarkably high. This is because the narrow micropores significantly diminish with N-polymer coating, which is a significant short-chain PFAS adsorption site (Ramos et al., 2022; Wang et al., 2023b). However, PFAS with ether or sulfonamide groups—regardless of being short-chained—exhibit higher adsorption onto positively charged surfaces (Sadia et al., 2024). Ether oxygen atoms are shielded by nearby fluorine atoms, which restrict their hydrogen bonding capability. As a result, the ether oxygen atoms function similarly to carbon atoms, effectively increasing the carbon chain length (Liu and Sun, 2021). In addition, ether groups in PFAS change the charge distribution of the molecules. The improved charge distribution and chain length are conducive for adsorbing onto the positively charged

surface. Similarly, the brunched PFAS that shows less adsorption affinity to the hydrophobic surface due to lower hydrophobicity and high polarity enhances adsorption on positively charged surfaces for higher electrostatic interactions (McCleaf et al., 2017; Belkouteb et al., 2020).

Functionalizing ACs with nitrogen has also been identified as an effective method for enhancing the PFAS adsorption selectivity. Natural water typically contains a high amount of natural organic matter (NOM), averaging around 5.7 mg/L, notably higher than the concentration of PFAS, which is usually several hundred ng/L (Kothawala et al., 2017). Consequently, NOM—including substances like fulvic acid, humic acid, and various macromolecules—as well as ions such as Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-}, SiO_3^{2-}, CO_3^{2-}, and metal cations (M^+), can substantially reduce the PFAS adsorption capacity of AC (Inyang and Dickenson, 2017; Shaikh et al., 2023). NOM's influence on PFAS adsorption capacity can be remarkably reduced by functionalizing AC with a positively charged N functional group, particularly with a hydrophobic long-chain amine group. The process involves increasing the adsorption driving force through intensifying adsorption energy or unlocking multiple adsorption mechanisms (Jiang et al., 2024b). Although the positively charged functional group can adsorb anionic NOMs, it has more selectivity for PFAS because of multiple adsorption driving forces, such as hydrophobic adsorption and electrostatic interaction (Shaikh et al., 2023). The research on improving N-doped ACs' PFAS adsorption selectivity is in the early stages. A controlled design of N-based polymers on the AC surface is crucial for advancing this technology. A recent study on N-grafted resin stated that tuning the adsorbent surface with a long alkyl chain quaternary amino group remarkably improved adsorption selectivity due to higher non-electrostatic interaction ascribed to the optimum utilization of the hydrophobic tail of polymer by the PFAS tail (Jiang et al., 2025). To replicate such an advantage in carbon-based materials, it is essential to graft amine groups with appropriate ligands while ensuring that the hydrophobic tails of the nitrogen polymers remain accessible to the PFAS tails.

Despite the remarkable advantage in PFAS removal and adsorption selectivity by N grafting in AC, the technology suffers from the limitation of causing secondary pollution by leaching polymers (Ramos et al., 2022). Some studies claim PDADMAC irreversible adsorption on AC surface, meaning a certain degree of stability (Hsieh and Pignatello, 2018). However, a low-energy sonication can scrape off the PDADMAC as it weakly aggregates over the surface rather than chemical bonding. Silane, as a cross-linked of AC and CTAB, was used by Wan et al. (2022) to improve the stability of polymer on carbon surface (Fig. 2a). However, leaching after treatment for an extended period and recycling is also observed, highlighting the limitations of the technology for column applications.

3.1.2 Defunctionalized AC

AC has a heterogeneous surface with a combination of mixed functional groups. While positive functional groups enhance the adsorption of anionic PFAS, acidic groups repel them. Although modifying AC with nitrogen or iron increases its surface positive charges, numerous acidic functional groups may still repel PFAS or reduce the effectiveness of the modification (Saeidi et al., 2021). Research indicates that the distribution of selective positive sites on adsorbent surfaces is more favorable than a complex of positive and negative sites for PFAS electrostatic interactions or anion exchange processes (Yuan et al., 2023). Nevertheless, achieving a surface with selective positive functional groups through doping is challenging due to the presence of stable oxygen functional groups on AC surface (Chernyak et al., 2016). In this regard, defunctionalizing AC by removing the acidic groups is an effective strategy. This process increases AC's charge-balancing sites for PFAS by enhancing π–π electron-based basicity through resonating π–electrons. Furthermore, it reduces the polarity of the AC by eliminating functional groups, which amplify water repealing properties. A study compared different AC modification strategies, such as oxidation, amination, and defunctionalization, in removing PFAS (Kim et al., 2024). The topography of the AC was kept almost unchanged, while the chemical properties were altered (e.g., polarity, functional groups, surface charge, and hydrophobicity). It was concluded that AC with basic surface properties and reduced polarity was most effective for multilayer adsorption of both long-chain and short-chain PFAS.

Typical oxygen functional groups on AC are C–O groups (e.g., hydroxyls, ether, epoxy) and C=O groups (e.g., carboxyl, carbonyl, lactone, and quinone). High-temperature post-pyrolysis can alter or reduce these functional groups and achieve surface basicity. At low temperatures, dehydration alters the hydroxyl and carboxyl groups to pyrones, lactones, and anhydrides. Lactones and anhydrides are strong acidic groups and decompose at a comparatively lower temperature, while phenol, quinoa, and carbonyls are weak acidic groups and require a high temperature to decompose (Shafeeyan et al., 2010; Chernyak et al., 2016). Inert

gas or H_2 gas can achieve AC defunctionalization at high temperatures. However, using H_2 is advantageous over inert gas because it is more effective in removing oxygen groups. In addition, H_2 gas can stabilize the carbon surface by gasifying unstable carbon atoms (Fig. 2(c)). A temperature-based decomposition of defunctionalized AC's surface functional groups showed a little release of CO_2 and CO ascribed to the negligible presence of carboxyl and pyrone or chromene-type oxygen, while the non-defunctionalized counterpart showed significant peaks for CO_2 and CO (Fig. 2(d)). The defunctionalization of AC remarkably improves the anion exchange capacity and reduces cation exchange capacity, which can perform better PFAS adsorption performance than higher point zero charged AC prepared through nitrogen modification (Saeidi et al., 2021). In addition, defunctionalizing AC is conducive to eliminating the potential of steric hindrance. In some cases, on the other hand, nitrogen-grafted AC displays decreased PFAS adsorption due to steric hindrance (Ateia et al., 2019; Saeidi et al., 2021). The defunctionalization rendered several beneficial properties on ACs' surface, including increasing hydrophobicity, tuning charge balancing sites for anions, and reducing steric hindrance, which is highly conducive to selective PFAS adsorption. Saeidi et al. (2021) reported that the NOM's influence on PFAS removal in environmental conditions can be completely avoided through AC defunctionalization. The advantages and mechanisms of PFAS adsorption onto defunctionalized AC are illustrated in Fig. 2(e).

3.1.3 Hydrophobicity amplified AC

Fluorination, silicon-doping, or coating hydrophobic polymers on pristine AC is the primary means of amplifying their hydrophobicity. Carbon surface fluorination can be achieved using either static or dynamic fluorination methods. The static fluorination method involves covalently bonding fluorine atoms to the carbon surface by treating the carbonaceous material with pure fluorine gas in a closed reactor. This process also includes thermal fluorination (Agopian et al., 2021). Because pure fluorine gas is highly reactive, carbon surface fluorination using this method can be performed at room temperature. It is worth mentioning that F_2 gas is considered hazardous, so a special protocol must be followed during fluorination with F_2 gas (Znidar et al., 2022). On the other hand, dynamic fluorination involves the formation of C–F bonds on the surface by treating the material with various fluorinating agents in chemical vapor deposition furnace reactors (Ha et al., 2022). Fluorine on the adsorbent surface is efficient in adsorbing PFAS due to the high fluorophilic interaction ascribed to the fluorine-fluorine noncovalent bond (He et al., 2022). Fluorine atoms have low attractive interatomic dispersion force and polarizability, resulting in self-aggregation and minimized contact with nonfluorine atoms. Therefore, fluorine-based adsorbents not only enhance PFAS adsorption but also ensure a high PFAS adsorption selectivity (He et al., 2024a). However, to adsorb PFAS to a fluorinated surface, it needs to be at a 1 nm distance. At this proximity, PFAS can form either trans/cis or L-shaped geometries with the adsorbent fluorocarbon, depending on the types of PFAS and fluorocarbon moieties. Both isolated and symmetric distributions of the fluorocarbon moieties can effectively remove PFAS, as they allow for head-to-head (flat) contact with the PFAS molecules (He et al., 2024b). Fluorination of carbon material can also enhance Lennard-Jones potential due to increasing van der Waals interactions, leading to more PFAS molecules settling on the surface. However, excessive fluorine atoms on carbon surface have a negative impact on PFAS adsorption because it creates very high water resistance and repeals PFAS (Fu et al., 2024). Medha et al. (2024) reported that doping of 0.8% to 3.3% fluorine on the AC surface is optimum for PFOS adsorption. Grafting nitrogen or positive functional groups can reduce the repulsion effects of highly fluorinated surfaces by tuning the landing sites for PFAS. The mechanism of PFAS adsorption onto nitrogen and fluorine-grafted AC is illustrated in Fig. 2(f).

Layering silicon-based polymers is another means of amplifying AC hydrophobicity. This can be achieved through aging hydrophobic silicon polymer with AC. In this process, polymers weakly aggregate over the AC surface without forming chemical bonds. To enable covalent bonding between the oxygen groups of the activated carbon and the organosilane, a controlled modification approach is required, such as hydrolysis at specific temperatures and pH (Dong et al., 2024). The silicon-grafted AC adsorbs PFAS through hydrogen bonding. The head of the PFAS molecule interacts with the silicon-oxygen (Si–O) groups of the layered polymer, while the tail adheres to the hydrophobic surface. Proper coating and the selection of an appropriate polymer are crucial for this modification. Huang et al. (2024) used inorganic and organic silicon-based polymers to modify AC surfaces. Although both polymers enhance hydrophobicity, inorganic silicon demonstrates significantly better PFAS adsorption compared to pristine AC and organic silicon-coated AC due to its ability to support multilayer adsorption and

semi-micelle formation.

3.2 Nanobubbles in AC's cavity and water

Microbubbles have low stability and tend to dissipate quickly, while Nanobubbles (NBs) can remain in the water for weeks or even months because of their boundary-accumulated positive ions shielding (Foudas et al., 2023). Besides, pinning NBs on a substrate can significantly enhance their stability (Favvas et al., 2021). Recent studies show that NBs in water and on AC surfaces significantly enhance PFAS adsorption by anchoring them. The energy at the air-water interface is 3–7 kcal/mol lower than that of bulk water, which promotes the migration of PFAS from the water matrices to the air-water interface (Yuan et al., 2023). Initially, the head group of the PFAS molecule approaches the NB interface, and subsequently, the hydrophobic tail enters the air interface due to hydrophobic interactions. The negatively charged sulfonic or carboxylic acid group of the PFAS head forms strong hydrogen bonds with water molecules, anchoring the PFAS at the interface (Emamian et al., 2019).

AC-NB-induced PFAS removal is highly related to the PFAS chain length. Generally, PFAS with more than six carbon atoms demonstrate better removal efficiency in this process (Meng et al., 2020). This is because short-chain PFAS do not readily accumulate on the NB surface (Fig. 3(a)). PFAS molecules on the NB surface can move in two directions: they can either migrate along the interface boundary or enter the bulk water phase (Fig. 3(b)). To leave the interface, PFAS must overcome the free energy barrier at the water-air interface. This energy is higher for long-chain PFAS (e.g., PFOS free energy barrier is 6.7 kcal/mol) and lower for short-chain PFAS (e.g., PFBS free energy barrier is 3.32 kcal/mol). The mean distance of the centers of long-chain PFAS molecules at the interface is greater than that of short-chain PFAS, resulting in higher Gibbs free energy for long-chain PFAS than short-chain ones (Fig. 3(c)). Therefore, short-chain PFAS can easily leave the NB upon applying slight external energy, while long chain PFAS remain attached. Furthermore, long-chain PFAS tend to align more homogeneously and compactly at the air-water interface due to the lower α-angle tail with the head group and higher force toward the air interface. In contrast, short-chain PFAS exhibit a more irregular arrangement (Fig. 3(d)). The more ordered arrangement

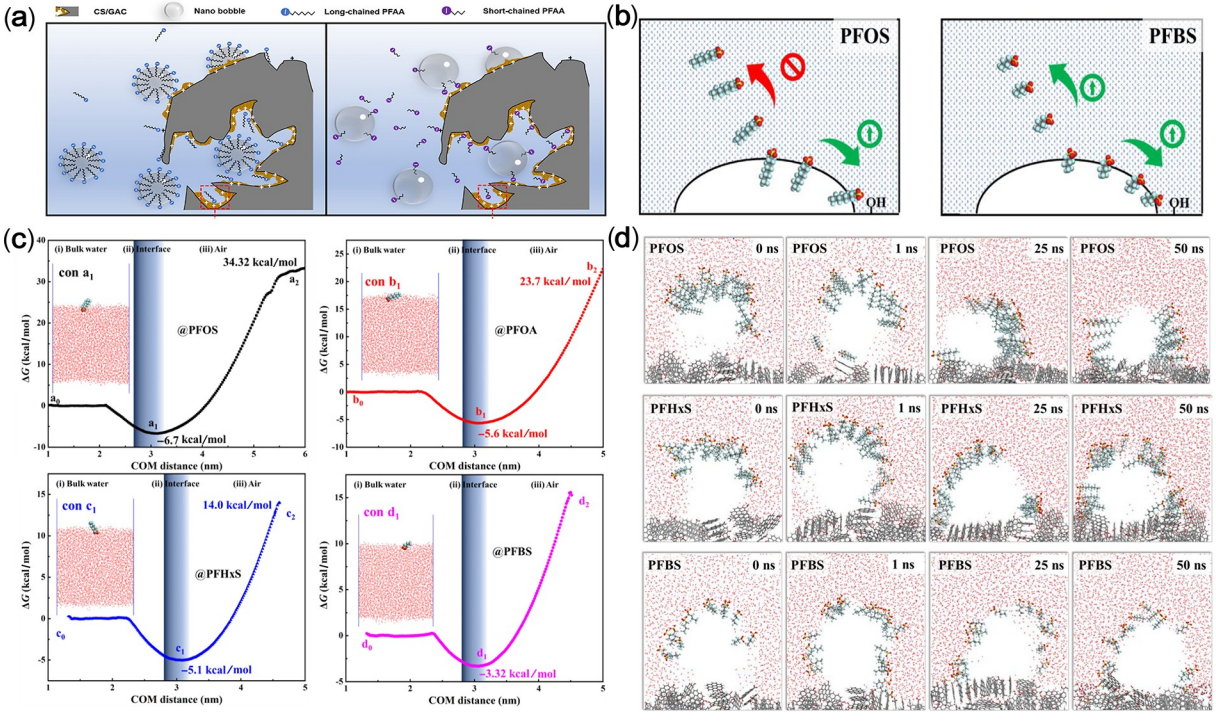

Fig. 3 (a) Long-chain and short-chain PFAS on NBs (Liu et al., 2022c); Copyright 2022, reprinted with permission from Elsevier, (b) movement of long-chain and short-chain PFAS on NB interface, (c) the center of mean distance of difference PFAS on the NB interface, (d) arrangement of different PFAS on NB interface (Yuan et al., 2023); Copyright 2023, reprinted with permission from Elsevier.

of long-chain PFAS contributes to increased stability by reducing the surface tension of the surfactants, making them more stable, charged, and conducive to adsorption on the AC surface (Yuan et al., 2023).

Both charged and hydrophobic surfaces are capable of adsorbing PFAS with the assistance of NBs. For hydrophobic surfaces or pores, NBs are initially pinned on the substrate, becoming active sites for adsorbing PFAS (Yuan et al., 2023). In contrast, positively charged surface functional groups attract negatively charged NB-PFAS colloids through electrostatic interactions. However, oxygen or negatively charged functional groups are unable to accommodate NBs due to their polarity and repealing NB-PFAS colloids (Jiang et al., 2021). The high stability and Brownian motion of NBs are beneficial for accumulating PFAS onto it and eventually adsorbing onto the AC surface (Liu et al., 2022c). Meng et al. (2017) reported an outstanding PFOS removal of 15.0 mmol/g by applying aeration in the PFAS-polyacrylonitrile fibers system, while sole aeration removes only < 0.06%. In addition, NBs are also efficient for the regeneration of PFAS-spent-AC. Meng et al. (2020) achieved 80% regeneration by removing surface and water NBs using degassing. It is worth mentioning that the AC cavity or surface and wastewater contain a small amount of NBs. Jiang et al. (2021) found that aeration improved 21%–29.2% PFAS adsorption, while degassing (water and AC) reduced it to 27.7%. Degassing only water or AC affected little, indicating the innate NBs in the water or AC's cavity performed little for PFAS adsorption and external NB supply was essential for the outstanding outcome.

3.3 Water confined nanopores

Smaller hydrophobic nanopores in AC are not filled with water due to low water pressure (e.g., < 80 MPa) in the water treatment column (Xu et al., 2011; Tinti et al., 2017). Water density inhomogeneity and vacuum clusters increase as the nanopore size decreases, with large vacuums forming at pore sizes of 0.9 to 0.6 nm (Liu et al., 2003). Water can be confined within these nanopores by altering its structure. Vapor diffusion is an effective method for confining water within AC's nanopores. The tetrameric structure of water molecules can be disrupted during vaporization, allowing the water to enter the nanopores, where it can reform into an anomalous water molecule (Fig. 4(a)). This process is not possible when treating AC with liquid water since the surface tension of the hydrophobic nanopores prevents the liquid from entering (Gao et al., 2020). When water is confined in nanopores, it exhibits unusual geometric properties related to pore size, which results in a low dielectric constant and reduced hydrogen bonds. This confinement restricts water's mobility to bulk water (Cuadrado-Collados et al., 2020). The water confined within nanopores can serve as an epicenter for the adsorption of short-chain PFAS.

Short-chain PFAS tend to remain in the water and do not easily adsorb onto hydrophobic surfaces due to their lower potential energy (Gagliano et al., 2020). In pristine AC, short-chain PFAS may adsorb through hydrophobic interactions. However, the performance of this adsorption is often poor, as water hinders the mass transfer of short-chain PFAS into the pores or onto hydrophobic surfaces. Additionally, adsorbed short-chain PFAS can be easily replaced by other pollutants or by long-chain PFAS. When water is confined into nanopores, the unique properties of the anomalous water enhance the driving forces for adsorption and facilitate the water-assisted slip effect, which allows short-chain PFAS to insert more easily into the pore structure. Understanding the adsorption behavior of PFAS in this complex environment—where water molecules can be deformed—is challenging. MD simulations can help elucidate the adsorption mechanisms involved (Cárdenas and Müller, 2021). MD simulations of confined nanopores-PFAS adsorption have revealed that the head of short-chain PFAS molecules is submerged in the confined water, while the tail remains near the pore wall. The confined water completely eliminates the mass transfer energy barrier, allowing short-chain PFAS to spontaneously adsorb onto the pores through hydrogen bonding and van der Waals interactions. Furthermore, the encapsulated water does not affect the pore wall, which reduces the surface energy of the hydrophobic wall and helps the short-chain PFAS slide along with the hydrogen bond network of the confined water (Fig. 4(b)). Interestingly, water-confined pores tend to favor the adsorption of hydrophilic PFAS over hydrophobic varieties. As a result, in a complex system, these pores effectively adsorb short-chain PFAS while minimizing the adsorption of other pollutants, ensuring high selectivity for short-chain PFAS. Currently, a highly microporous water-confined AC showed 2.2–27.9 times better short-chain PFAS removal performance than benchmark adsorbents (e.g., AC, resins, or covalent triazine-based framework) (Shi et al., 2023). The confined water in (< 0.7 nm) nanopores has a low melting point (Wang et al., 2021); thus, a low temperature can remove anomalous water and it's adsorbed PFAS. Consequently, low-temperature heating followed by alcohol washing can achieve excellent recycling performance.

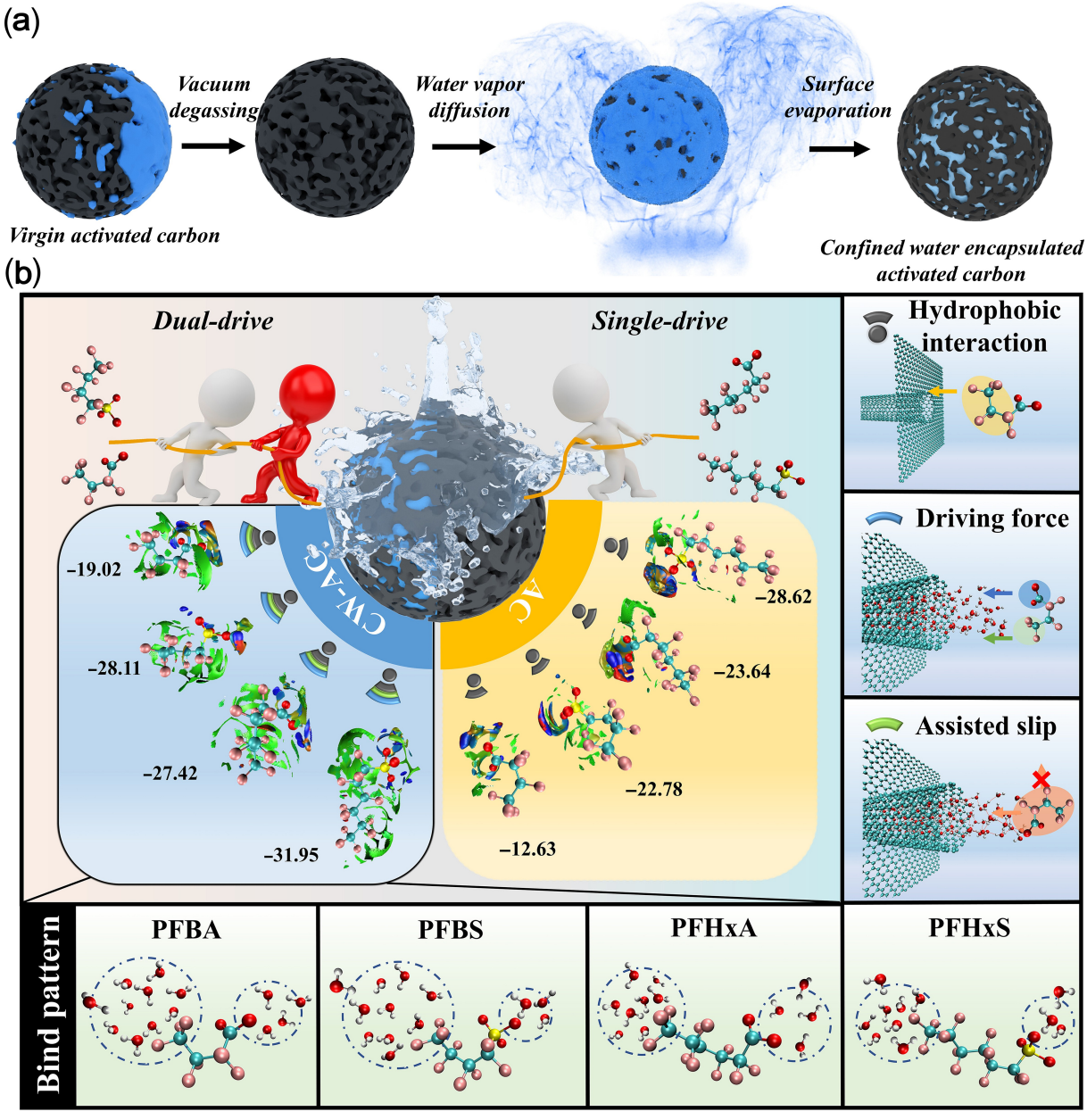

Fig. 4 (a) The vapor diffusion process of water confining into nanopores, (b) potential mean force of PFAS adsorption onto AC and confined nanopores AC, and different driving forces for adsorption into confined nanopores (Shi et al., 2023); Copyright 2023, reprinted with permission from PNAS.

4 Advances in carbon-based PFAS adsorbents

4.1 Comparative adsorption performances

The maximum adsorption capacity of AC found in batch experiments may not accurately reflect its actual performance in environments with very low concentrations of PFAS or in the presence of competing ions and NOMs (Dong et al., 2024). Nonetheless, it remains an important criterion for comparing AC with other adsorbents. The box chart shown in Fig. 5 illustrates the maximum adsorption capacity of various carbonaceous materials for PFOA, PFOS, PFBA, and PFBS. The average adsorption capacity of reported GACs is as follows: 0.32 mmol/g for PFOA, 0.62 mmol/g for PFOS, 0.12 mmol/g for PFBA, and

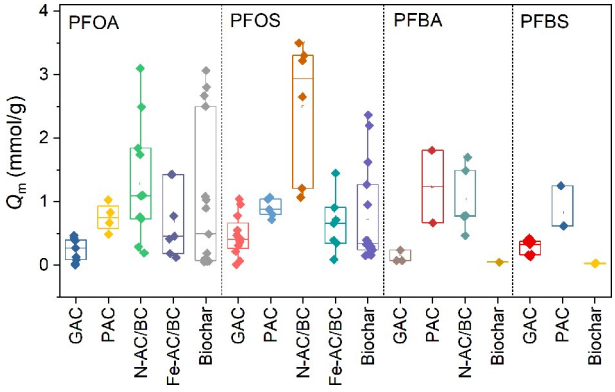

Fig. 5 The comparative performance of different ACs in removing PFAS. The chart was prepared using ACs' maximum PFAS adsorption capacities as reported in the literature (Yu et al., 2009; 2023; Carter and Farrell, 2010; Chen et al., 2011; Zhao et al., 2011; Deng et al., 2012a; 2012b; 2013; 2015; Yao et al., 2014; Wang et al., 2015; Zhang et al., 2016; Wu et al., 2018; Meng et al., 2019; Gagliano et al., 2020; Li et al., 2020; 2022; 2023; Xu et al., 2020; Zhou et al., 2021; Pauletto and Bandosz, 2022; Ramos et al., 2022; Wan et al., 2022; Yadav et al., 2022; Lei et al., 2023; Shaikh et al., 2023; Shi et al., 2023).

0.28 mmol/g for PFBS. It is noteworthy that GAC's adsorption capacity for short-chain PFAS is relatively low. The adsorption process of pristine GACs primarily involves physisorption through hydrophobic interactions and pore-filling. Thus, factors such as a larger specific surface area (SSA) and appropriate pore size are crucial for achieving high performance (Yu et al., 2023). Zhao et al. (2011) obtained 0.18 mmol/g PFOA and 0.18 mmol/g PFOS adsorption capacity for 670 m^2/g SSA of F600, while Chen et al. (2017) obtained 0.43 mmol/g PFOA and 0.78 mmol/ g PFOS adsorption capacity for 815 m^2/g SSA of GAC. The pore distribution and size also influence adsorption performances. For example, a GAC with an SSA of 712 m^2/g adsorbed 0.37 mmol/g of PFOS, while a PAC with an SSA of 812 m^2/g adsorbed 1.04 mmol/g of PFOS, which is 2.8 times higher than the GAC performance (Yu et al., 2009). On average, the PACs perform better than the GACs. The calculated mean adsorption capacity of different PACs for PFOA and PFOS are 0.72 and 0.93 mmol/g, ascribed to their high SSA and pore volume. The performance of PAC can be enhanced further by applying engineering technology. For example, confining water into PAC nanopores achieved 1.81 and 1.25 mmol/g PFBA and PFBS removal ability, which was a much greater performance than GACs and other N-grafted ACs (Shi et al., 2023).

Nitrogen-modified AC or biochar (N-AC/BC) are efficient adsorbents for PFAS, particularly for long-chain PFAS. The mean value of the reported performances of N-AC/BS is much higher than pristine GACs or PACs, such as 3.1 mmol/g for PFOA (Deng et al., 2012b), 3.3 mmol/g for PFOS (Deng et al., 2012b), 1.7 mmol/g for PFBA (Deng et al., 2013), and 0.4 mmol/g for PFBS (Li et al., 2023). While N richer polymer grafting on GAC diminishes pores and reduces SSA and porosity, N tuning over smaller particles or fiber (e.g., PACs, super fine ash, fiber, film) is beneficial because even the polymer-diminished pores, it still has high SSA and exposed active sites for PFAS adsorption (Ramos et al., 2022; Wan et al., 2022).

On the other hand, biochar is a cost-effective material for the removal of PFAS (Fan et al. 2025). It can achieve adsorption performance comparable to that of commercial AC, and some engineered biochar are designed with highly porous and functional surfaces that outperform conventional ACs. For instance, Deng et al. (2015) prepared a highly porous bamboo-derived biochar through KOH activation, resulting in a specific surface area of 2450 m^2/g and a pore volume of 0.67 cm^3/g. This biochar demonstrated a maximum adsorption capacity of 2.2 mmol/g PFOS and 1.03 mmol/g PFOA. Zhou et al. (2021) developed KOH-activated coconut shell biochar with an SSA of 1322 m^2/g, demonstrating a PFOA removal capacity of 3.06 mmol/g. While activated biochar shows impressive performance in removing long-chain PFAS, it is less effective for the removal of short-chain PFAS and the chemical activation process is a costly. As an alternative, physical activation of biomass is more efficient. Ao et al. (2024) enhanced the SSA of bamboo biochar to 1085 m^2/g through a combination of vacuum pyrolysis and steam activation, resulting in an adsorption capacity of 1.33 mmol/g for PFOS.

4.2 Regeneration processes

Desorption of PFAS for adsorbent regeneration is typically achieved in two ways: 1) by weakening the adsorption forces, allowing the PFAS to be removed from the surface, or 2) by breaking down the PFAS from the adsorbent surface using thermal energy (Jafarinejad et al., 2025). The thermal regeneration process involves breaking down the C–C, C–S, and C–F chains of PFAS and their subsequent volatilization. The energy required for decomposition primarily depends on PFAS types and partially on chain length. PFSAs require higher decomposition temperatures than PFCAs, while long-chain PFAS exhibit higher thermal stability compared to short-chain ones (Sonmez Baghirzade et al., 2021). For instance, PFOA begins to decompose at temperatures above 175 °C, while PFOS decomposition starts at 475 °C (Xiao et al., 2020b). Although the bond cleavage energy of the C–S bond

(275 kJ/mol) is lower than that of the C–C bond (348 kJ/mol), sulfonic PFAS shows higher resistance to thermal decomposition due to additional C–F bonds (two more C–F bonds in sulfonic PFAS compared to carboxylic PFAS with the same chain length) and stronger interaction with the carbon surface resulting from higher hydrophobicity (Sonmez Baghirzade et al., 2021). Besides, the head of sulfonic PFAS is densely surrounded by O groups comparing carboxylic PFAS, making it more resistant to decomposition (Turner et al., 2021). The outcome of thermal regeneration processes is also influenced by the heating conditions, such as temperature, gas, water or air environment, and the presence of catalysts. An oxidative environment decomposes PFAS more efficiently at lower temperatures compared to an inert gas environment (Sun et al., 2024). However, regenerating PFAS-laden AC in a high-temperature air environment is not advisable, as it can lead to the gasification of AC's surface carbon, resulting in pore enlargement and a reduction in yield. To lower energy consumption, hydrothermal technology was explored for PFAS-laden AC regeneration. PFAS can react with water molecules at hydrothermal conditions to eliminate HF and produce short-chain byproducts (Wang et al., 2022). However, dissolved PFAS decomposition requires comparatively higher hydrothermal temperature. Adsorption of PFAS on the AC surface and the presence of a catalyst, such as NaOH, can reduce the PFAS decomposition temperature and regenerate AC. At alkaline hydrothermal conditions, the OH– attacks the alpha carbon of PFAS and cleavages the bond between the head group and tail, and forms perfluorinated alcohol, which is subsequently converted into perfluoroacyl fluoride through HF elimination. The perfluoroacyl fluoride then undergoes hydrolysis and decarboxylation, resulting in mineralized products and fluorine ions (Wu et al., 2019; Wang et al., 2022). Using an alkaline hydrothermal process at 120 °C, Zhang et al. (2024a) achieved 81% regeneration efficiency after recycling GAC five times. Microwave irradiation (MWI) is another promising method of regenerating PFAS-laden AC, and it can be treated as an alternative to the conventional heating regeneration method (Vakili et al., 2024). The dielectric nature of ACs allows the conversion of low-powered microwave energy into high heat. Besides, the material is heated uniformly from the inside out during MWI, while in traditional heating, heat diffuses from the heat source to the inside of the material, resulting in the MWI achieving the desired heat faster (Mian et al., 2022). Gagliano et al. (2021) achieved over 90% regeneration efficiency for PFOA- and PFOS-laden AC using 500 W microwave power for 3 min.

The solvent elution regeneration process involves weakening hydrophobic interactions, electrostatic interactions, and/or ion exchange processes. This can be achieved by replacing water with alcohol to weaken hydrophobic interactions, as well as introducing ionic liquids or altering the pH to reduce electrostatic interactions and ion exchange processes (Chularueangaksorn et al., 2014; Deng et al., 2015; Wan et al., 2022; Shaikh et al., 2023). A percentage of alcohol ranging from 10% to 100% (e.g., methanol or ethanol) (Punyapalakul et al., 2013), a combination of alcohol with ionic liquids (e.g., ammonium hydroxide, ammonium chloride, or sodium chloride), or pH-altering bases (e.g., potassium hydroxide or sodium hydroxide) (Zaggia et al., 2016) have been applied in this processes at room temperature, mild heating (e.g., below 80 °C), or through sonication. It has been reported that the optimal dose can achieve over 100% regeneration efficiency, likely due to some activation during the regeneration process (Gagliano et al., 2020). To achieve the optimum regeneration, it is essential to understand the adsorption mechanisms involved. Strong hydrophobic adsorption may not be suitable for regeneration when using ionic liquids or altering the pH of the aqueous solution. Similarly, electrostatically adsorbed PFAS on the adsorbent surface may not respond well to alcohol treatment. For instance, a solution of NH_4OH and NH_4Cl effectively desorbs PFOA, PFOS, PFBA, and PFBS from a quaternary amine-functionalized resin, whereas the same solution is ineffective for hydrophobic resin (Zaggia et al., 2016). Mild heating can be used to enhance desorption process by increasing molecular interactions. Deng et al. (2015) reported an increase in PFOS desorption from 93% to 98% by raising the temperature from 25 to 45 °C.

Both the thermal and solvent elution regeneration processes have their advantages and disadvantages. Thermal regeneration can achieve a certain degree of defluorination, and its effectiveness can be enhanced by increasing the temperature (Xiao et al., 2020b). However, this method can gasify some surface carbon, reduce porosity, and release volatile fluorine and carbon compounds (Xiao et al., 2020b; Sonmez Baghirzade et al., 2021). Furthermore, thermal regeneration may not be suitable for most polymer-grafted PFAS sorbents because grafted polymer could be unstable at high temperatures and transform into aromatic structures, which can impair their subsequent PFAS adsorption performance (Mian et al., 2022). In contrast, solvent elution regeneration is efficient for regenerating functional PFAS adsorbents, but it results in a

significant amount of PFAS-containing organic solvents.

5 Challenges

5.1 Removing short- or ultra-short-chain PFAS

There has been an increased focus on long-chain PFAS, particularly PFOA and PFOS, from policymakers, researchers, and health advisors. This has led to a surge in research aiming at long-chain legacy PFAS removal, while less attention has been given to short-chain PFAS treatment. Removing short-chain PFAS is challenging with pristine ACs due to their high mobility and ease of replacement (Gagliano et al., 2020). Meanwhile, growing research emphasizes the potential health risks of short-chain PFAS and their notable presence in drinking water. Research by PAN (Pesticide Action Network-Europe) members indicates that 98% of PFAS in tap water is trifluoroacetic acid (TFA), with an average concentration of 740 ng/L (Helmut et al., 2024). Short- or ultra-short-chain PFAS tend to remain in drinking water sourced from surface water, whereas long-chain PFAS are removed efficiently (Sadia et al., 2023). Introducing a charged surface can enhance electrostatic interactions, which aids more in the adsorption of long-chain PFAS through both hydrophobic and electrostatic forces than short-chain PFAS. Some functional AC and water-confined nanopores have demonstrated improved removal short-chain PFAS removal performance, but the application of these technologies at the pilot or industrial scale has yet to be validated (Shi et al., 2023).

5.2 Adsorption and regeneration performance

The commercial GAC used in wastewater and drinking water treatment processes exhibits lower performance in removing PFAS compared to various modified carbons, as discussed in Section 4.1. GAC columns with low performance necessitate early regeneration, which incurs significant costs related to transportation, installation, and heating. By enhancing GAC performance, it is possible to extend its lifespan and lower treatment costs. Functionalizing AC may improve its efficacy in either column or batch adsorption studies. This process also offers the advantage of selective adsorption of specific PFAS compounds (Xu et al., 2020). However, the heating regeneration process may alter the functional groups present in the functional AC (Mian et al., 2022). On the other hand, the feasibility of solvent-induced regeneration of functional AC has not yet been assessed for large-scale applications (Lei et al., 2023). It is important to note that ACs are utilized for dynamic micropollutants, and enhancing one particular property, such as increasing particular functional groups, may negatively impact the removal of other pollutants because of diminishing pore structure and surface area. Furthermore, establishing new infrastructure in wastewater or drinking water treatment plants is capital intensive, as the costs associated with equipment and material production are significant (Lai and Ngu, 2020). Increasing surface area (e.g., exceeding 2000–3000 m^2/g with high microporosity) through different activation methods can significantly enhance adsorption performance, making the AC suitable for heating regeneration (Mian et al., 2023). However, for industrial applications, it is crucial that the porous carbon maintains adequate physicochemical stability. AC with low bulk density is prone to attrition during treatment and regeneration. Additionally, the treatment facility needs to expand to utilize low-bulk-density AC to maintain a similar AC weight, which may be highly cost-intensive (Capareda, 2022).

5.3 Cost and environmental sustainability

The cost and environmental sustainability are crucial for AC manufacturing and applications. Although AC is considered cost-effective and environmentally friendly, it causes high costs for PFAS removal due to its limited adsorption performances. Medina et al. (2022) pilot study revealed that GAC costs 30%–40% more than resin and 70% more than modified clay material. The high cost is associated with its large unit installment, often recycling, and lower performance than other adsorbents. Besides coal-based AC usage is also considered a way of carbon emission as it converts stable carbon into gaseous carbon. A life cycle assessment of single-use- and regeneratable AC and resin for PFAS removal by Ellis et al. (2023) concluded that single-use resin was more environmentally friendly and cost-effective than others, while single-use AC was the most costly and least sustainable treatment method. This result was highly influenced by the recent regulation of breakthrough criteria. Since the current regulation is stricter than the previous one, many countries want to achieve zero PFAS emissions, achieving that standard and economic sustainability using current GAC or PACs is a great challenge.

6 Conclusions and outlook

AC is a versatile material used in water treatment, but it

has several limitations when it comes to remediating PFAS. Significant advancements are necessary to achieve current targets of a few nanograms per liter or to eliminate PFAS from water entirely. In this review, we present the latest technologies focused on AC-based PFAS adsorption. We critically discuss various adsorption mechanisms, introduce innovative materials that tackle key challenges, and outline key criteria for developing new carbon-based adsorbents. Despite the progress made, many challenges remain. Therefore, future research should adhere to the recommendations below to advance the new carbonaceous adsorbents and overall PFAS remediation.

1) More efforts are needed to develop effective technologies for removing short-chain PFAS. One possible approach can be engineering narrow cavities that allow the intrusion of short-chain PFAS while blocking large-size pollutants. Besides, small-scale pilot testing should be conducted on recently reported technologies, such as water-confined nanopores, grafting quaternary amine groups, and enhancing hydrophobicity, which help evaluating the effectiveness of these methods in removing PFAS and other pollutants.

2) NBs play a positive role in PFAS removal. However, the method of improving NB pinning on the AC surface is unknown. Developing a feasible method for producing NBs and optimizing their distribution on the AC surface within a column system may improve PFAS removal.

3) Further empirical research and computational simulations can enhance our understanding of how different types of pores—such as pore size and functional groups on pore walls—affect the adsorption of various PFAS compounds. One possible approach involves designing the pore size through controlled methods, functionalizing the pores with heteroatom doping without compromising surface topography, and then applying them for PFAS removal. This strategy may help us understand how pore characteristics influence the removal of PFAS.

4) Economic sustainability is crucial for PFAS treatment. AC performance and recycling should be enhanced greatly. Low-cost synthesis or modulation approaches and waste biomass instead of coal or high-value biomass should be incorporated in PFAS adsorbent preparation.

5) For environmental sustainability, adsorbed PFAS should efficiently defluorinate and close their lifecycle. Simultaneously, PFAS emissions must be halted, and alternatives to this chemical must be developed and utilized.

CRediT Authorship Contribution Statement

Md Manik Mian: Conceptualization, Data curation, methodology, visualization, writing–original draft and editing. **Jiaxin Zhu**: Data curation, investigation, and review. **Xiangzhe Jiang**: Data curation, visualization, investigation, and review. **Shubo Deng**: Conceptualization, funding acquisition, writing–review, editing, and supervision.

Conflict of Interests Shubo Deng is an Editor of *Frontiers of Environmental Science & Engineering*. The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Availability of Data and Materials All data analyzed or generated are included in this article.

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