Review Article

A Comprehensive Review of Surface-Modified Adsorbents for Improving Per- and Polyfluoroalkyl Substances (PFAS) Removal: Materials, Mechanisms, and Regeneration Strategies

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Per- and polyfluoroalkyl substances (PFAS) are persistent organic pollutants that create environmental threats for both human health and natural ecosystems. Currently, the adsorption method remains one of the most effective and scalable technologies for PFAS removal. However, conventional adsorbents such as activated carbon often show limited affinity for short-chain PFAS and reduced performance in complex water matrices. To fill this performance gap, researchers are turning to surface-engineered adsorbents, including functionalized carbons, biopolymer-based materials, minerals, and metal-organic frameworks (MOFs). Carbon-based adsorbents exhibit high adsorption capacities for long-chain PFAS. At the same time, mineral and clay modifications create selective adsorbents through tailored surface charges, biopolymer systems achieve better PFAS adsorption through grafted functional groups, and MOFs provide adjustable pore structures that form strong bonds with short-chain PFAS. These modifications introduce positive surface charges and create fluorophilic domains, improving PFAS removal through electrostatic and hydrophobic mechanisms. Surface modifications enhance host-guest interactions. The functional groups on the adsorbent form stronger connections with PFAS headgroups, improving binding strength and PFAS removal efficiency. Furthermore, the modified adsorbents show strong regeneration potential after both solvent washing and pH-based desorption. Research needs to validate performance through actual water testing while creating low-cost regeneration methods to enable large-scale deployment of the technology. This review synthesizes current evidence on surface-modified adsorbents, encompassing material design approaches, primary adsorption mechanisms, and restoration methods, to inform the development of future PFAS cleanup methods.

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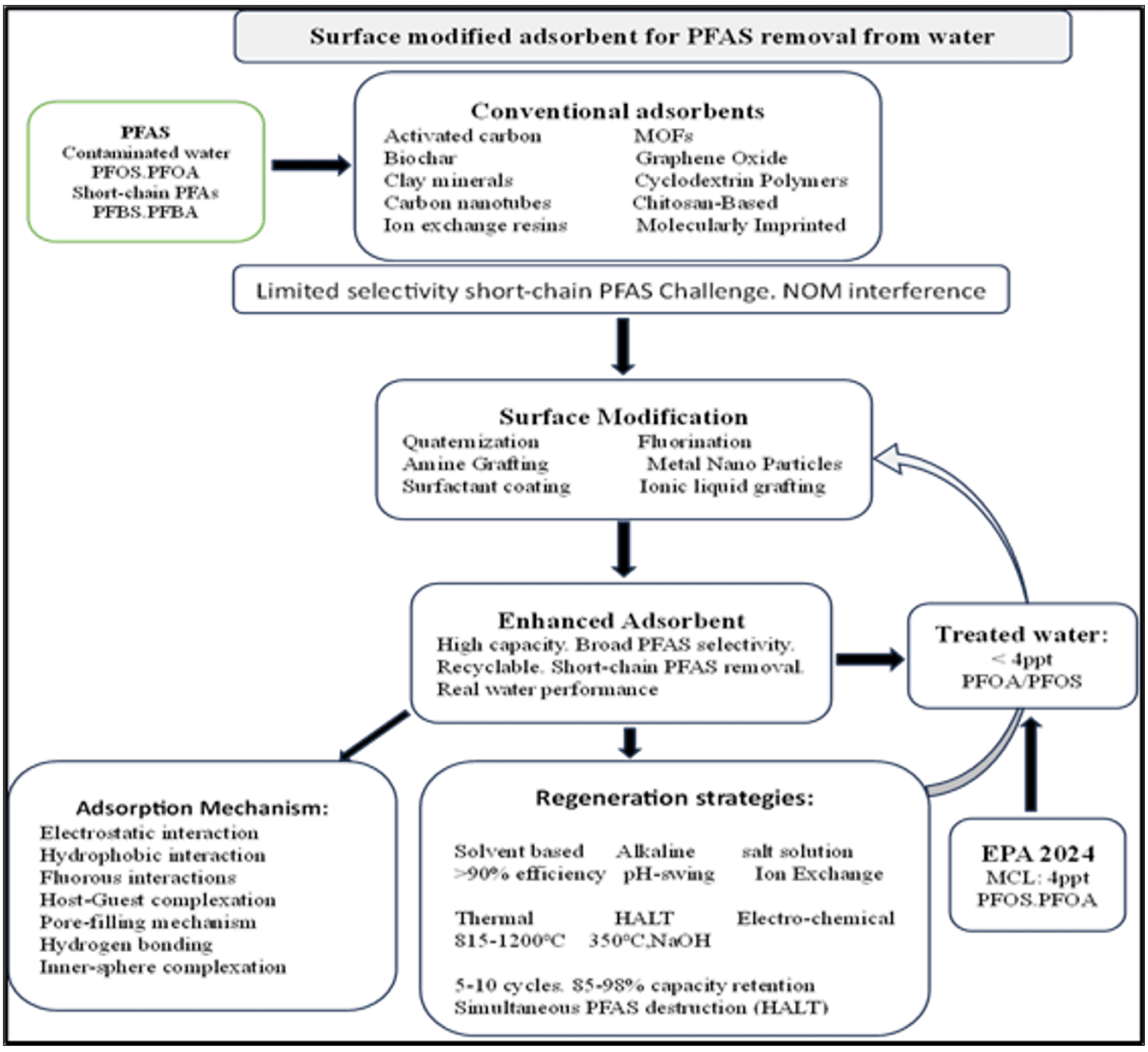
Highlights

- Surface-modified adsorbents exhibit superior PFAS removal efficiency compared to conventional materials.
- Electrostatic, hydrophobic, and fluorous interactions govern enhanced PFAS adsorption performance.
- Fluorous domains provide selective affinity toward short-chain PFAS compounds.
- Carbonaceous, mineral, biopolymer, and MOF-based modifications enable tailored surface functionality and pore architecture.
- Solvent, alkaline, and electrochemical regeneration methods achieve >90% recovery efficiency.
- The HALT process enables simultaneous PFAS mineralization and adsorbent restoration.
- Modified adsorbents maintain 85–98% adsorption capacity over multiple regeneration cycles.
- Surface engineering offers scalable solutions for meeting ppt-level PFAS regulatory standards.

Graphical Abstract



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1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are synthetic persistent organic pollutants, and their worldwide distribution has caused environmental damage and health problems for humans[1]. PFAS compounds include short chains with fewer than eight carbon atoms and long chains with eight or more carbon atoms [2]. The long-chain compounds like perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) pose more severe environmental issues than the short-chain compounds [3][4]. PFOS displays stronger hydrophobic properties and a greater overall negative charge than PFOA because its perfluoroalkyl chain is longer and includes a sulfonate functional group[5]. Furthermore, these properties of PFOS enable it to bind more effectively to various sorbent materials [6]. Previous research has demonstrated that carbon- and mineral-based adsorbents exhibit better removal performance for PFOS and PFOA due to this tendency [7][8]. The sorbent surfaces exhibit better PFOS partitioning due to increased hydrophobicity, and the strong negative charge of PFOS enhances its electrostatic attraction to positively charged sorbents [9]. The different structural characteristics of PFOS and PFOA help account for the observation that PFOS removal generally occurs more effectively across various sorption mechanisms [10]. To understand the varied sorption behavior, it is essential first to analyze the molecular architecture of PFAS, characterized by a carbon backbone in which hydrogen atoms are partially or fully replaced by fluorine [11]. The compound forms a carbon-fluorine bond that exhibits strong polarity, with a bond strength of 485 kJ/mol, thereby enhancing chemical and thermal stability. PFAS materials exhibit both hydrophilic and hydrophobic characteristics because their structure contains a hydrophilic head group and a hydrophobic C-F tail. PFAS materials have become widespread in industrial applications through their use in household products, industrial products, flame retardants, and waterproof clothing, resulting in significant environmental distribution [12][13]. PFAS can bioaccumulate in human and animal bodies, leading to multiple harmful health effects, such as lower birth weights in infants and a higher risk of kidney cancer during adulthood[3][14].

In light of increasing evidence regarding these health risks, regulatory agencies have begun to set enforceable limits on PFAS in drinking water. The U.S. Environmental Protection Agency (EPA) established its first National Primary Drinking Water Regulation (NPDWR) for PFAS in April 2024 under the Safe Drinking Water Act, in response to growing health risks and widespread environmental contamination. The regulation established Maximum Contaminant Levels (MCLs) of 4 parts per trillion (ppt) for PFOA and PFOS individually, and 10 ppt for perfluorohexane sulfonic acid (PFHxS), perfluorononanoic acid (PFNA), and hexafluoropropylene oxide dimer acid (HFPO-DA, also known as GenX chemicals). A new Hazard Index was established by the rule to control PFAS mixtures of two or more compounds, which produce cumulative health impacts when found together in drinking water. Public water systems must finish their first monitoring by 2027 according to the regulation, while they must reach total compliance by 2031. The regulation requires advanced remediation technologies to achieve ultra-

low removal thresholds while efficiently eliminating both long-chain and short-chain PFAS because it uses detection limits that operate at low parts-per-trillion levels [15].

Meeting these strict regulatory standards has increased demand for advanced, efficient PFAS removal technologies. PFAS remediation technologies operate through two distinct methods: destructive and non-destructive. Destructive technologies break C-F bonds to produce harmless molecules from PFAS. These technologies include chemical oxidation, advanced reduction, electrochemical oxidation, photocatalysis, sonochemical treatment, hydrothermal reaction, plasma treatment, and thermal destruction[16]. In contrast, non-destructive technologies use physical separation methods to extract PFAS from materials, producing concentrated PFAS products. The non-destructive technologies use three methods: sorption, reverse osmosis, and nanofiltration [17]. While each approach offers distinct advantages, the two technologies function as separate methods, but their combination yields optimal PFAS removal. Adsorption has emerged as the top PFAS removal technology because it offers a cost-effective solution that achieves high removal rates while protecting the environment. Previous studies utilized specific adsorbents, including biochars, granular and powdered activated carbon, carbon nanotubes, magnetic biochars, natural clays, metal nanoparticles, soils, and anion exchange resins [18].

The effectiveness of these adsorbents is not uniform; it varies significantly based on specific material characteristics. PFAS adsorption depends on adsorbent properties, such as surface functional groups, aromaticity, anion exchange capacity, and pore size and pore volume[19]. PFAS molecules exhibit complex sorption behavior due to their hydrophilic, hydrophobic, and oleophobic C-F chains. The main driving forces behind PFOA and PFOS adsorption are electrostatic interactions and the hydrophobic effect [20]. Despite these interactions, conventional adsorbents frequently fail to deliver adequate performance under real-world conditions. Scientists modified adsorbents by altering surface morphology or chemical composition because the adsorbents exhibited performance drops when faced with organic matter and water constituents and showed slow sorption rates and reduced capacity to distinguish between different substances [21].

This limitation has driven innovation toward a new generation of engineered materials, such as amine-modified carbonaceous materials, β-cyclodextrin-based polymers, and organic-inorganic hybrid materials [22]. Researchers have studied modified adsorbents extensively, but they have failed to gather all existing evidence into a comprehensive synthesis. To guide this review, three questions were identified: 1) What type of surface modifications are most effective for enhancing the removal of different PFAS classes? 2) What are the dominant mechanisms reported for these enhanced adsorbents? 3) What regeneration strategies have been reported to demonstrate both efficiency and sustainable performance? The review systematically evaluates the available evidence on PFAS removal using surface-modified adsorbents that employ different modification strategies, adsorption mechanisms, and regeneration approaches.

2. Review methodology

This review provides a comprehensive assessment of surface-modified adsorbents for PFAS removal from aqueous solutions. A systematic literature search was conducted using the Web of Science, Scopus, and Google Scholar databases, covering publications from 2008 to 2025. Search terms included combinations of PFAS, PFOA, PFOS, adsorption, surface modification, functionalization, and regeneration. Studies were included if they: [1] reported experimental data on modified adsorbents for PFAS removal, [2] provided quantitative data on adsorption capacity or removal efficiency, and [3] were published in peer-reviewed journals. The research focused on studies examining how modifications alter material properties, their impact on performance, and the potential for material reuse. Additionally, citation tracking and reference list screening were used to identify additional articles containing this information.

3. Conventional adsorbents in PFAS removal: overview

Previous studies showed that conventional adsorbents such as activated carbon, ion exchange resins, biochar, clay minerals, and carbon nanotubes have been widely investigated for PFAS remediation. However, these materials exhibit significant limitations that compromise their practical application (**Table 1**). Activated carbon, despite its high surface area, exhibits limited selectivity for short-chain PFAS [23][24], reduced efficiency in the presence of natural organic matter [23], and requires frequent regeneration [24]. Ion exchange resins face challenges, including high operational costs [24], performance degradation due to competing ions [25], and limited capacity for certain PFAS compounds [25]. Biochar varies in quality depending on the feedstock, has a lower adsorption capacity than activated carbon, and is insufficient to remove short-chain PFAS. Clay minerals are limited by low surface area [26], poor selectivity in complex water matrices, and challenging separation after treatment [24]. Carbon nanotubes, while offering unique structural properties, are extremely costly and face challenges in dispersion and recovery in aqueous solutions. Graphene and graphene oxide share similar drawbacks, including high production costs, aggregation issues in water, challenging scale-up, and limited regeneration potential, despite their theoretical surface area [26]. Metal-organic frameworks (MOFs) offer tunable pore structures, particularly suited to short-chain PFAS capture, but face high production costs, poor water stability, challenging scale-up, and risks of metal leaching [24]. Chitosan-based materials provide biocompatible and functionally versatile platforms through their amine and hydroxyl groups, yet exhibit limited mechanical strength, pH-dependent performance, poor stability in acidic conditions, and low adsorption capacity for certain PFAS [27]. Cyclodextrin-based polymers achieve selective PFAS binding through host-guest inclusion complexation within their hydrophobic cavities. Still, they are limited by the high cost of pure cyclodextrins, limited stability at extreme pH, and challenges in water solubility without crosslinking [28]. Molecularly imprinted polymers (MIPs) offer high selectivity through

template-guided recognition sites. Still, their complex synthesis, elevated production costs, limited binding-site density, and concerns about template leaching currently limit their broader application [29][30].

The effectiveness of conventional adsorbents varies dramatically depending on the molecular structure and chain length of target PFAS compounds. Long-chain PFAS molecules typically exhibit higher removal rates due to their greater hydrophobicity. In contrast, short-chain PFAS compounds present a particularly stubborn challenge owing to their greater water solubility and reduced hydrophobic interactions [31]. This performance gap, combined with the specific limitations of each adsorbent class, has driven extensive research into surface modification strategies aimed at enhancing adsorption capacity, selectivity, and regeneration potential for both legacy long-chain compounds and increasingly prevalent short-chain alternatives.

Adsorbent	Limitation	Need for Modification
Activated carbon	Limited selectivity for short-chain PFAS[29][32][33]; competes with natural organic matter[23]; requires frequent regeneration[24]; decreased efficiency at higher pH[34].	Surface functionalization to enhance selectivity[25][34], pore size optimization for short-chain PFAS[32], regeneration methods development [24][29], and quaternary ammonium compounds[25].
Ion exchange resins	High cost compared to other compounds[24][29], performance affected by competing ions[35], limited capacity for certain PFAS compounds[35], regeneration generates concentrated waste streams[36], and resin fouling by organic matter [36].	Development of PFAS-specific functional groups[31], hybrid materials to reduce fouling, improved regeneration techniques[24], and enhanced hydrophobic and electrostatic interactions[35].
Biochar	Variable quality based on feedstock, lower adsorption capacity than AC, poor mechanical strength, limited selectivity, and insufficient removal of short-chain PFAS[3].	Chemical activation to increase surface area, metal oxide impregnation, [37]amino-functionalization[31], controlled pyrolysis conditions, and surface modification with quaternary ammonium [3].
Clay minerals	Limited surface area[24], low adsorption capacity for neutral PFAS, poor selectivity in complex water matrices, challenging separation after treatment[24], and interlayer collapse in some conditions [22].	Organo-modification with quaternary ammonium, pillaring to increase interlayer spacing, acid activation to increase surface area, magnetic modification for easier separation[24], and combination with other adsorbents.
Metal-organic frameworks	High production cost[24], poor stability in water[24], challenging scale-up[37], potential metal leaching[24], and limited research on long-term stability[24].	Hydrophobic ligand incorporation[24], mixed-metal approaches[24], development of water-stable MOFs, functionalization with fluorophilic groups[24], and integration with other materials for stability[38].
Chitosan-based materials	Limited mechanical strength[27], pH-dependent performance[27], poor stability in acidic conditions[27], low adsorption capacity for some PFAS[27], and degradation over time[39].	Cross-linking to improve stability, grafting with functional groups, composite formation with other materials, magnetic modification for separation, and development of chitosan-based beads/hydrogels[39].
Graphene and graphene oxide	High production cost[26], difficult separation from water[26], aggregation issues[26], challenging scale-up[26], and limited regeneration potential[26].	Functionalization with amino groups[31], formation of 3D structures to prevent aggregation, integration with other materials (composites)[40], development of magnetic graphene derivatives[41], and surface modification to increase selectivity[26].

Adsorbent	Limitation	Need for Modification
Minerals and mineral oxides	Variable performance based on mineral type, limited surface area for some minerals, sensitivity to solution pH[42], poor selectivity in complex matrices, and low capacity for short-chain PFAS[43].	Surface modification with organic compounds [31], nanosizing to increase surface area[44], composite formation with carbon materials[45], controlled morphology development, and incorporation of active metal sites[41][46].
Molecularly imprinted polymers	Complex synthesis process[30], high production cost, limited binding sites, template leaching concerns, and less effectiveness for structurally diverse PFAS[29].	Optimization of template-monomer interactions, development of universal PFAS recognition sites, integration with other materials for enhanced stability, scale-up of production methods, and improvement of regeneration techniques [29].
Carbon nanotubes	Extremely high cost, potential environmental risk, challenging dispersion in water, difficulty in recovery after use, and limited adsorption capacity for some PFAS[47].	Surface functionalization to increase hydrophobicity, development of CNT-polymer composites, magnetic modification for easier separation, oxidation to increase surface functionality, and creation of hierarchical structures[47][48].
Cyclodextrin-based materials	High cost for pure cyclodextrins[28], limited stability in extreme pH[28], relatively small cavity size for longer PFAS, water solubility challenges, and lower capacity than some conventional adsorbents[49].	Polymer cross-linking to improve stability[50], functionalization to enhance selectivity, development of cyclodextrin-based networks, integration with other materials, and optimization of cavity size for different PFAS[48].

Table 1. Limitations of conventional adsorbents and modification strategies

4. Mechanism and regeneration of enhanced adsorbents

4.1. Surface Modification Strategies

Surface modification is a promising technique for overcoming the limitations of conventional adsorbents. Notably, this part summarizes the major modification approaches for various material classes to improve PFAS removal.

4.1.1. Modified carbonaceous materials for PFAS removal

Biochar modification

A 2021 study by J. Steigerwald investigated the potential to use spent coffee grounds (SCG) to engineer a cost-effective adsorbent for PFOS removal from water. Importantly, the conversion of SCG to biochar was achieved by activating it with potassium hydroxide (KOH) to obtain the final product, SCGKOH. This activation with KOH increases the surface area of SCGKOH, leading to an efficiency 300 times higher than that of untreated pyrolyzed SCG. Accordingly, the study found that, without treatment, pyrolyzed SCG exhibited low PFOS adsorption under regular environmental concentrations. Nonetheless, KOH activation of pyrolyzed SCG can be performed under optimal conditions, achieving PFOS removal and uptake of up to 99.6%. The SCG:KOH ratio of 1:1, being optimal, proved advantageous for producing SCG-KOH with a maximum adsorption capacity of 43.4 mg/g. The SCGKOH adsorbent exhibits excellent efficiency and a high surface area due to its small pore size. Moreover, it exhibited a negative surface charge and very few surface functional groups, indicating a high carbon content. Furthermore, due to its structural features and the presence of longer hydrophobic regions, SCGKOH could be a good candidate for adsorbing PFOS via hydrophobic binding via the CF chain, despite its lack of surface functionality. When activated, this waste material was shown to enhance the removal of trace-level PFOS[51]. In addition, according to a 2023 study by I. Militao and colleagues, rice straw-derived biochar and albumin from Moringa oleifera seeds encapsulated in alginate beads were highly effective at adsorbing PFOS and PFBS from water. In this study, both albumin and biochar beads achieved high PFOS removal, at 87% and 99%, respectively, within 16 hours. The evidence indicates that the albumin beads had lower PFBS removal (10%), whereas the biochar beads removed 40% in under 48 hours, outperforming other natural adsorbents. In general, pH and natural organic matter did not significantly affect PFAS removal with biochar beads, and hydrophobic interactions played a dominant role over electrostatic interactions [52].

Carbon modifications

In 2016, researchers Y. Zhi et al. increased the basicity of activated carbon to assess its ability to remove PFOS and PFOA pollutants. Through this study, several activated carbons extracted from coal, coconut shell, wood, and phenolic polymers were treated with high-temperature and ammonia gas to increase their surface basicity. This led to the finding that the ammonia treatment is more effective than the heat treatment in enhancing basicity and improving PFOS/PFOA adsorption affinity. Notably, the higher point-zero charge and total basicity led to better adsorption performance. However, the effectiveness of surface modifications varied across different activated carbon precursor materials. For instance, wood-based carbon and activated carbon fibers showed a 1-3 order of magnitude enhancement in adsorption, while some materials exhibited reduced PFOS or PFOA uptake after modification [25]. In addition, in 2017, W. Chen et al. successfully prepared Polyacrylonitrile Fiber (PANF) and

derived activated carbon fibers (PACF) as efficient adsorbents for removing PFOS and PFOA from water. As a result, the PACFs showed significantly higher sorption capacities than PANF, with 1.52 mmol/g for PFOS and 0.73 mmol/g for PFOA, outperforming commercial coal-based activated carbons. This efficiency is due to the higher number of available sorption sites on PACF surfaces. The sorption studies have shown that multilayer adsorption occurred after the monolayer, through electrostatic and hydrophobic mechanisms, forming micelle-like structures. Significantly, the spent PACFs can be effectively regenerated and reused over multiple sorption-desorption cycles, which is a significant advantage[53].

Carbon nanotube modification

In several studies, mechanisms such as hydrophobicity (which also leads to hemimicelle/micelle formation), electrostatics, and hydrogen bonding are the primary mechanisms of removal by activated carbon adsorbents. These mechanisms depend on the source material of the activated carbon, surface chemistry, and type of carbon used[47]. However, in 2018, L. Liu and colleagues grafted different metal nanoparticles (iron, copper, and zinc) onto multiwalled carbon nanotubes (MWCNTs) to remove PFOA. This facilitates the removal of PFOA via inner-sphere complexation, hydrophobic interactions, and electrostatic mechanisms between PFOA and the modified MWCNT surfaces [54].

Graphene-based materials

In a comprehensive study conducted by Pervez et al. (2024), eight graphene oxide (GO)-based adsorbents were developed and evaluated for their effectiveness in removing PFAS mixtures from river water. Of all the materials tested, GO modified with the cationic surfactant cetyltrimethylammonium chloride (GO-CTAC) demonstrated the most promising results, achieving nearly 100% removal of all 11 PFAS compounds examined, including both short-chain and long-chain varieties, which is crucial for practical applications. The adsorption kinetics were best described by the pseudo-second-order model, suggesting rapid uptake driven more by chemisorption than by simple physisorption. In terms of equilibrium, the Toth isotherm provided the best fit for the data, indicating a complex arrangement on the GO-CTAC surface, rather than a single uniform site, likely due to heterogeneous multi-site interactions. Overall, GO-CTAC's superior performance can be attributed to the positive surface charges introduced by the cationic surfactant, which enhanced electrostatic attraction toward the anionic PFAS headgroups. Additionally, hydrophobic interactions between the surfactant's alkyl chains and the perfluorinated tails of PFAS further strengthened the binding. These findings underscore the significance of surface charge engineering in graphene adsorbents and suggest that cationic modification of GO can effectively address the common challenges associated with unmodified graphene oxide, such as aggregation tendencies and low affinity for short-chain PFAS. Ultimately, GO-CTAC appears to be a highly viable candidate for real-world PFAS cleanup, particularly in challenging conditions where maintaining high performance is essential[40].

A landmark study by Xu et al. (2025), published in the National Science Review, introduced an innovative concept for a 2D adsorbent. They developed a highly charged, monodispersed material called PAGO by attaching 1-nm-thick polyamine adlayers to graphene oxide (GO) nanosheets. This novel material exhibited an impressive PFAS adsorption capacity of approximately 3,070 mg/g, which is several dozen times greater than that of unmodified GO and commercial activated carbon. As a result, PAGO has emerged as one of the leading graphene-based adsorbents documented in the literature to date. The material also demonstrated remarkable removal kinetics, achieving about 57–95% of its equilibrium capacity within a single minute and nearly 100% PFAS removal from contaminated water in just a few minutes. Furthermore, the researchers did not stop there. Through a combination of experimental characterization and theoretical modeling, they proposed that the robust adsorption occurs due to a synergistic, multi-mechanism interaction. This includes electrostatic attraction between protonated polyamine surface sites and the anionic polar headgroups of PFAS, as well as hydrogen bonding and hydrophobic interactions involving the non-polar perfluorinated tails. Notably, the study also indicated that the spent adsorbent could be regenerated, with the desorbed PFAS subsequently destroyed. This approach suggests a closed-loop treatment strategy that effectively addresses both removal efficiency and long-term PFAS management. Overall, these findings position PAGO as a next-generation graphene-based material capable of overcoming the common capacity, kinetic, and regeneration limitations typically associated with conventional GO adsorbents[55].

According to the research of T. Wu et al., it has been established that PFOS adsorbs on few-layered porous graphite (FPG) via both electrostatic and hydrophobic mechanisms. In particular, PFOS inserts itself between the hydrophobic lamellae of FPG through intercalation. Additionally, PFOS may also penetrate hydrophobic pores on the particle surfaces. Finally, this dual electrostatic-hydrophobic binding model explains the high multilayer PFOS uptake capacity of FPG[56].

4.1.2. Natural and bio-derived materials:

Clay minerals modification

The researchers M. Wang et al. (2021) conducted a study to determine the most effective adsorbent for PFAS removal: modified montmorillonite clay with carnitine and choline versus unmodified clay. They used both experimental and modeling techniques to optimize interactions and predict sorbent efficacy. The study found that, in addition to hydrophobic and electrostatic interactions, hydrogen bonding significantly enhanced the binding of PFOA and PFOS to modified clays. Specifically, PFOA formed hydrogen bonds between its carboxyl group and the functional group on clay carnitine, while its fluorinated tail bonded to the interlayers. On the other hand, PFOS is primarily held together by hydrogen bonds between its sulfonate group and hydroxyl groups on the organic amendment [13].

In addition, a new type of adsorbent, called fluorinated montmorillonite (F-MT), was developed by Du et al. (2016) by replacing cationic surfactants on clay with fluorinated ones. Consequently, the F-MT adsorbent is highly effective at removing PFOS and PFOA, trace-level pollutants, with a capacity that outperforms that of activated carbon and resin adsorbents. The spent F-MT can be fully regenerated by washing with methanol and reused for up to five cycles without any reduction in performance. Moreover, the most important point is that F-MT is selectively efficient at removing both PFOS and PFOA, even in the presence of other organic pollutants, such as phenol, pyridine, SDS, and phenanthrene, which have a minimal effect on adsorption. The selectivity is due to PFOS and PFOA binding to the hydro-oleophobic fluorinated chains of F-MT, while hydrocarbons adsorb other organics. In brief, this provides a new understanding of the mechanisms of PFAS adsorption on fluorinated adsorbents[57].

Furthermore, Khodabakhshloo and Biswas in 2023 developed oleylamine-modified composites from naturally occurring Iranian zeolite and diatomaceous earth (DE) for PFOS removal from simulated wastewater. Characterization via TGA, SEM, FTIR, and surface/pore size analysis confirmed successful surface modification with oleylamine. The modified materials demonstrated substantially enhanced PFOS adsorption capacities compared to their unmodified counterparts. Oleylamine-zeolite achieved 25.5 mg/g versus 0.39 mg/g for raw zeolite, while oleylamine-diatomite reached 14.1 mg/g versus 4.72 mg/g for raw DE. Adsorption isotherm modeling indicated predominantly monolayer binding, and post-adsorption analysis attributed the improved performance primarily to electrostatic and hydrophobic interactions between PFOS and the modified surfaces[21].

Similarly, Dong et al. (2021) conducted a study synthesizing a modified natural clay with imidazolium-type ionic liquids (ILs) to remove both PFOS and PFOA. As a result, this modified natural clay with ionic liquids achieved maximum adsorption capacities of 66.2 mg/g for PFOA and 151.5 mg/g for PFOS. This new adsorbent could be regenerated with >98% desorption efficiency using three different eluents: 50 mM NaCl, 500 mM NaCl, or a methanol/water (50%/50%) mixture[22].

Mineral oxide modification

In a 2020 study by M. Hassan et al., the efficacy of red mud-modified sawdust (RMSDN600) and unmodified sawdust (SDN600) in eliminating PFAS from water was investigated. The RMSDN600 was prepared by incorporating red mud, a material containing minerals such as magnetite (Fe_3O_4), ferrihydrite, and desilicated minerals. The objective of this study was to determine the difference in PFOS removal capacity between RMSDN600 and regular SDN600. The adsorption mechanism is governed by hydrophobic interactions, which are stronger here than PFOS-PFOS electrostatic repulsion. Thus, the higher adsorption of RMSDN600 and SDN600 was 194.6 mg/g and 178.6 mg/g, respectively, at pH 3.1. This difference in adsorption capacity is due to the

abundance of protonated metal-based functional groups and a more ordered graphitic carbon structure resulting from catalytic degradation and transformation of cellulose and hemicellulose on the RMSDN600[19].

4.1.3. Polymer-based modifications

Chitosan-based materials

In 2008, Q. Yu used an epichlorohydrin crosslinker and PFOS as a template molecule to synthesize a chitosan-based molecularly imprinted polymer (MIP) adsorbent. This adsorbent is highly effective for PFOS adsorption, with a capacity of 560 µmol/g, higher than that of the non-imprinted polymer (258 µmol/g). As a result, this MIP showed good selectivity with minimal negative effects from other anionic pollutants. However, electrostatic interactions were found to play a crucial role in PFOS sorption. Therefore, the MIP can be regenerated and reused for at least five cycles without any loss of performance[58]. Similarly, S. Deng et al. (2012) grafted long polymer brushes containing quaternary ammonium groups onto cotton using surface-initiated ATRP (Atom Transfer Radical Polymerization). This synthesized quaternized cotton displayed exceptional adsorption efficiency for removing PFOS and PFOA from water, with adsorption equilibrium achieved within 4-12 hours. At pH 5, the quaternized cotton exhibited high adsorption capacities of 3.3 mmol/g for PFOS and 3.1 mmol/g for PFOA, surpassing those of most other adsorbents. Additionally, the adsorption was effective over a wide pH range of 3-10. Finally, the results showed that the quaternary ammonium groups facilitated PFOS/PFOA removal through anion-exchange mechanisms [9].

Cyclodextrin-based polymer

A **study conducted by A. Yang et al. (2020)** revealed that incorporating amino groups into cyclodextrin (β-CD) polymer crosslinkers can enhance the binding of anionic PFAS. To examine the individual contributions of the crosslinker groups and the β-CD cavities to binding anionic PFAS, adsorbents were synthesized using tripodal crosslinkers containing either three amino or three amido groups that crosslink the β-CD polymer networks. These amino- and amido-crosslinked β-CD adsorbents were prepared with comparable physical and chemical properties, enabling an investigation of how the crosslinker functionality and the β-CD host affect the binding affinity and capacity for anionic PFAS compounds. The study compared β-CD polymers with amino or amido crosslinkers and found that amines provided superior removal for anionic PFAS over amides. Furthermore, both β-CD polymers outperformed activated carbons for PFOA removal, indicating the importance of β-CD host-guest inclusion complexes. While the amido-functionalized adsorbents exhibited low GenX binding, the amino polymer exhibited outstanding GenX affinity/capacity due to electrostatic interactions that are critical for short-chain/branched PFAS. Notably, the amine-β-CD polymer demonstrated 100-fold higher affinity. It doubled the

capacity for PFOA over GenX, the highest values reported, highlighting the synergistic effects of electrostatic and host-guest binding for efficient anionic PFAS removal [49].

4.1.4. MOF-based materials

Recent advances highlight the potential of metal-organic frameworks (MOFs), a highly tunable adsorbent for PFAS remediation. A comprehensive review by Lee et al. (2025) emphasized that MOFs exploit multiple adsorption mechanisms, including **electrostatic attraction, hydrophobic interactions, hydrogen bonding, and Lewis acid–base coordination**, to achieve enhanced selectivity, particularly for short-chain PFAS. However, the review also noted persistent challenges in aqueous stability, regeneration efficiency, and scalability, underscoring the need for the rational design of functionalized MOFs incorporating amine, fluorophilic, or hydrophobic groups. Complementing this synthesis, an experimental study published in *Small* (2025) demonstrated the efficacy of a **bio-derived, water-stable MOF, $CuII_2$(S, S)-hismox·$5H_2O$**, synthesized from histidine. This material achieved **80–100% removal of long-chain PFAS** (PFOS, PFDA, PFUnDA) and up to **86% removal of short-chain PFAS** (PFBS, PFBA), with **rapid adsorption kinetics (<30 seconds under continuous flow)** and excellent reusability across multiple regeneration cycles. Structural analysis via single-crystal X-ray diffraction confirmed specific binding modes between PFAS molecules and MOF frameworks, providing mechanistic insight into their high affinity. Together, these studies establish MOFs as a promising class of adsorbents, combining **selectivity, efficiency, and durability**, while highlighting the importance of continued innovation to overcome stability and scale-up barriers[59][60]

4.1.5. Ion exchange resin functionalization

Based on previous studies, ion exchange resins remove long-chain PFAS via hydrophobic interactions and hydrogen bonding, whereas short-chain PFAS rely more on electrostatic interactions [38][61]. Furthermore, S. Woodard et al. (2017) found that anion exchange resins can effectively eliminate PFAS via two mechanisms: dual ion exchange and adsorption. This is due to the characteristics of PFAS, with a hydrophobic nonionic carbon tail and a negatively charged head, and the resin beads containing neutral copolymers with positive sites. Consequently, the PFAS tail attaches to the hydrophobic resin backbone, and the head bonds to the positive sites. This dual mechanism of ion exchange and adsorption enables higher PFAS removal than with solely adsorption media [62].

4.2. Adsorption mechanisms

The results of earlier research indicate that adsorbent surface modifications enhance PFAS removal through multiple mechanisms. The design process for optimized adsorbents targeting specific PFAS compounds requires an understanding of these mechanisms.

4.2.1. Electrostatic Interactions

PFAS compounds maintain their anionic state at typical environmental pH levels because of their low acid dissociation constant (pKa) properties[53]. PFAS removal through electrostatic interactions achieves its highest efficiency. The process of surface modification by introducing a positive charge, achieved via quaternization with quaternary ammonium groups or amino-functionalization, results in stronger electrostatic attraction between the adsorbent material and the anionic PFAS headgroups.

The 2016 study by Zhi et al. showed that cotton treated with quaternary ammonium groups achieved PFOS/PFOA removal primarily through anion exchange [25]. Moreover, the research of Yang et al. (2020) demonstrated that amino-crosslinked cyclodextrin polymers outperformed amido-functionalized variants because electrostatic interactions enabled better removal of short-chain and branched PFAS[49]. Electrostatic interactions were the key factor enabling PFOS detection during chitosan-based MIP adsorption experiments, as confirmed by the study by Yu et al. (2008) [58]. In addition, the dual mechanism of ion exchange resins occurs because PFAS, being negatively charged, binds to positive sites on neutral copolymer beads. At the same time, its hydrophobic tail attaches to the resin backbone, according to Ateia et al. (2019) [32].

The electrostatic mechanism is particularly important for removing short-chain PFAS. Electrostatic attraction increases in strength with decreasing PFAS chain length because it compensates for diminished hydrophobic interactions in short-chain compounds, according to Lei et al. (2023) [63].

Finally, cationic adsorbents demonstrate outstanding performance for PFBA and PFBS, which are difficult to adsorb with standard activated carbon materials, according to this research.

4.2.2. Hydrophobic Interactions

Hydrophobic interactions between the perfluorinated carbon tail of PFAS and the hydrophobic regions of adsorbents primarily drive adsorption processes. This mechanism is effective for removing long-chain PFAS because these substances exhibit greater hydrophobicity than their shorter counterparts [22]. Steigerwald et al. demonstrated in 2021 that SCGKOH could efficiently adsorb PFOS via hydrophobic interactions with its carbon-fluorine chain because the material lacked surface functional groups [51]. According to the research conducted by Militao et al. (2023), hydrophobic interaction served as the primary mechanism for PFOS removal from biochar, while electrostatic interaction functioned as a secondary mechanism [52]. Chen et al. (2017) studied multilayer adsorption via electrostatic and hydrophobic interactions, leading to micelle-like structures that formed on PACF surfaces [53]. The research conducted by Hassan et al. (2020) demonstrates that adsorbent surfaces exhibit stronger hydrophobic interactions that govern adsorption than PFOS-PFOS electrostatic repulsion between PFOS molecules [19]. Recent computational studies have provided molecular-level insights into hydrophobic

interactions. Lei et al. (2023) demonstrated that PFAS molecules prefer to align their long hydrophobic perfluoroalkyl chains with graphite-like aromatic structures on biochar surfaces, resulting in binding energies that increase with chain length [63]. This study explains that hydrophobic interactions remain the main mechanism for long-chain PFAS adsorption on carbon-based materials, as organic matter competes with this process. The extent of hydrophobic interactions depends on the length of the PFAS chains. Hydrophobic binding is more effective for longer chains because shorter chains rely on electrostatic interactions for removal [49]. The research conducted by Minervino et al. (2024) demonstrated that hydrophobic interactions are the main mechanism by which both anionic and zwitterionic PFAS bind to carbon-based materials, based on their systematic studies of zwitterionic PFAS [64].

4.2.3. Fluorous Interactions

Fluorous interactions, which describe how fluorinated compounds interact more strongly with one another than with other materials, are a specialized method for removing PFAS from contaminated sites. The study by Du et al. (2016) demonstrated that fluorinated montmorillonite (F-MT) could effectively bind PFOS and PFOA via its hydro-oleophobic fluorinated chains that exhibit fluorous affinity. In contrast, other organic pollutants could only attach to the hydrocarbon regions of the material. [57].

4.2.4. π-π Interactions

The π-π stacking mechanisms, though less commonly discussed in the PFAS literature than in other organic pollutant literature, can contribute to adsorption on aromatic carbon surfaces and graphene-based materials. Aromatic adsorbents attract PFAS aromatic moieties via their π-electron systems, thereby creating this interaction. Studies showed that graphene oxide and reduced graphene oxide materials with higher π-electron density can adsorb more organic contaminants via π-π stacking. The electron-withdrawing nature of fluorine atoms can influence the π-electron distribution, potentially altering PFAS adsorption system interactions [3][22].

4.2.5. Pore-Filling Mechanisms

Pore-filling is known as a critical physical adsorption mechanism, particularly for microporous adsorbents. The size-selective nature of this mechanism makes it especially important for targeting specific PFAS chain lengths.

The study conducted by Kebede et al. (2025) showed that ball-milled colloidal activated carbon (CACBM) achieved better PFAS elimination because its pore-filling mechanism moved to a more efficient stage. The Polanyi-Dubinin-Manes (PDM) isotherm model demonstrated that micropore-filling was the main mechanism, as CACBM achieved 7.4× better PFOS extraction and 6× better PFOA extraction than granular activated carbon [65]. The relationship between pore size and PFAS adsorption has been systematically investigated. Furthermore, the research by Liu Z et al. (2024) established that ultra-micropores with dimensions less than 1.2 nm serve as the

primary PFOA adsorption sites because they exhibit higher adsorption efficiency than small mesopores, which have dimensions in the 2.0–3.0 nm range. The molecular dynamics simulations showed that PFOA molecules bind to pore walls in ultra-micropores with maximum interaction energies of -567.32 kcal·mol^{-1} because the overlapping potential barriers from both sides of the pore walls create more effective adsorption sites. PFOA forms micelles and hemi-micelles in small mesopores, which enables these pores to serve as both transport pathways and effective adsorption sites [66]. Moreover, the thermodynamic analysis by Wang et al. (2024) established that PFOA adsorption on biochar proceeds through spontaneous physical pore-filling, and Gibbs free energy changes ($\Delta G°$) show a range between -2.24 and -5.38 kJ/mol[67].

However, the physical nature of pore-filling also means that desorption can occur relatively easily when environmental conditions change, highlighting the importance of considering both adsorption and desorption characteristics in remediation applications.

4.2.6. Hydrogen Bonding

Hydrogen bonding contributes to PFAS adsorption through interactions between functional groups on the adsorbent and PFAS molecules. Hydrogen bonds can form between –COOH and –OH groups on adsorbents and oxygen atoms in PFAS anions. Fluorine atoms can also form hydrogen bonds with –OH groups of adsorbents[14]. Wang et al. (2021) showed that hydrogen bonding increased the binding strength of PFOA and PFOS to modified clays. PFOA formed hydrogen bonds between its carboxyl group and functional groups on clay carnitine, while PFOS bonded through hydrogen bonds between its sulfonate group and hydroxyls on organic amendments[13]. The studies indicate that the methyl group C–H bonds create hydrophobic interactions with PFOS, whereas the carbonyl groups C=O enable hydrogen bonding[52].

4.2.7. Host-Guest Complexation

The materials that scientists create from cyclodextrin use their ability to form host-guest inclusion complexes as a special method for achieving adsorption. The cyclodextrin types, including α, β, and γ, all possess hydrophobic inner cavities that can form protective shields around PFAS molecules, while β-cyclodextrin is the most effective because its cavity size matches the dimensions of multiple PFAS substances. The research conducted by Yang et al. (2020) demonstrated that both crosslinker functions and interactions with the β-CD guest molecule determine PFAS binding strength. The combined effects of electrostatic interactions arising from amino crosslinkers and the host-guest binding mechanism produced strong binding and high capacity; the amine-modified β-CD polymer demonstrated an affinity for PFOA that was 100 times that of GenX [49]. Choudhary et al. (2022) used molecular dynamics simulations to show how cyclodextrin interacts with PFAS using mechanistic details. The computational studies demonstrated that the adsorption mechanism comprises two components that operate in parallel: electrostatic attraction between protonated amino groups and the anionic PFAS headgroup, and

hydrophobic interactions between the fluorinated tail and the cyclodextrin cavity. The superior performance of amino-functionalized cyclodextrin polymers stems from their binding mechanism, which is more effective than that of single-mechanism adsorbents [68].

4.2.8. Inner-Sphere Complexation and Ligand Exchange

Metal-modified adsorbents remove PFAS by forming inner-sphere complexation bonds. Metal nanoparticle-grafted MWCNTs removed PFOA via inner-sphere complexation, hydrophobic interactions, and electrostatic attraction, according to research by Liu et al. (2018) [54].

In their 2014 research study, Zhang and colleagues discovered that PFOS binds with kaolinite through an initial rapid outer-sphere complex bond, which later transforms into a slow ligand exchange process that removes adsorbed H_2O. PFOS chemisorption to clay mineral surfaces occurs through specific bonding with sulfonate groups to hydroxyl groups while also allowing hydrophobic and electrostatic forces to bind nonspecifically. Researchers have established that effective adsorbent design requires multiple interaction mechanisms to function properly[69]. Recent research has emphasized the importance of designing adsorbents that integrate multiple interaction mechanisms. For example, in their study, He Y. et al. (2024) established that the combination of hydrophobic, fluorous, and electrostatic segments achieves optimal sorption capacity. The electrostatically modified porous aromatic frameworks showed 14.8 times improved PFOA adsorption performance compared to the non-quaternized parent material and 24.1 times better performance than activated carbons. The study found that commercial adsorbents (GAC and ion exchange resins) use hydrophobic and electrostatic interactions as their main removal mechanisms. In contrast, fluorous interactions have emerged as the most effective technique for short-chain PFAS removal [70]. Finally, scientists now recognize computational modeling as a valuable tool for studying how different mechanisms work together. The DFT calculations, together with molecular dynamics simulations, show that effective PFAS adsorbents function by simultaneously engaging multiple binding sites. In contrast, their binding performance depends on the solution chemistry, pH, ionic strength, and the presence of competing organic matter [12][13].

4.2.9. Multi-Mechanism Synergy

The majority of enhanced adsorbents function by employing several combined mechanisms that operate simultaneously. As confirmation, the study by Wu et al. (2018) showed that multilayer PFOS sorption on few-layered porous graphite (FPG) occurs through electrostatic attraction and hydrophobic interactions, as PFOS inserts between hydrophobic lamellae via intercalation and hydrophobic pores on the particle surface [56].

To conclude, the mechanism contributions depend on adsorbent attributes, including surface charge and functional groups, hydrophobicity and pore structure, and PFAS molecular weight, head group, and molecular

structure. The next generation of adsorbents needs to be designed through research into the mechanisms that govern interactions among different materials.

Adsorbent class	Modification strategy	PFAS compound	Capacity before (mg/g)	Capacity after (mg/g)	Enhancement factor	Key mechanisms	References
Activated carbon	Ball milling (physical modification)	PFOA, PFOS, PFBA, PFBS	Not explicitly quantified GAC removal (12% for PFOA, 0% for PFOS)	Colloidal Activated Carbon (CAC_{BM}) removal: Up to 89% for PFOS, 73% for PFOA, 55% for PFBS, 30% for PFBA	7.4x for PFOS and 6x for PFOA (based on removal %)	Increased surface area and microporosity; enhanced dispersion; electrostatic and hydrophobic interactions; intraparticle diffusion	[65][71]
Biochar	Magnetic modification (Fe_2O_3 nanoparticles, ~100 nm, pre-pyrolysis at 200°C)	PFOS	~20–30 mg/g (unmodified sugarcane bagasse BC at 200°C)	120.44 ± 12.37 mg/g	4–6x	Electrostatic interaction, ion exchange, hydrophilic groups, hemimicelle formation, pore filling, bilayer formation	[72]
	Post-pyrolysis thermal treatment (air oxidation at 400°C for 30 min)	PFBS	17–34 (various feedstocks at 800°C, 1st saturation)	40–170 (after 400°C treatment)	2–5x	Increased SSA and porosity, oxidative removal of tarry deposits, increased C/O and C/H ratios (enhanced hydrophobicity)	[73]
Clay minerals	Surfactant-modified montmorillonite	PFUnA, PFBS,	~5–10	~40–50	~5–8×	Hydrophobic surface	[74]

Adsorbent class	Modification strategy	PFAS compound	Capacity before (mg/g)	Capacity after (mg/g)	Enhancement factor	Key mechanisms	References
	(CTAC)	PFHxA, PFHpA, PFOA, PFOS, PFNA, PFDA, GenX, PFHxS				creation; ion exchange; interlayer expansion	
Chitosan	Surface quaternization (chemical modification)	PFOS; PFOA	PFOS: 12.3 mg/g, PFOA: 10.1 mg/g	PFOS: 38.7 mg/g, PFOA: 32.4 mg/g	~3.1x for PFOS, ~3.2x for PFOA	Enhanced electrostatic attraction, increased surface charge density, improved hydrophilicity, and regenerability	[75][76]
MOFs	Post-synthetic modification (PSM) vs. preassembled (PAM)	PFOA	493 (PAM)	783 (PSM)	1.59x	Increased pore accessibility; Lewis acid–base interactions; electrostatic attraction	[77]
Graphene oxides	Cationic surfactant grafting (GO-CTAC); polyamine adlayer coating (PAGO); amine functionalization (EA, DETA, HMT)	PFBA, PFBS, PFHxA, PFHpA, PFOA, PFOS, PFNA, PFDA, GenX, 6:2	GO baseline: 50–90% removal after 48 h; adsorption capacity ~75 mg/g (PFOA)	GO-CTAC: ~100% removal for 11 PFAS in 1 h; PAGO: 2180–3070 mg/g (PFOA/PFOS), 602 mg/g	GO-CTAC: ~10× faster kinetics; PAGO: **30–40× capacity increase** vs. GO	Electrostatic attraction (quaternary ammonium or protonated amines), hydrophobic interactions, hydrogen	[40][55]

Adsorbent class	Modification strategy	PFAS compound	Capacity before (mg/g)	Capacity after (mg/g)	Enhancement factor	Key mechanisms	References
		FTSA, PFPrA		(PFBA), 220 mg/g (PFPrA)		bonding, F–F interactions	
Ion exchange resins	Quaternary ammonium functionalization (MIEX® GOLD); cross-linked DADMAC/BisAAm polymer beads (DR4); porogen-enhanced porosity; strong-base anion exchange	PFBA, PFBS, PFHxA, PFHxS, PFOA, PFOS	MIEX® GOLD: >99% removal at 1 ppm; capacity ~1.01–1.05 g/g (PFHxS, PFOA)	DR4: **3300 mg/g** (PFOA); MIEX® GOLD: **1.01–1.05 g/g**; >99.9% removal for all PFAS at 50 BV	DR4: ~30× higher than commercial resins; MIEX® GOLD: ~10× higher than many AERs	Strong electrostatic attraction; ion exchange with Cl^-; hydrogen bonding (DR4); hydrophobic interactions	[78][79]
Cyclodextrin polymer	Covalent grafting to graphene oxide (GO-Poly-BCD); crosslinking with benzyl chloride, HDI, or EPI	PFBA, PFPeA, PFHxS, PFHpA, PFOA, PFOS, PFDA, PFUdA, PFOSA, N-MeFOSAA, N-EtFOSAA	GO baseline: ~1.3 µg/g; β-CD-EPI/HDI: ~10–20% removal; β-CD-Cl: up to 75% PFOA	GO-Poly-BCD: up to 4.0 µg/g; β-CD-Cl: log Ka up to 3.3 L/g	~3× (GO-Poly-BCD vs. GO); ~2–6× (β-CD-Cl vs. β-CD-EPI/HDI)	Hydrophobic interactions, van der Waals forces, host–guest inclusion, electrostatic attraction (amine groups)	[80][81]
Molecular Imprinted Polymer	Electropolymerization of o-phenylenediamine (o-PD) with PFOS template on hydrophobic gold; bulk/surface imprinting using AAM, TFMAA, 4-VPy, chitosan, or	PFOS, PFOA, PFHxS, PFNA, PFTeDA	NIP baseline: <1–5 mg/g	PFOS: up to 1455 mg/g; PFOA: 5.4–12.4 mg/g	~10× to >1000×	Selective cavity recognition; electrostatic attraction (PFAS anion ↔ protonated monomers); hydrogen bonding; fluorophilic and	[82][83]

Adsorbent class	Modification strategy	PFAS compound	Capacity before (mg/g)	Capacity after (mg/g)	Enhancement factor	Key mechanisms	References
	DADMAC-based matrices					hydrophobic interactions; surface imprinting enhances accessibility	

Table 2. Performance Enhancement Through Surface Modification

4.3. Regeneration and Reusability

The ideal regeneration technique enables complete PFAS recovery via desorption while restoring the adsorbent's capacity for PFAS adsorption without damaging its physical or surface properties. Therefore, the development of optimized regeneration strategies is a fundamental requirement for operational PFAS adsorption systems. Furthermore, surface modifications enhance an adsorbent's initial adsorption capacity, but they also affect the effectiveness of regeneration processes and the durability of adsorbents across multiple usage cycles. The selection of a modification method establishes the primary binding pathways and the most effective regeneration procedure; therefore, combining modification design with regeneration planning becomes essential for real-world implementation[1][2].

4.3.1. Role of surface modification on regeneration performance

Surface modifications in PFAS remediation work to improve adsorption capacity but create obstacles to regeneration processes. Modified adsorbents, which rely on strong electrostatic interactions through quaternized materials and amino-functionalized polymers, require different regeneration methods than those that rely on hydrophobic interactions. Recent studies show that the characteristics and strength of surface functional groups determine both the energy required for desorption and the success of regeneration [3][22].

For instance, adsorption materials modified with quaternary ammonium groups require salt solutions or pH-swing methods to break electrostatic bonds for efficient regeneration. Fluorinated modifications, which use organic solvents to break fluorous-fluorous interactions, require specific regeneration methods that must be known for designing effective PFAS treatment systems [12][13]. The regeneration process for modified biochar and activated carbon materials improves structural integrity because their modified forms provide superior

mechanical stability compared to their unmodified counterparts [16]. The adsorbent's ability to withstand extreme regeneration conditions depends on the specific modification method used. Covalent modifications via grafted polymers and chemically bound functional groups provide greater resistance to regeneration cycles than electrostatically bound surfactants, which lose some material during regeneration [76].

4.3.2. Solvent-Based Regeneration

In several studies, researchers have employed methods to regenerate organic solvents during PFAS desorption. In 2021, Dong et al. achieved >98% desorption efficiency for imidazolium IL-modified clay using a methanol/water (50%/50%) mixture[22]. Du et al. (2016) showed that fluorinated montmorillonite (F-MT) could be completely regenerated through methanol washing, enabling five consecutive performance cycles without any decrease in efficiency [57]. In their 2018 study, Wu et al. achieved 85.3% regeneration efficiency for few-layered porous graphite using 50% ethanol at 40°C for 12 hours [56]. In addition, multi-component solvent systems have been identified as the most effective chemical regeneration approach in several recent comprehensive reviews, achieving >90% desorption efficiency compared to single-component solvents (12.5%), salt solutions (2.34%), or alkaline/acidic solutions (1.17%)[84]. The enhanced efficiency of multi-component systems is due to their capacity to concurrently disrupt multiple binding mechanisms that regulate PFAS adsorption on modified adsorbents [85]. For modified adsorbents, the organic solvent concentration and composition must be optimized based on the modification type. As confirmation, the study by Kashani et al. (2025) showed that quaternized chitosan hydrogels achieved more than 98% PFAS desorption over 10 cycles when tested with dilute NaCl solutions at 0.025 M because their surface-modification design reduced the need for chemical regeneration [76]. In contrast, Li et al. (2024) developed a stepwise methanol-acid regeneration train that operated by desorbing PFAS and co-adsorbed heavy metals from wood-derived biochar through two distinct processes, resulting in more than 90 percent pollutant recovery while allowing adsorbent reuse for three complete cycles [86].

Mixed solvent systems that combine organic solvents with salts exhibit enhanced regeneration. To confirm this, in 2021, Liu et al. conducted research in which a 1:1 (v/v) ethanol-1 mmol/L Na_2SO_4 mixed solution was used for Cu/F-rGa regeneration [12]. In general, the combined action of organic solvent molecules, which disrupt hydrophobic interactions, together with salt ions, which enable ion exchange, creates a synergistic effect in mixed solvents. The ion exchange process requires this dual-modification feature in adsorbents because both hydrophobic and charged surface groups are present.

4.3.3. Alkaline Regeneration

In the regeneration process, alkaline solutions are known to disrupt electrostatic binding and facilitate PFAS desorption. Yu et al. (2008) showed that chitosan-based MIP adsorbents retained their ability to remove PFOS at

full capacity through five adsorption-regeneration cycles, which used a 0.5 M NaOH/acetone (90/10, v/v) mixture at 40°C for 24 hours. The alkaline regeneration process destroyed chitosan's electrostatic binding but enabled complete regeneration without loss of performance [58].

The effectiveness of alkaline regeneration is particularly pronounced for adsorbents modified with amine or amino groups, where pH adjustment can protonate/deprotonate functional groups to reverse electrostatic attractions. However, excessive alkalinity or prolonged exposure can damage certain modifications, particularly those involving ester linkages or acid-labile crosslinks.

Based on previous work, it has been concluded that alkaline regeneration is most effective with adsorbents modified with amine or amino groups because pH changes enable functional groups to switch between their protonated and deprotonated states, thereby canceling electrostatic binding.

Furthermore, the research by Yang et al. (2020) demonstrated that amino-crosslinked β-cyclodextrin polymers preserved 89% of their original capacity after 6 cycles using a methanol/water (70/30) mixture. The polymers exhibited greater stability under conditions resembling harsh alkaline environments [49].

4.3.4. Salt Solution Regeneration

In salt solutions, high ionic strength can effectively desorb PFAS via ion-exchange mechanisms. Dong and co-workers (2021) demonstrated that both 50 mM and 500 mM NaCl solutions achieved over 98% desorption efficiency for ionic-liquid-modified adsorbents [22]. The optimal salt concentration depends on the strength of PFAS-adsorbent interactions. Salt-based regeneration works best for ion-exchange resins and quaternized materials, which bind PFAS via electrostatic interactions. Zhang et al. (2024) showed that quaternary ammonium-modified porous organic polymers with structural tunability achieved 90% PFAS removal efficiency after 5 regeneration cycles using a 10% NaCl–30% methanol mixture. The modification process improved both adsorption capacity and regeneration efficiency compared to unmodified polymers [85].

Surface modification plays a critical role in salt regeneration. The adsorption of heavily quaternized adsorbents requires stronger salt solutions and extended contact times because their electrostatic interactions with salts are stronger than those of less heavily quaternized adsorbents. Thus, the optimal salt concentration represents a trade-off between desorption efficiency and the preservation of the modification layer's structure [16][17].

4.3.5. Thermal Regeneration

High-temperature treatment is an effective method for the complete regeneration of adsorbents, as it decomposes adsorbed PFAS compounds.

Wu et al. (2018) achieved 99.8% regeneration efficiency by heating exhausted few-layered porous graphite to 800°C [56]. However, the process requires significant energy resources due to the degradation of the adsorbent

material after multiple usage cycles. To address this challenge, scientists have developed advanced thermal regeneration techniques that create specific temperature profiles that effectively break down PFAS while preserving the material's integrity. Thermal reactivation processes carried out at designated treatment centers, operating between 815 and 1200 degrees Celsius, can achieve over 99 percent elimination of PFAS contamination. However, these high temperatures can result in a 10 to 20 percent weight reduction of activated carbon and may alter its surface chemical properties. Additionally, modified adsorbents with organic functional groups attached via grafting are particularly susceptible to thermal degradation, as their modification layer begins to disintegrate at temperatures exceeding 400 degrees Celsius [84]. In addition, the implementation of temperature-control systems on thermally reactivated GAC significantly reduces its environmental impact over its lifespan compared with single-use GAC and regenerable ion-exchange systems. Operating at lower temperatures, starting at 815 degrees Celsius, reduces energy requirements while still achieving effective regeneration. The sustainability benefits of this system hinge on two primary factors: the transportation distance of materials to centralized reactivation plants and the frequency of regeneration processes [87].

4.3.6. Hydrothermal Alkaline Treatment (HALT)

The hydrothermal alkaline treatment (HALT) regeneration technology offers an innovative solution for effectively removing PFAS contamination while simultaneously restoring adsorbent materials. This method employs a strong alkali and operates at near-critical conditions (e.g., 350°C, 16.5 MPa, 1 M NaOH) to achieve complete PFAS mineralization. A study conducted by Soker et al. in 2023 demonstrated that HALT resulted in over 99% destruction of PFOS absorbed by granular activated carbon, achieving a defluorination rate of 96 ± 4% with no fluoroorganic intermediates detected. Additionally, the process operates at significantly lower temperatures than conventional thermal regeneration (350°C versus 815–1200°C), resulting in energy savings while providing the dual benefits of PFAS destruction and adsorbent regeneration [88].

The thermal and chemical stability of surface modifications under alkaline hydrothermal conditions determines how HALT works for modified adsorbents. Carbon-based adsorbents exhibit good HALT tolerance, although their organic polymer, surfactant, and metal nanoparticle modifications will undergo partial degradation during the treatment. Research is needed to evaluate how different modification strategies interact with HALT to identify the optimal combination of modification and regeneration methods [19][20].

4.3.7. Electrochemical Regeneration

Electrochemical methods provide an effective means to regenerate adsorbents by restoring their operational capacity. Liu et al. (2021) demonstrated electrosorption-desorption of Cu/F-rGa by applying a reverse voltage, yielding a 75.5% desorption efficiency [12]. This method enables direct regeneration without introducing chemical substances into the system. The method demonstrates promise as an efficient regeneration solution that requires

less than 1 kWh per regeneration cycle, uses only a few chemical reagents, and enables in situ regeneration. The process requires either anodic or cathodic polarization to change surface charge or create reactive materials that support PFAS desorption. Electro-regeneration achieves optimal performance when used with modified adsorbents containing conductive materials, such as metal nanoparticles and graphene. Recent studies have investigated electro-regeneration for PFAS-contaminated adsorbents and found that ion-exchange resins and electroconductive carbon materials achieved the greatest success. The technique works best on modified adsorbents with surface charges that can be adjusted electrochemically. The method requires non-conducting modifications to achieve its full functionality, while co-contaminants pose obstacles to electrochemical methods [89]. The process of coupled electrochemical degradation purifies PFAS contaminants in regeneration brine through an integrated system. Researchers achieved over 99% PFOA degradation using boron-doped diamond electrodes within 3 hours, with 84% defluorination. This method enables researchers to eliminate PFAS contamination in concentrated regeneration waste streams through waste treatment [21][31].

4.3.8. Regeneration Efficiency and Cycling Stability

Multiple studies have demonstrated excellent cycling stability for modified adsorbents:

MIP adsorbents: Maintained full capacity over five cycles[58]. Molecular imprinting ensures that binding sites remain structurally intact through multiple regeneration cycles, with 98.3% regeneration efficiency using NaOH/acetone mixtures [29][32].

Fluorinated montmorillonite: No performance reduction over five cycles[57]. The covalent bonding of fluorinated chains to clay surfaces provides exceptional stability, allowing complete regeneration with simple methanol washing [32].

PACF: Effective regeneration and reuse over multiple cycles[53]. Polyacrylonitrile-based activated carbon fibers maintain structural integrity during solvent regeneration, though capacity gradually decreases to ~85% by the fourth cycle[23].

Few-layered porous graphite: High regeneration efficiency maintained[56]. Both thermal (99.8% at 800°C) and solvent-based (85.3% with 50% ethanol) regeneration methods prove effective, with the thermal method showing superior but more energy-intensive results[25].

Quaternized chitosan hydrogels: Maintained >98% performance over 10 cycles using a dilute 0.025 M NaCl solution, demonstrating the exceptional regenerability of properly designed cationic modifications[76].

Amino-crosslinked β-CD polymers: Retained 89% capacity after 6 cycles with methanol/water (70/30) regeneration, showing the good long-term stability of host-guest complexation systems[49].

Porous organic polymers (POPs): Maintained 90% PFAS removal through 5 cycles using 10% NaCl + 30% methanol, demonstrating that rigid polymer frameworks preserve structure better than flexible materials [85].

Wood-derived biochar: Achieved >90% pollutant recovery for at least 3 cycles using stepwise methanol-acid regeneration, though capacity gradually declined due to irreversible pore blocking[86].

Graphene oxide-CTAC: Rapid adsorption kinetics (~30 minutes) were maintained through multiple cycles, though regeneration efficiency declined from 95% (1st cycle) to 78% (5th cycle) due to gradual surfactant loss[40].

4.3.9. Impact of Surface Modification on Long-Term Stability

The stability of surface modifications during regeneration processes is essential for modified adsorbents to maintain long-term performance across multiple adsorption-regeneration cycles. The stability of modifications depends on several essential factors, which include:

Chemical Stability: Covalently grafted modifications (e.g., ATRP-grafted polymers, silanized surfaces) demonstrate superior stability compared to physically adsorbed modifications (e.g., surfactant coatings). Kashani et al. (2025) demonstrated that covalently cross-linked quaternized chitosan retained 98% of its original capacity after 10 cycles. In contrast, clays with physically adsorbed surfactants typically experience a 10–20 percent capacity loss after 5 cycles due to ongoing surfactant desorption during regeneration [24].

Thermal Stability: Organic modifications become incompatible with high-temperature regeneration, which exceeds 400°C. Metal oxide modifications and fluorinated surfaces exhibit better thermal stability. Still, even these materials may undergo phase changes and sintering when exposed to standard thermal reactivation temperatures, which range from 815 to 1200°C [25][34].

Mechanical Stability: The handling and regeneration processes that follow ball-milling create abrasive conditions that degrade the physical properties of all modified materials. The handling of ball-milled activated carbon yields better regeneration performance than granular forms because this process creates more favorable mass-transfer conditions and reduces diffusion limitations [65].

Structural Integrity: Porous structures face risks of damage through their exposure to extreme conditions during regeneration processes. Research findings demonstrate that activated carbon micropore volume declines by 5% to 15% after five regeneration cycles. In contrast, modified carbons exhibit better preservation, with only 8% to 12% loss because their modification layers provide structural strength [35].

4.3.10. Sustainability and Techno-Economic Considerations

The efficiency of PFAS remediation systems depends on their ability to remove pollutants, the energy required for regeneration and chemical use, the production of resulting waste, and their overall environmental effects

throughout their operational life.

Energy Consumption: Thermal regeneration requires substantial energy, as it uses between 0.5 and 2.0 kilowatt-hours to process 1 kilogram of adsorbent. Chemical regeneration requires between 0.1 and 0.5 kilowatt-hours to process 1 kilogram of material, including solvent recovery. The electro-regeneration process requires the least energy, requiring less than 0.1 kilowatt-hours to produce 1 kilogram of output. The technology is not yet advanced enough for commercial use, and its current state of development makes it difficult to operate [87].

Chemical Requirements: The twelve-solvent system operates as a multi-component system, achieving more than 90% efficiency but generating waste streams that require treatment or disposal. The environmental burden of regeneration chemicals can offset the benefits of extended adsorbent life. The sustainability of modified adsorbents using salt-based regeneration systems can be enhanced because their chemical toxicity impacts are reduced, while their waste treatment becomes easier to manage [22].

Life-Cycle Analysis: The life-cycle assessment results indicate that regenerable ion exchange resins cause less environmental harm than single-use granular activated carbon. The thermal reactivation process requires GAC to operate within its optimal temperature range while also recycling its materials to achieve sustainability results that match or exceed the environmental impact of traditional methods. The environmental impact of modified adsorbents needs to be assessed by examining how their production process affects the performance of their regeneration system and their total operational lifespan [87].

Cost-Effectiveness: An economic analysis shows that regeneration is cost-effective when adsorbents reach their third to fifth cycle limits, with less than 20% performance degradation. The initial price of modified adsorbents becomes competitive when their regeneration efficiency exceeds 85% across multiple usage periods. The break-even point depends on adsorbent cost, regeneration method, PFAS concentrations, and treatment volume[39].

4.3.11. Challenges and Future Directions

The field of PFAS adsorbent regeneration has made substantial progress, but it still faces multiple obstacles.

Incomplete Desorption: The optimized regeneration process fails to recover complete capacity because certain PFAS molecules form permanent bonds with the material. This situation becomes most serious when materials undergo strong electrostatic or chemical-bonding modifications. The research path to solving this problem requires scientists to investigate pulsed regeneration methods and sequential multi-stage regeneration systems[47].

Modification Loss: The aggressive regeneration process will result in the removal of some physical modifications that require remodification over time. The research field is currently investigating the development of self-healing modification technology as its next focal point [48].

Co-contaminant Interference: Real-world PFAS applications require treating complex mixtures containing both organic and inorganic contaminants. Regeneration strategies need to develop either selective desorption methods or systems for treating mixed waste materials. The stepwise regeneration technique shows potential for the successful separation of different contaminant groups [90].

Scale-Up Challenges: The majority of regeneration research uses synthetic waters, which researchers test at the bench scale. Full-scale and pilot demonstrations are necessary because they establish real-world regeneration performance through testing with flow-through columns, variable water quality, and extended operation lasting over 100 cycles [91].

PFAS Destruction in Regeneration: Regeneration reduces PFAS material to smaller volumes, which makes disposal or destruction more difficult. The combination of advanced oxidation, electrochemical degradation, and HALT systems leads to complete PFAS mineralization, thereby completing the treatment process [49][51].

Regulatory Compliance: The regeneration processes must comply with hazardous waste handling regulations that protect worker safety and govern the discharge of regeneration waste streams. The use of modified adsorbents requires additional regulatory oversight when modification chemicals or byproducts pose toxicological risks [52].

The selection of a regeneration technique depends on the specific adsorbent used, the characteristics of PFAS substances, the costs involved, and the resulting environmental effects. Research must develop regeneration methods that achieve sustainability and cost efficiency while producing minimal secondary waste and sustaining adsorbent function during extended cycling periods. Furthermore, the process of developing new adsorbent materials requires designers to include all regeneration methods in their initial design work rather than treating them as a later consideration. The most effective PFAS remediation systems will use a comprehensive design that integrates modification chemistry, adsorption mechanisms, and regeneration methods, thereby enhancing performance, sustainability, and economic viability throughout the adsorbent material's entire lifespan [25][53].

Adsorbent type	Modification	Regeneration method	Efficiency (%)	Cycles tested	Capacity retention	References
Quaternary nitrogen	Quaternary nitrogen-grafted activated carbon (SNQ)	Ethanol	65	-	73%	[92]
Quaternary Ammonium Flax Fibers	Poly (METAC) grafting via ATRP	MeOH/1% NaCl solution	>78% PFBS >97% PFBA	**1**	>85% after 1 cycle	[93]
MIP-Chitosan	Molecular imprinting	NaOH/acetone (90/10)	98.3	7	95% after 7 cycles	[58]
Porous Organic Polymer (POP)	Structurally tunable quaternary ammonium	10% NaCl + 30% methanol	90%	5	90% PFAS removal maintained through 5 cycles	[85]
Graphene oxides (rGO)	Reduced graphene oxide-modified zinc ferrite-immobilized chitosan beads (rGO-ZF@CB)	0.1 M ethanol	80.35	5	Slightly altered	[94]
β-CD polymer	Amine cross-linked	Methanol/water (70/30)	93.6	6	89% after 6 cycles	[95]
Surface-modified quaternized chitosan hydrogels (MQCGs)	Surface-modified bio-based hydrogels derived from cross-linked quaternized chitosan (MQCGs)	0.025 M NaCl solution	>98%	10	Performance maintained >98%	[76]

Table 3. Regeneration Methods for Surface-Modified Adsorbents

5. Conclusion

This comprehensive review has systematically evaluated surface-modified adsorbents for PFAS remediation, focusing on modification strategies, adsorption mechanisms, and regeneration approaches. The evidence

demonstrates that surface modifications, such as amine functionalization, quaternization, surfactant coating, and fluorination, significantly enhance PFAS removal capacity and kinetics compared to conventional adsorbents. Enhanced performance arises from synergistic mechanisms that combine electrostatic interactions with anionic PFAS headgroups, hydrophobic interactions with perfluorinated tails, and emerging fluorous interactions, which provide superior selectivity, particularly for challenging short-chain PFAS compounds. Furthermore, modified adsorbents demonstrate excellent regeneration efficiency and cycling stability when appropriate methods are employed. Solvent-based regeneration achieves >90% desorption efficiency, while salt solutions and alkaline treatments effectively restore capacity with minimal chemical requirements. Advanced approaches, including hydrothermal alkaline treatment (HALT) and electrochemical regeneration, offer promising pathways for simultaneous PFAS destruction and adsorbent recovery, with properly designed materials maintaining 85–98% of their initial capacity over 5–10 cycles. On the other hand, despite significant progress, several critical challenges must be addressed for practical implementation. Future research should prioritize: [1] performance evaluation in complex real-world water matrices containing natural organic matter and competing ions, [2] development of scalable and cost-effective modification protocols suitable for large-scale production, [3] sustainable regeneration technologies that minimize energy consumption and secondary waste generation, [4] comprehensive techno-economic and life-cycle assessments to validate commercial viability, and [5] long-term stability studies under realistic operating conditions (>20 cycles). The stringent regulatory limits established by the U.S. EPA in 2024 (4 ppt for PFOA/PFOS) underscore the urgent need for advanced remediation technologies. Surface-modified adsorbents offer a promising pathway to meet these requirements while addressing emerging short-chain PFAS that evade conventional treatment. Successful next-generation adsorbents will integrate multifunctional surface modifications, hierarchical pore structures, robust regeneration compatibility, and sustainable synthesis from bio-derived precursors. Collaborative efforts between materials scientists, environmental engineers, and regulatory agencies will be essential to translate laboratory innovations into field-deployable technologies that provide scalable, cost-effective, and environmentally responsible solutions for PFAS contamination.

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