Journal of Water Process Engineering 63 (2024) 105471

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Structure and mechanism of nanoengineered membranes toward per- and polyfluoroalkyl substances (PFAS) removal from water: A critical review

Md. Nahid Pervez, Tao Jiang [*], Yanna Liang

Department of Environmental and Sustainable Engineering, University at Albany, State University of New York, Albany, New York 12222, USA

ARTICLE INFO

Keywords:
Per- and polyfluoroalkyl substances (PFAS)
Nanocomposite membrane
Electrospun nanofibers membrane
Adsorption
Degradation
Filtration

ABSTRACT

As a group of hazardous and bioaccumulative molecules, per- and polyfluoroalkyl substances (PFAS) threaten the health of ecosystems and humans. A growing body of research suggests that nanoengineered membranes can effectively remove PFAS from water media over conventional technologies. However, there has been a lack of discussion on the different types of membrane materials and the mechanisms involved in PFAS removal. This review thus seeks to fill the knowledge gap by critically assessing different membrane materials, particularly nanocomposites and electrospun nanofiber. Besides discussing the mechanisms underlying PFAS removal, this work also reviewed environmental and operational factors that can impact the efficiency of PFAS removal from water. In addition, future research directions were provided upon analyzing the challenges and opportunities in this membrane field.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals made intentionally, which are partially or fully fluorinated surfactants. They have been widely used in numerous industrial and consumer products, including cosmetics, lubricants, paper packaging, membranes, textiles, fabric finishing agents, and aqueous film-forming foams (AFFFs) because of their unique chemical and thermal properties (Fig. S1) [1,2]. These compounds possess a hydrophobic-lipophilic carbon chain that incorporates fluorine instead of hydrogen atoms in some or all carbon atoms. PFAS can be categorized into several classes and subclasses based on the functional groups at different positions and the length of the carbon chain [3,4]. There is a wide range of nearly 15,000 different types of PFAS, with perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) being the most frequently detected compounds [5,6]. Due to their widespread use, PFAS have often been found in drinking water samples around the globe, leading to an increased apprehension over their substantial risks to human health and the environment [7–9]. The U.S. Environmental Protection Agency (EPA) recently significantly lowered the lifetime health advisory threshold for PFOA in drinking water from 70 to 0.004 ng/L and PFOS from 70 to 0.02 ng/L [10]. Therefore, purifying PFAS-contaminated water from various sources on a global scale is an urgent need.

Given the potential risk of long-term exposure to PFAS in water, a number of treatment technologies have been documented for PFAS removal. Chemical degradation approaches, including sonochemical, electrochemical, photocatalytic, and advanced oxidation-reduction, may be useful for PFAS removal since they can transform PFAS into innocuous molecules [11]. However, when treating PFAS-rich landfill leachate and other complex matrices with high concentrations, slow reaction kinetics and water-matrix effects continue to be major obstacles to degradation [12]. Significant concerns include the high operational cost due to the large energy and chemical consumption and the potential secondary contamination by the degradation products [13]. Conventional methods for drinking water treatment, such as coagulation, flocculation, sedimentation, and sand filtration, are ineffective in removing PFAS (<20 %) from water, according to experiments conducted on a large scale [14]. Adsorption methods using adsorbent materials, such as activated carbons [15,16], ion exchange (IX) resins [17], and engineered clays [6,18], are frequently employed for removing PFAS. However, one major drawback of these traditional methods is that they may not be very effective for removing short-chain PFAS from water. Another issue is that they are expensive to handle and treat, especially when the adsorbents are regenerated or reactivated. Lastly, there is a concern about competitive adsorption with other ions and natural organic matter, the concentrations of which are often orders of magnitude higher than PFAS in polluted water [19–21]. These challenges have prompted researchers to explore efficient and cost-effective methods for

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[*] Corresponding author at: 1400 Washington Avenue, Albany, New York 12222, USA.
E-mail address: tjiang2@albany.edu (T. Jiang).

https://doi.org/10.1016/j.jwpe.2024.105471
Received 21 February 2024; Received in revised form 11 May 2024; Accepted 11 May 2024

their removal from water sources.

Membrane technologies have been recognized as efficient methods for treating wastewater from various sources worldwide because of their compact size, low energy consumption, small footprint, and high removal efficiency [22,23]. Subsequently, a significant focus has been on membrane-based filtration methods for removing PFAS from water. Among different approaches, high-pressure membrane filtration processes that utilize nanoporous membranes like nanofiltration (NF) and reverse osmosis (RO) have proven to be highly effective in separating both short- and long-chain PFAS compounds with rejection efficiencies typically exceeding 90 % [24,25]. The majority of commercial NF membranes have shown exceptional removal efficiency, ranging from 90 % to 99 %, for conventional PFOS and PFOA compounds. Literature reviews indicate that the NF method is less efficient in removing short-chain compounds than long-chain ones [26]. Conversely, there is a scarcity of literature documenting the removal of PFAS by RO compared to the abundance of research on NF. Both academic and industrial research have shown that the RO membrane method may achieve a removal rate of 96 % to almost 100 % for PFAS compounds with varying chain lengths and functional groups [27]. This removal rate is much greater than that achieved by NF membranes. The main reason is the comparatively low molecular weight cutoff (MWCO) of RO membranes, typically about 98–100 Da. However, although RO can remove PFAS to a higher degree, NF has the benefit of not requiring re-mineralization of the treated water. Both NF and RO face constraints associated with permeate flow and fouling influenced by the membrane's properties and the hydrodynamics inside the membrane module [28]. Among various constraints, fouling is the greatest obstacle in implementing NF and RO, although this may be mitigated with appropriate pre-treatment and membrane modification, thus leading to increased total capital expenditure and expenses associated with operation and maintenance [29]. Therefore, it is essential to develop effective membranes with less fouling and a high removal efficiency of PFAS using economical and eco-friendly methods.

There has been a notable rise in the commercialization of nanotechnology over the past decades. The nanotechnology-integrated membranes, referred to as “nanoengineered membranes” are superior to conventional membranes for water purification in terms of better permeability, reactivity, high efficiency, and antifouling properties [30,31]. These structures provide organized transportation channels and include several forms, such as nanopores spanning across, structural flaws, interlayer spacing in 2D laminar membranes, and intrinsic and regular pores in porous materials [32,33]. Accordingly, many efforts have been made to prepare nanoengineered membranes and apply them to remove PFAS from water. Nanoengineered membranes may be categorized into two groups: (1) nanocomposite membranes, which are prepared by mixing nanomaterials and polymeric support using common techniques, such as phase inversion, blending, and interfacial polymerization [34], and (2) electrospun nanofiber membranes, which are prepared using polymeric substances and nanomaterials with advanced properties through the electrospinning technology [35]. It has been demonstrated that the incorporation of nanomaterials into polymers could not only tune the structure and physicochemical properties (hydrophilicity, porosity, charge density, chemical, thermal, and mechanical stability) of membranes but also introduce multifunctionalities such as sorptive, catalytic, and filtration characteristics into the membranes, which is beneficial for PFAS removal. Besides, compared with conventional membranes, this type of membrane is less prone to fouling and offers more environmental benefits, a genuine attribute highly demanded for the efficient removal of PFAS on a large scale (Fig. 1a and b) [36].

A thorough critical evaluation of the removal performance of innovative nanoengineered membranes on PFAS has not yet been published, even though several publications have dealt with membrane technology for PFAS removal. This assessment would be especially helpful for differentiating these novel nanoengineered membranes from more traditional ones because of their multifunctional properties. A review of these new nanoengineered membranes designed to remove PFAS would

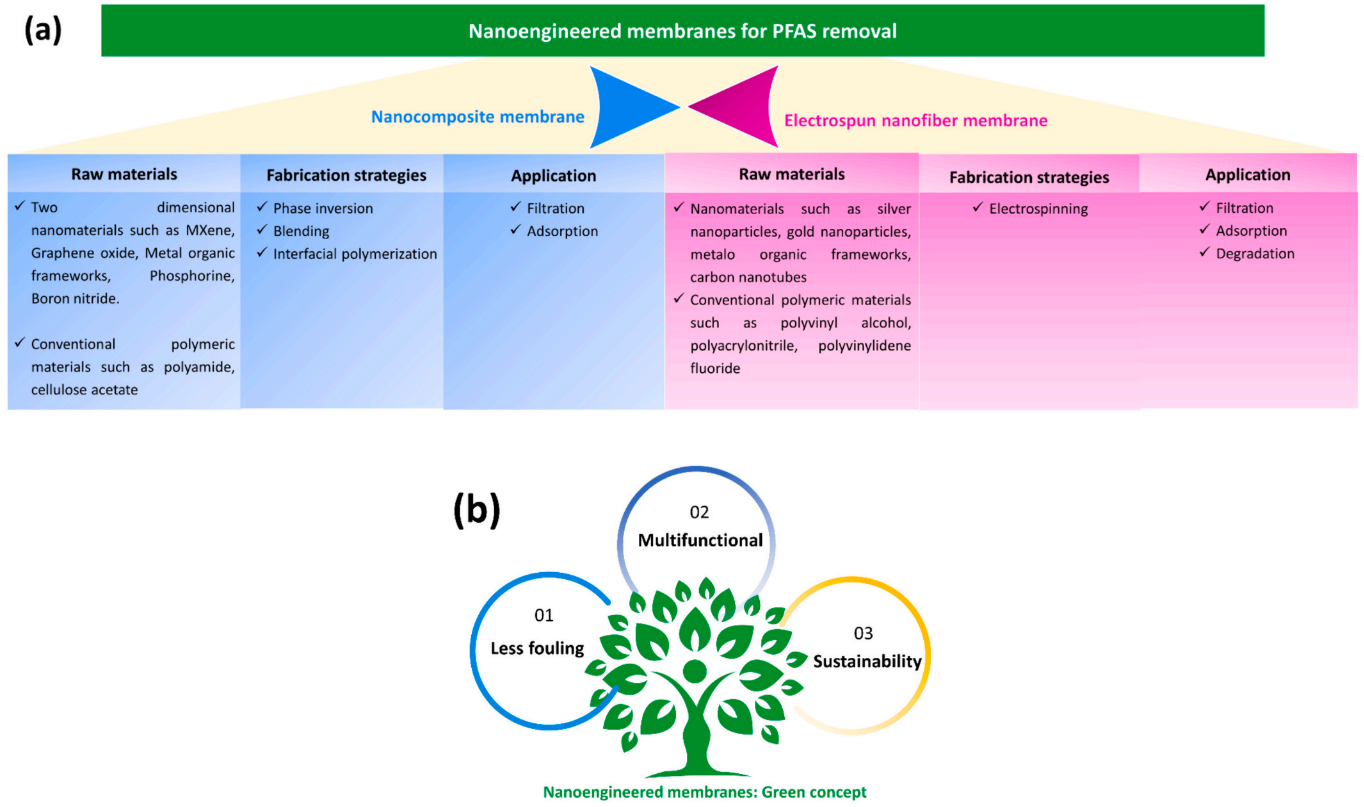

Fig. 1. (a) Features of nanoengineered membranes and (b) their benefits for PFAS removal.

benefit researchers interested in the continuous development of new membrane materials. Given these considerations, this paper aims to contribute to the existing body of literature by offering a critical analysis of nanoengineered membranes for PFAS removal, which is the first time reported in the literature to the best of the authors' knowledge. It accomplishes this by reviewing the principles and fabrication processes involved in nanoengineered membranes and influencing factors associated with PFAS removal processes. The challenges and future perspectives of using nanoengineered membranes for PFAS (Table S1) removal are discussed in detail.

2. Fundamental of nanoengineered membranes

2.1. Nanocomposite membranes

The production of nanocomposite membranes primarily relies on phase inversion (PI), blending, and interfacial polymerization techniques. Phase inversion procedures typically involve converting a polymer from a thin solution film into a solid matrix by subjecting it to a different gas/liquid phase or a temperature change. (Fig. 2a). To create a nanocomposite membrane using the PI method, the nanofillers are evenly distributed inside the polymer solution before the PI process takes place and are usually prepared in flat sheets or hollow fiber forms [37,38]. Blending is the most straightforward technique involving incorporating nanoscale components (such as metal nanoparticles) into polymers. A common solvent is used to disperse all components, including the polymer and nanoscale entities [39]. The nanocomposite membrane (Fig. 2b) was produced after the solvent evaporated, leaving the polymer chains intact [34]. The interfacial polymerization (IP) technique involves the dissolution of two distinct monomers in solvents incapable of mixing, forming a continuous polymer layer via chemical interaction (Fig. 2c). The nanoscale entities are introduced before the polymerization process. The polymerization occurs at the boundary between the two stages. The interfacial polymerization process forms a thin and distinct layer on the substrate, contributing to its high membrane flux [40]. Applying emerging IP techniques can potentially lead to redesigning nanocomposite membranes by varying their internal structure, nanofillers, reaction conditions, and surface coating (Fig. 2d). Furthermore, the new trends in the fabrication of nanocomposite membranes, including 3D printing, surface-initiated polymerization, and layer-by-layer assembly, hold enormous promises for developing nanocomposite membranes (Fig. S2).

2.2. Electrospun nanofibers membrane

Electrospinning is a promising and adaptable method for manufacturing continuous nanofiber membranes with diameters ranging from 20 nm to a few hundred nanometers, offering ease of use and high efficiency [42,43]. The fundamental components of an electrospinning setup consist of a syringe equipped with a nozzle, a power supply to generate the electric field, a reference electrode or a grounded object, and a pump (Fig. 3a). The electrospinning technique leverages the electrostatic repulsion forces inherent in a high-intensity electric field to generate nanofibers. A notable electric field is generated between the nozzle of the syringe and the counter electrode, which houses the solution intended for electrospinning. The divergence in electric potential between the ejector and the grounded target induces a deformation in the solution droplet at the nozzle, resulting in its expulsion in the form of a conical shape. The solvent inside the solution undergoes evaporation as the charged jet rapidly moves toward the counter electrode [44,45]. This process forms solid and uninterrupted nanofiber membranes on the grounded target, which are collected for PFAS removal. In addition, different types of electrospinning can be set up using needless, multi-needle, coaxial, and emulsion.

Importantly, several processing parameters (Fig. 3b), such as the solution concentration and solvent (viscosity, surface tension, and conductivity), operating conditions (flow rate, applied voltage, working distance, and needle gauge), and environmental conditions (temperature, relative humidity, and atmospheric pressure) must be carefully adjusted to produce an ideal electrospun nanofibers membrane for efficient PFAS removal.

3. Factors influencing PFAS removal by nanoengineered membranes

3.1. Membrane material

3.1.1. Nanocomposite membranes

The efficacy of PFAS removal may vary depending on the membrane material. So far, a range of two-dimensional (2D) nanomaterials have been incorporated into polymeric support for the preparation of nanocomposite membranes and applied to remove PFAS (Table 1). MXene-

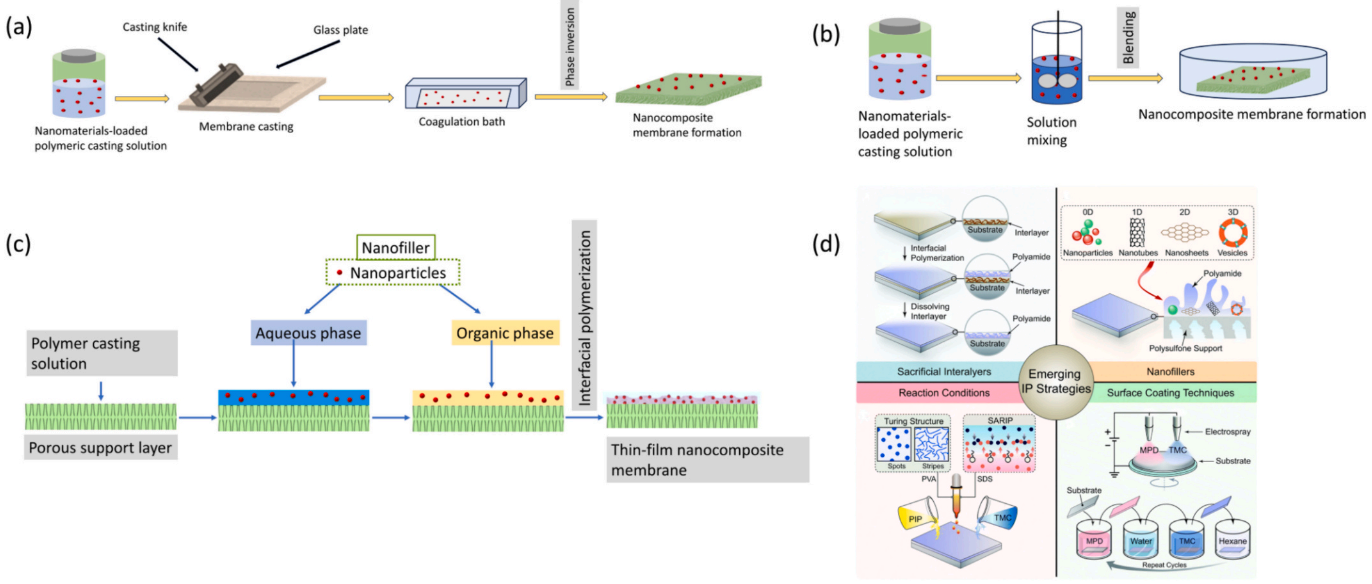

Fig. 2. Nanocomposite membrane fabrication types, (a) phase inversion, (b) blending, (c) interfacial polymerization, and (d) emerging IP strategies, reprinted from ref. [41] under the open access policy, Royal Society of Chemistry, 2021.

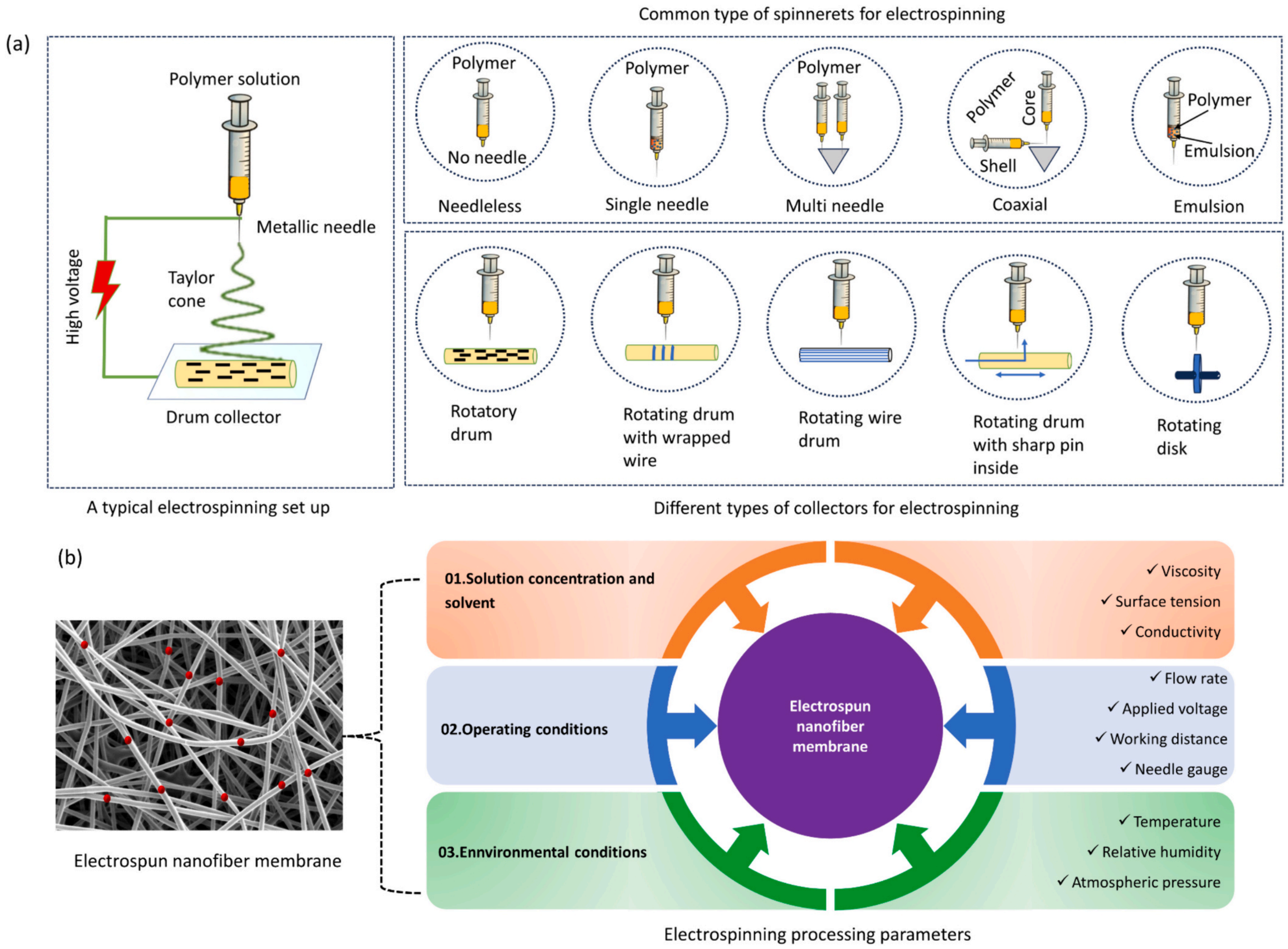

Fig. 3. (a) A typical electrospinning set up with different nanofiber membranes fabrication process and (b) processing parameters. Red circles represent the nanoparticles deposited on the membrane surface.

based thin-film nanocomposite membranes for PFOS removal through a nanofiltration module were prepared, and it was found that the introduction of MXene nanosheets influenced the selective layer characteristics by forming a lamellar structure of the membrane. It should be noted that the rejection efficiency for perfluorohexanesulfonic acid (PFHxS) and perfluorohexanoic acid (PFHxA) was 92–96 %, which is higher than that of the membrane (around 72 %) [46,47]. Significant interest has been noticed in using MOFs to develop high-selectivity nanocomposite membranes due to their varied and controlled topologies, uniform pore widths, and variable functional sites. In a recent research by Zhao et al. [48], a new capillary-assisted interfacial polymerization (CAIP) technique was used to produce nanocomposite membranes of MOF-PA. This regulation leads to the efficient exposure of MOF nanochannels on the surface of the membrane, as well as the creation of a PA matrix with a pronounced cross-linking gradient in the vertical direction. These factors collectively contribute to the exceptional selectivity of PFAS, surpassing a threshold of 95 %.

Previous studies have shown that GO membranes possess a high degree of hydrophilicity and a multilayer laminated structure, which enables efficient transport of water molecules while impeding the passage of other substances [49,50]. For example, an engineered GO membrane was developed to remove PFOS from water, and the rejection efficiency of PFOA increased by 70 % compared to the unmodified GO membrane [51], which may be ascribed to the increased resistance to diffusion resulting from the decreased width of the nanochannels in the modified GO membrane. Comparably, reduced graphene oxide (rGO) membranes have been synthesized and applied for PFOA removal [52]. The membrane exhibited a consistent rejection rate of 72 % for PFOA. Introducing amine functional groups onto hexagonal boron nitride (h-BN) significantly modified the electrostatic interactions between the BN (NH_2) and the fluorinated alkyl chains, thus leading to the potassium nonafluoro-1-butanesulfonate ($C_4F_9SO_3K$) rejection percentages of 93 % [53]. Phosphorene-modified sulfonated polyether ether ketone (SPEEK) was synthesized and achieved an average rejection rate of 99 % for PFOA [54]. The presence of organically modified montmorillonite (OMMT) enhanced the nanocomposite membrane performance of poly (*m*-phenylene isophthalamide) (PMIA), achieving a PFOS rejection efficiency of >95 % [55].

A recent study showed that the thin film nanocomposite membrane functionalized with ultra-fine activated carbon can be used for multifunctional purposes, namely filtration and adsorption for PFAS removal [56]. The resultant membranes with optimum loading showed a remarkable PFOS rejection effectiveness of 94 %, with over 80 % attributable to adsorption. The results provide insights into the interplay between adsorption and separation processes by nanocomposite membranes. However, many nanomaterials, such as MOFs, GO, MXene, etc., may be employed to enhance the performance of adsorptive nanocomposite membranes for PFAS removal due to their controllable chemical and physical properties. Adsorptive membranes integrate the functions of adsorption and filtration into a unified separation process. Compared to traditional filtration membranes, adsorptive membranes provide many benefits, such as better retention of contaminant ions,

Table 1
Effect of nanocomposite membrane characteristics on PFAS removal.

Membrane materials	Membrane module	Surface charge	Surface roughness (nm)	Surface wettability	PFAS compounds	PFAS concentration (μmol/L)	pH	Applied pressure (bar)	PFAS removal (%)	Ref
MXene-PA	NF	Negatively	82	Hydrophilic	PFHxS PFHxA	0.25; 0.32	10	3	96.85; 93.35	[47]
PMIA-OMMT	NF	Negatively	164	Hydrophilic	PFOS	0.20	7	5.5	>90	[55]
MXene-PA	NF	Negatively	85.4	Hydrophilic	PFOS	4.00	5.5	4.8	>96	[46]
Amine functionalized-BN	NF	Negatively	6	Hydrophilic	Potassium nonafluoro-1-butanesulfonate	295.70	7	6	>93	[53]
SPEEK-phosphorene	NF	Negatively	59.9	Hydrophilic	PFOA	241.51	7	2.06	99	[54]
MOF-PA	NF	Negatively	60.5	Hydrophilic	PFBA PFBS PFHpA PFHxS PFOA PFNA PFOS	0.093; 0.067; 0.055; 0.050; 0.0483; 0.043; 0.040;	6	5	56.2; 68.3; 67.4; 88.5; 80.6; 92.6; 98.9	[48]
MXene-CNT	NF	Negatively	32	Hydrophilic	PFOA	0.24	7	1	98.27	[58]
PDDA-GO	NF	Positively	–	Hydrophilic	PFOA	120.75	8	4.5	98	[51]
PSS-GO	NF	Negatively	–	Hydrophilic	PFOA	120.75	8	4.5	95	[51]
rGO	NF	Negatively	78	Hydrophilic	PFOA	2.42	3	3.44	90	[52]
AlFu-MOF	MD	Negatively	779	Hydrophilic	PFBS; PFPeS; PFHxS; PFHpS; PFOS; PFPeA; PFHxA; PFHpA; PFOA	0.070; 0.060; 0.053; 0.047; 0.042; 0.080; 0.067; 0.058; 0.050;	8.5	–	45.0; >94.4; 91.3; >96.8; 90.0; 0.0; 72.3; 88.3; 93.5; 70.5	[59]
PES-PA-activated carbon	Adsorption + filtration	Negatively	3372	Hydrophilic	PFOS	4.00	6	–	Adsorption capacity = 475 mg/g Rejection = 94	[56]

Note: Poly(diallyldimethylammonium chloride) (PDDA), Perfluorobutanoic acid (PFBA), Perfluorobutanesulfonic acid (PFBS), Perfluorononanoic acid (PFNA), Perfluoropentane sulfonic acid (PFPeS), Perfluoroheptane sulfonic acid (PFHpS), Perfluoropentanoic acid (PFPeA).

lower energy usage, and greater permeate flow [57]. Therefore, developing adsorptive nanocomposite membranes could be an effective way to remediate PFAS.

3.1.2. Electrospun nanofiber membranes

Various nanomaterials and polymers have been employed to fabricate electrospun nanofiber membranes for the efficient remediation of PFAS (Table 2). They are usually applied in the filtration, adsorption, and catalytic processes for removing PFAS. Electrospun membranes with porous architectures have the potential to effectively address the issue of PFAS through a gravity-driven filtration mechanism while requiring very low-pressure levels. For example, Wan et al. [60] synthesized ultrathin fibers by electrospinning, utilizing PVDF, and including quaternary ammonium moieties through grafting. A notable affinity toward the PFOA/PFOS compounds (PFOX) was detected, resulting in the removal of 97.9 ± 1.4 % of PFOX from synthetic groundwater. In addition, polydiallyldimethylammonium chloride (PDDA)-polyvinyl alcohol (PVA)-based composite electrospun nanofiber membranes demonstrated the rapid capturing ability for GenX (>97 %), PFOS (>99 %), and PFOA (>99 %) through the gravity-driven filtration with a tendency for regeneration [61]. Recent studies indicate that electrospun nanofiber membranes have favorable adsorption properties toward PFAS compounds. The adsorption isotherms revealed that the adsorption capacity of the MWCNTs-electrospun nanofiber membranes for PFOS (16.29 ± 0.26 μmol/g) exhibited an approximately 18-fold increase in comparison to the pure electrospun nanofiber membranes (0.92 ± 0.06 μmol/g) [62]. Composite electrospun nanofibers comprising fluorinated carbon nanotubes and silk fibroin were fabricated for the adsorption and extraction of PFAS [63]. The analysis of adsorption isotherms indicated that the primary mode of adsorption on the uniform surface of F-CNTs/SF was monolayer adsorption, exhibiting a notable affinity toward PFAS. This study presents a unique concept for developing adsorbent-mediated extraction routes targeting short-chain PFAS. Similarly, amidoxime surface-functionalized electrospun polyacrylonitrile (ASFPAN) had the highest GenX removal efficiency, reaching a maximum of 35 % [64], higher than the reported value of aminated PEGDA (0.0987 mmol/g) [65]. Furthermore, it is comparable to the established value of imine-linked 2D COFs (0.606 mmol/g) [66], the ionic fluorogel resin (0.84 mmol/g) [67], and GAC and PAC (0.79 mmol/g) [68]. Moreover, the removal percentage of GenX (around 72 %) was removed in the presence of an algae-based PAN electrospun nanofiber membrane [69]. Recently, a polyethylenimine-polyvinyl chloride (PEI-PVC)-based electrospun nanofiber membrane demonstrated an improved adsorption capacity for perfluorobutanoic acid (PFBA), perfluorobutanesulfonic acid (PFBS), PFOA, and PFOS. By combining a PEI functional group with a nanopore structure, the scientists were able to maximize electrostatic attraction, leading to impressive adsorption capabilities [70].

3.2. Membrane surface characteristics

The effectiveness of membranes is heavily influenced by factors such as surface characteristics, sub-layer morphology, and the thickness and compactness of the top layer [75]. The significance of surface roughness as a critical metric in evaluating the efficacy of nanocomposite membranes for removal efficiency has been established. The surface roughness of the membrane also significantly impacts its water permeability. An increased level of surface roughness might facilitate the deposition of pollutants on the membrane surface, whereas a significantly reduced roughness may hinder the passage of water molecules across the membrane [76]. In general, an increase in surface roughness is associated with a corresponding increase in the percentage of material removal. As seen in Table 1, membranes with greater surface roughness are associated with a larger proportion of PFAS removal. For instance, using a TFN membrane composed of MXene in combination with PA showed a significantly increased surface roughness of 85.4 nm. This enhanced surface roughness led to a remarkable PFAS rejection rate exceeding 96 %. In contrast, the bare TFC membrane exhibited a much lower surface roughness of 15.3 nm and a less pronounced trend of PFAS rejection, measuring 72 % [46]. Similarly, the surface of the MXene/CNT membrane exhibited more roughness than the pure MXene membrane. This roughness was shown to facilitate an augmentation in water penetration, resulting in a higher removal efficiency exceeding 90 % [58]. The oxygen-rich functional groups present in MXene demonstrated exceptional interfacial attraction and adhesion to polymer chains. Due to the heightened rate of IP reaction, the presence of the solution caused a delay in forming the PA layer. Additionally, the hydrophilic MXene facilitated the retention of water molecules during the IP reaction, creating a bubble-like nodular structure. Consequently, this process induced a modification in the surface roughness of the membrane [46,77]. In addition, it was shown that CAIP facilitated the formation of MOF-PA nanocomposite membranes with increased surface roughness. This characteristic was found to be advantageous as it led to enhanced water permeability and superior rejection ability, surpassing that of the TIP membrane with a rejection rate above 95 % [48,78].

3.3. Effect of electrospinning conditions

Besides, it has been observed that various variables influence the electrospinning process (Table 2). These factors directly impact the production of electrospun fibers that are smooth and free of beads and ultimately determine their performance for capturing pollutants [79]. Hence, in order to gain a more comprehensive comprehension of the electrospinning technology and the production of polymeric nanofiber membranes, it is essential to be aware of the impacts of all these controlling factors. As described in Table 2, each parameter is strongly interconnected with the electrospun nanofiber membrane's response (i.e., diameter). The diameter of fibers is of significant importance in the context of wastewater treatment using electrospun nanofiber membranes. In general, the reduction of pollutant levels is more effective with smaller-diameter composite polymer nanofiber membranes [80,81]. This is because membranes with smaller diameters possess a greater specific surface area relative to their volume than those with larger diameters. Consequently, the increased surface area provides more active sites for the adsorption of organic pollutants during the adsorption process [82]. The removal efficiency of PFAS was shown to be greater when using a smaller diameter electrospun nanofiber membrane. According to the data presented in Table 2, a quaternary ammonium grafted PVDF electrospun nanofiber membrane with a diameter of 99.9 nm exhibited a removal efficiency of 75 % for PFOA and 97 % for PFOS.

Similarly, the same type of membrane with a smaller diameter of 67.2 nm demonstrated a removal efficiency of 50 % for PFOA and 99 % for PFOS [60]. Mantripragada et al. [64] observed that introducing surface modification to PAN led to a marginal reduction in nanofiber size. Specifically, the modified PAN nanofiber membrane, ASFPAN10, exhibited an average diameter of 516 ± 48 nm. Furthermore, this modified membrane showed better efficiency in removing GenX, with a removal rate of 35 %. The pure electrospun PAN nanofiber membrane had a larger average diameter of 542 ± 66 nm, with a GenX removal rate below 20 %. Furthermore, in their study, Dai et al. [62] found that including MWCNTs in electrospun nanofiber membranes significantly increased specific surface area. Specifically, the specific surface area of the MWCNT-electrospun nanofiber membranes was observed to be twice that of the pure poly (D, L-lactic acid) (PDLLA) electrospun nanofiber membranes. This increase in specific surface area indicates that adding MWCNTs led to a reduction in membrane diameter and an enhancement in PFOS adsorption efficiency. In fact, the adsorption efficiency of PFOS was found to be >75 % in the presence of MWCNTs, while the absence of MWCNTs only allowed for the elimination of approximately 20 % of PFOS. According to the research, a decrease in the diameter of electrospun nanofibers results in an increase in specific

Table 2
Effect of electrospun nanofiber membrane characteristics on PFAS removal.

Membrane materials	Electrospinning conditions	Membrane diameter (nm)	Testing type	Surface charge	Surface wettability	PFAS compounds	PFAS concentration (μmol/L)	pH	PFAS removal (%)	Ref
PVDF-g-QA	• Voltage = 23 kV • 10 cm work distance • Flow rate = 0.15 mL/h (syringe pump) • Needle with an inner diameter of 0.337 mm • The relative humidity (RH) 60 %.	67.2 ± 27.3	Glass sand core filtration	Positively	Hydrophobic	PFOA; PFOS	0.12; 0.12	3.3 to 8.9	97.9 ± 1.4; 99.1 ± 0.4	[60]
Poly (D,L-lactide) (PDLLA)	• Voltage = 20 kV • 15 cm work distance • Flow rate = 1.5 mL/h (syringe pump) • Needle with an inner diameter of 0.8 mm. • The relative humidity (RH) 20 ± 2 %.	100 ± 20	Sorption	Negatively	Hydrophobic	PFOS	0.20	6	20	[62]
MWCNTs		100 ± 20	Sorption	Negatively	Hydrophobic	PFOS	0.20	6	>75	
Polyacrylonitrile nanofibrous	• Voltage = 18 kV • 15 cm work distance • Needle with an inner diameter of 1.22 mm.	542 ± 66	Filtration medium	Negatively	Hydrophilic	GenX	288.12	4	18	[64]
Amidoxime surface-functionalized electrospun polyacrylonitrile nanofibrous		516 ± 48	Filtration medium	Negatively	Hydrophilic	GenX	288.12	4	35	
Polyethylenimine-polyvinyl chloride	• Voltage = 25 kV • 15 cm work distance • Flow rate = 0.5 mL/h (syringe pump) • Needle with an inner diameter of 0.26 mm • The relative humidity (RH) 40 %.	200	Adsorption	Positively	Hydrophilic	PFBA; PFBS; PFOA; PFOS;	23.86–1168.01; 16.66–833.06; 12.08–603.76; 10.00–499.87	7	Adsorption capacities (qmax, 98.70 mg/g for PFBA, 222.36 mg/g for PFBS, 234.85 mg/g for PFOA, and 319.82 mg/g)	[70]
PVA/PDDA	• Voltage = 18 kV • 15 cm work distance • Flow rate = 0.18 mL/h (syringe pump).	–	Filtration	Positively	Hydrophilic	PFOA; PFOS;GenX	0.48; 0.40; 0.58	0.49 kPa	99; 99; 97	[61]
Ag/Au-PAA/PAH	• Voltage = 8.7 kV • 12 cm work distance • Flow rate = 0.04 mL/h (syringe pump).	–	Electrochemical oxidation	–	–	PFOA; PFOS	2.4×10^{-3}; 2.0×10^{-3}	7	72; 91	[71]
UiO-66-F4/ polyacrylonitrile	• Voltage = 16 kV	30–150	Adsorptive-Extraction	–	–	4:2 FTSA; 6:2 FTSA; FBSA;	3.05; 2.34; 3.34;	3	29; 38; 5;	[72]

(*continued on next page*)

Table 2 (*continued*)

Membrane materials	Electrospinning conditions	Membrane diameter (nm)	Testing type	Surface charge	Surface wettability	PFAS compounds	PFAS concentration (μmol/L)	pH	PFAS removal (%)	Ref
	• 20 cm work distance • Flow rate = 0.8 mL/h (syringe pump).					PFBS; PFHpS; PFHxS; PFOS; PFPeS; PFHpA; PFHxA; PFOA; PFUnA	3.33; 2.22; 2.50; 2.00; 2.86; 2.75; 3.18; 2.42; 1.77		20; 70; 60; 45; 72; 8; 7; 10; 30	
Lignin/PVA/bi-MOFs	• Voltage = 15 kV • 15 cm work distance • Needle with an inner diameter of 0.40 mm.	240 ± 12	Degradation under solar light	Negatively	Hydrophilic	PFOA	48.30	7	89.6 % of PFOA was degraded within 3 h	[73]
MIP Co/Fe@CNF	• Voltage = 14 kV • Needle with an inner diameter of 0.7 mm.	–	Electrochemical Degradation	Negatively	–	PFOA	48.30	4	89.7	[74]
Fluorinated carbon nanotubes/silk fibroin	• Voltage = 20 kV • 10 cm work distance • Flow rate = 0.01 mL/h (syringe pump) • Needle with an inner diameter of 0.26 mm. • The relative humidity (RH) lower than 50 %.	500–800	Adsorptive-Extraction	–	Hydrophilic	PFOA; PFOS; 6:6 PFPi	24.15–241.51; 19.99–199.95; 14.24–142.44	3	Adsorption capacities 28.48, 31.83 41.26 mg/g	[63]
PAN/Algae	• Voltage = 18 kV • 15 cm work distance • Needle with an inner diameter of 1.22 mm.	295 ± 23	Adsorption	Negatively	Hydrophobic	GenX	288.12	6	Pure PAN = 12 % Algae = 50 % PAN/Algae = 72 %	[69]

surface area, thereby enhancing the adsorption capacity [83].

3.4. Effect of pH

The pH can influence the removal rate by changing the electrical interaction between PFAS and the membrane surfaces via various functional groups. Nanocomposite membranes exhibited a positive correlation between the rejection of PFAS and the pH level [47]. The rejection of MXene-PA membranes for PFHxA and PFHxS was found to be greatly improved at a pH of 10, indicating that electrostatic repulsion played a prominent role as a mechanism under circumstances of high pH (Table 1). Conversely, when the pH value is <5, the surface of the membrane exhibits a positive charge. Consequently, the electrostatic attraction between the MXene-PA membrane and PFHxA or PFHxS was increased, thus reducing the rejection efficiency.

In typical wastewater, PFAS often occur as anionic species, which are negatively charged and fully dissociated in the aqueous environment. When the adsorbents possess a positive charge, an electrostatic attraction is established between them and the anionic PFAS molecules [84]. The pH of a solution substantially influences the surface charge potential, which, in turn, profoundly affects the adsorption capacity. The essential active sites for PFAS adsorption have been identified as the surface functional groups of adsorbents. Under circumstances of low pH, the functional groups mentioned may undergo protonation and transform into positively charged ions. These ions exhibit increased susceptibility to higher amounts of PFAS adsorption due to electrostatic attraction [85] (Table 2). As the pH of the solution rises, the functional groups tend to undergo deprotonation, resulting in a decrease in their positive charge and a potential acquisition of a negative charge. The surface attraction affinity may be diminished, or the negatively charged functional groups might induce repulsion of the PFAS anions by electrostatic forces [86].

3.5. Effect of PFAS concentration

The efficiency of membrane filtration and, by extension, the degree to which PFAS are removed is affected by the concentration of the PFAS solution. Initial PFAS concentrations may be different depending on the original PFAS source. The concentration of PFAS in the feed solution may have an impact on the ultimate efficacy of the removal process [87].

An example of this may be seen in the study conducted by Ma et al.

[47], where they observed a correlation between the rate of PFAS removal and varying concentrations. The impact of varying concentrations is evident in the membrane's removal efficiency, as shown by the minor rise in retention rates for PFHxS (from 95.5 % to 97.2 %) and PFHxA (from 93.07 % to 93.85 %) when the concentration was raised from 0.02 to 0.50 μmol/L and from 0.03 to 0.64 μmol/L, respectively. Evidently, the decline in acceptance of PFHxA and PFHxS exhibited a positive correlation with escalating concentrations, while the augmentation of PFHxS retention showed significant prominence. Likewise, it has been shown that the rejection of PFOS exhibited an upward trend as the concentration of PFOS rose across various trans-membrane pressures. The observed phenomenon may be attributed to the direct relationship between the initial concentration of PFOS in water and the adsorption of PFOS molecules onto the membrane surface. As the PFOS concentration increases, more PFOS molecules are adsorbed onto the membrane surface [11,88]. This leads to a reduction in the pore size of the membranes and an intensified screening action on the membrane surface. Consequently, the rejection performance of the membranes is enhanced [55]. Moreover, increased concentrations promoted the development of a cake layer on the membrane's surface. The inclusion of a cake layer may act as a barrier, increasing the probability of rejecting PFOA [58]. During the filtering process, coexisting molecules like humic acid (HA) and impurities in the feed may accumulate on the surface of the membrane, creating a layer of cake and increasing the barrier to the passage of PFOA.

3.6. Applied pressure

The use of pressure has been shown to be crucial in the membrane filtering process [89], as it facilitates the transportation of solutes onto the surface of the membrane and directly impacts the efficiency of PFAS compound removal. An observation was made regarding the rise in pressure from 3 bar to 6 bar, resulting in a reduction in the rejection of the MXene-PA membrane. Specifically, the rejection fell from 99.53 % to 96.94 % for PFHxS and from 94.98 % to 90.97 % for PFHxA. The reduction in PFAS removal may be attributed to the increasing dominance of pressure as the driving force [47]. In contrast, the study by Xu et al. [58] revealed minimal impact of pressure on PFOA rejection. This implies that the MXene/CNT membrane exhibits exceptional resistance to compaction and maintains steady separation performance, perhaps attributed to the presence of carbon nanotubes successfully mitigating excessive deformation and structural damage of the membrane.

On the one hand, the extensive use of NF/RO in PFOX remediation is limited by its relatively high energy requirements, requiring substantial operating pressure differentials to overcome natural osmotic pressure, as well as its poor water recovery and risk for membrane fouling [90–92]. Comparatively, electrospun nanofiber membranes are a viable option due to their capacity to customize the effectiveness of PFAS removal while operating in a gravity-driven membrane filtration (GDM) mode, which requires less energy. According to Wan et al. [60], the gravity-driven electrospun PVDF-g-QA membranes exhibited significantly higher efficiency in removing PFOX than the pressure-driven UF membranes with a MWCO of 10 kDa. Furthermore, the removal efficiency of the gravity-driven electrospun PVDF-g-QA membranes was comparable to that of the commercial NF 270 membranes, which achieved a PFOX removal rate of over 95 % [93].

3.7. Influences of membrane surface hydrophilicity/phobicity

Hydrophilicity or hydrophobicity are crucial features that significantly affect the membrane performance. The hydrophilicity or hydrophobicity of a membrane is determined by its level of attraction to water molecules on its surface [94]. Membranes with a hydrophobic surface have a higher susceptibility to fouling due to the accumulation and attachment of bacteria, proteins, and suspended particles. However, hydrophilic membranes resist the stated medium components, maintaining their retention-permeation efficiency [95].

As presented in Table 1, hydrophilic nanocomposite membranes have been used for PFAS removal in filtration systems. Several research studies have concentrated on improving nanocomposite membranes' hydrophilicity using different surface modification approaches to enhance their filtering efficiency [96,97]. For example, Le et al. [46] prepared a hydrophilic nanocomposite membrane and found that the addition of highly hydrophilic nanosheets into the PA layer enhanced the hydrophilicity of the membrane, resulting in enhanced water permeability and PFAS rejection efficiency [98]. All studied nanocomposite membranes exhibited hydrophilic surfaces, and PFAS removal efficiency was very high (i.e., above 90 %) (Table 1).

Similarly, as listed in Table 2, hydrophilic electrospun nanofiber membranes exhibited higher PFAS removal efficiency than their hydrophobic counterparts. Besides the hydrophilicity/phobicity, the charge of the membrane surface is important. For example, Dai et al. [62] prepared a hydrophobic nanofiber membrane with a negative surface charge and found only 20 % removal of PFOS, but the removal percentage increased to 99 % when the surface charge was positive [60]. Mantripragada et al. [64] used a hydrophilic nanofiber membrane with a negative surface charge and observed a mere 18 % elimination of GenX. A significant improvement in removal efficiency, reaching 97 %, was reported for a positively charged hydrophilic nanofiber membrane [61].

3.8. Influences of co-existing anions

The influence of co-existing anions is a crucial aspect to consider when assessing the suitability of nanoengineered membranes for the removal of PFAS. Numerous anions, including humic acid (HA), Cl^-, SO_4^{2-}, and PO_4^{3-}, are often encountered within the hydrosphere. These anions have the potential to engage in competitive interactions with PFAS on membrane surfaces [99,100]. It was reported that an increase in the rejection of PFHxA and PFHxS when introducing HA. Furthermore, they found a positive correlation between the concentration of HA and the rejection of these compounds [47]. As the concentration of HA grew from 0 to 15 mg/L, the rejection rate of PFHxA and PFHxS increased from 93.35 % to 95.48 % and from 96.85 % to 99.3 %, respectively. Moreover, Xu et al. [58] observed a significant increase in the rejection of PFOA when HA and Ca^{2+} were present, compared to the PFOA system in isolation. The presence of HA in the solution resulted in significant alterations in the ΔG_θ value, with more pronounced negative changes. This suggests that PFOA-Ca-HA compounds released a greater amount of energy compared to the other reactions. This observation indicates that the presence of HA and Ca^{2+} in the solution facilitated the combination of HA with Ca^{2+} and PFOA, resulting in a rise in the size of the compound. This explains why the rejection of PFOA increased when HA and Ca^{2+} were both present. According to the principle of sieving, larger-sized complexes have significant difficulty traversing the MXene/CNT membrane and are readily rejected. Furthermore, the simultaneous presence of opposing electrical charges brought by Ca^{2+} and HA results in charge screening, reducing the electrostatic repulsion between the membrane and HA and increasing the barrier to PFOA transit [26,101].

In another study, Luo et al. [55] observed that including phosphate (PO_4^{3-}) had the greatest impact on eliminating PFOS. The observed phenomenon may be ascribed to the heightened electronegativity of the membrane surface, resulting in an amplified electrostatic repulsion between the membrane surface and PFOS molecules. Furthermore, the rejection of PFOS has been seen to rise when the concentration of Pb^{2+} increases across all trans-membrane pressures. The observed enhancement may be ascribed to the formation of complexes between lead ions and PFOS molecules [55]. In the study involving PVDF-g-QA electrospun nanofiber membrane [60], it was observed that anionic organic competitors, such as macromolecule HA and FA substances, had a lesser detrimental effect on PFOX removal compared to anionic surfactant SDS (with a smaller molecular weight of 288 Da). This difference can be attributed to the lower adsorption affinity from the hydrophilic components in HA and FA substances [102].

3.9. *Effect of water metrics*

Various water matrices have been investigated to assess the practical efficacy of nanoengineered membranes in removing PFAS. Xu et al. [58] studied the efficacy of PFOA removal using a membrane composed of MXene and CNT. The researchers examined the membrane's performance in the presence of several genuine water samples, including settling water, charcoal water, and filtered water sourced from Changdang Lake waterworks in Changzhou, China. This experimental approach allowed for observing the membrane's practical use and effectiveness in purifying water. The results clearly indicate that the MXene/CNT membrane exhibited a removal efficiency of over 91 % for PFOA. Moreover, the membrane had a superior removal performance for PFOA in settling water samples compared to charcoal and filtered water. The observed outcome may be ascribed to the elevated concentration of organic matter in the settling water. PFOA readily forms a bigger complex coordination with organic matter due to the bridging action of inorganic salts, enhancing the rejection rate. The findings support using the MXene/CNT membrane to remove PFOA.

4. Removal mechanism

4.1. *Nanocomposite membranes*

Nanocomposite membranes efficiently eliminate both long-chain and short-chain PFAS from water and wastewater. Comprehending the PFAS removal mechanism of nanocomposite membranes is crucial to developing new nanocomposite membranes with enhanced qualities and performance. The primary mechanisms involved in nanofiltration using nanocomposite membranes include size exclusion, electrostatic exclusion, and the hydrophobic effect [19]. The size exclusion strategy prevails throughout all stages of the PFASs separation process. As the membrane's molecular weight cut-off (MWCO) decreases, the rejection rate increases due to steric hindrance, which excludes particular PFAS molecules with a very large molecular sizes (Fig. 4a) [103,104]. Another significant process leading to the elimination of PFAS using membranes with a charged surface, is electrostatic exclusion (Fig. 4b). The charged surface is created from the fabrication process which typically involves polymerization by reacting diamine and triacyl chloride at the interface. In this process, any unreacted acyl chloride groups can undergo hydrolysis to form carboxyl groups, which impart a negative charge to the membrane's surface. The Donnan effect suggests that negative charges on the membrane surface can cause repulsion between the anionic PFAS molecules, leading to their removal [105].

In addition, the surface hydrophobicity/hydrophilicity of the nanocomposite membranes can aid in the elimination of PFAS from water and wastewater. For example, nanocomposite nanofiltration membranes exhibit higher hydrophilicity in comparison to reverse osmosis membranes due to variations in their functional groups [47]. Conversely, PFAS are widely recognized for their hydrophobic characteristics, particularly long-chain PFAS, since they have longer hydrophobic tails in the C—F backbone. Hence, using more hydrophobic membranes can offer greater benefits in removing PFAS from water and

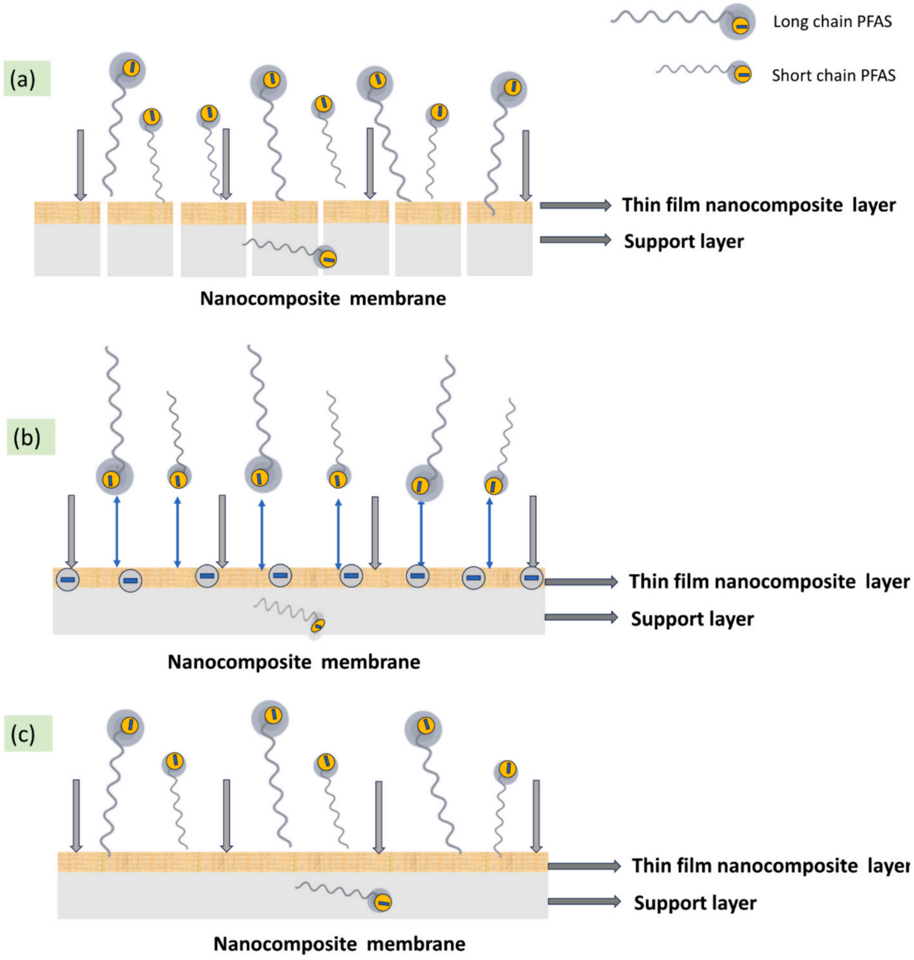

Fig. 4. PFAS removal mechanism, namely (a) size exclusion, (b) electrostatic exclusion, and (c) hydrophobic effect by nanocomposite membrane.

wastewater (Fig. 4c). Nevertheless, this could have an impact on the rate at which the permeate flows through [106].

On the other hand, adsorptive membranes have been observed to provide a potential method for in situ removal of PFAS with fast kinetics and membrane separation tendency. Recently, nanocomposite membranes have been used in both adsorption and filtration modes. It has been discovered that the removal mechanism is driven by hydrophobic-hydrophobic adsorption, which is facilitated by the C—F bonds of PFAS and the benzene rings of the nanofiller. Additionally, size exclusion and electrostatic repulsion play a role in the removal process [56].

4.2. *Electrospun nanofiber membranes*

The underlying mechanisms responsible for removing PFAS by electrospun nanofiber membranes can be analyzed from many perspectives. The strong electrostatic interaction between the positively charged nanofiber and negatively charged PFAS enhances adsorption, leading to improved adsorption efficacy (Fig. 5) [107]. The nanofiber membranes' surface charge and pore structure are the primary factors influencing this phenomenon. The nanofiber membranes' high porosity and interconnected structure contribute to their effectiveness in gravity-driven PFAS filtering, which is significantly more energy-efficient than pressure-driven membrane processes [70]. Quaternary ammonium grafting causes the membrane surface to become positively charged. This positive charge attracts ionized PFAS as an anion exchanger, resulting in improved adsorption capacity [60].

Nanofiber membranes have a multifunctional separation process encompassing screening, adsorption, and depth filtration [108]. Prior research has shown that adsorptive electrospun membranes are efficient in treating heavily contaminated industrial wastewater because of their ability to achieve many separation objectives [109]. Depth-type membranes containing nanofibers are highly porous materials densely packed with adsorbents [110,111]. These filters trap molecules both on the surface and inside the medium. The process of removing pollutants from liquids involves a combination of filtration or mechanical trapping and adsorption, which leads to an increased capacity for removal. It is worth noting that there are currently no studies available for the removal of PFAS using nanofiber membranes in a depth filtration method. Creating a multi-layer structure for nanofiber membranes has the potential to enhance the effectiveness of depth filtration in removing pollutants [112], such as PFAS.

Electrospun nanofiber membranes can also be utilized for the catalytic degradation of PFAS [74]. In this case, the nanofiber was coated with nanoparticles in order to mitigate agglomeration and enhance the number of reaction sites. The addition of oxidant (PMS) to the reaction system in the absence of light greatly enhanced the reaction rate at which PFAS was removed. This suggests that the transition metal ions in the bimetallic-supported nanofiber successfully stimulated PMS to generate many free radicals (Fig. 5) [73]. Furthermore, the investigation revealed that the mineralization efficiency was above 80 %, suggesting that the composite nanofiber membrane developed was highly effective in mineralizing PFAS.

Nevertheless, there may still be a need to process the effluent by employing nanofiltration as a post-treatment method to further remove degradation and mineralized products in order to release pollutant-free streams to the environment. Bifunctional photocatalytic nanofiltration membranes have emerged as a prominent area of research in the field of purifying water contaminated by micropollutants [113,114]. All of these findings provide compelling evidence that the catalytic membrane's dual functionalities can work simultaneously in a continuous permeation process, resulting in enhanced PFAS removal and effluent free of contaminants in practical situations.

5. Membrane fouling and scaling

Fouling and scalability are significant challenges in membrane technology that must be addressed to achieve practical implementation. Fouling develops when pollutants are deposited on the surface of a filtration membrane, impeding the passage of liquids through the membrane's pores. This phenomenon can be attributed to physical and chemical interactions or mechanical forces, leading to the reduction in size or obstruction of membrane pores (Fig. 6) [116].

While nanomaterials integrated into the membrane surface have been found to decrease membrane fouling, the use of nanoengineered membranes may enhance the membrane's ability to reject PFAS chemicals through an improved adsorption method [93]. Additionally, it might result in the accumulation of substances on the membrane surface and the formation of mineral deposits, which decreases the effective area of the selective layer. The occurrence of membrane fouling is directly influenced by the surface characteristics of the membranes [117]. For example, amphiphilic nature-based membranes exhibit surface fouling when used for PFAS filtering [118], but hydrophilic membranes display antifouling properties for PFAS even after recovery [119]. Additionally, fouling has been observed to be positively correlated with the concentration of PFAS. Increased PFAS concentrations may saturate membrane pores to capture PFAS molecules and effectively impede water flow. Reduced quantities of PFAS in the feed decrease the probability of PFAS molecules causing blockages [120]. Given the presence of PFAS with other pollutants and ions in the aquatic environment, examining the impact of their co-existence toward fouling is crucial. It was found that the presence of HA in the feed solution resulted in the formation of an organic cake layer on the surface of the membrane, which acted as an additional barrier, impeding the transit of PFAS. Also, the membrane surface's electronegativity increased, leading to a heightened electrostatic repulsion between the membrane and PFAS [47]. Another study tried to understand the reason for membrane

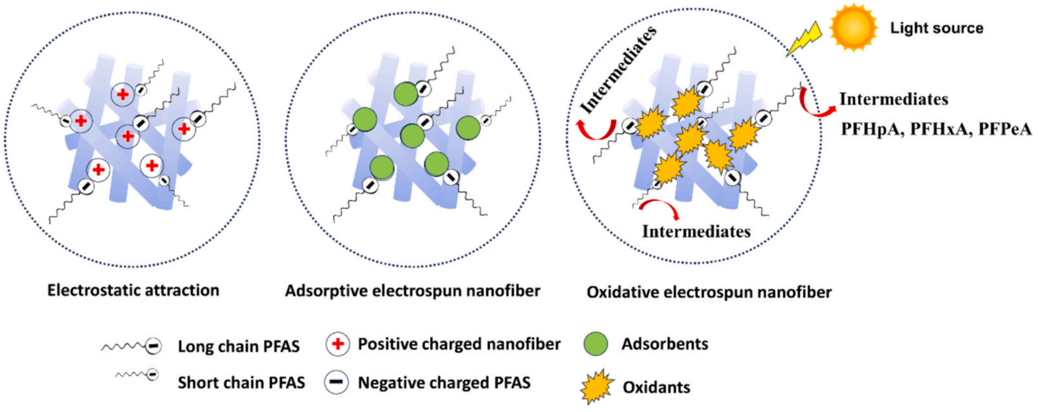

Fig. 5. Mechanisms of PFAS removal by electrospun nanofiber membrane [115].

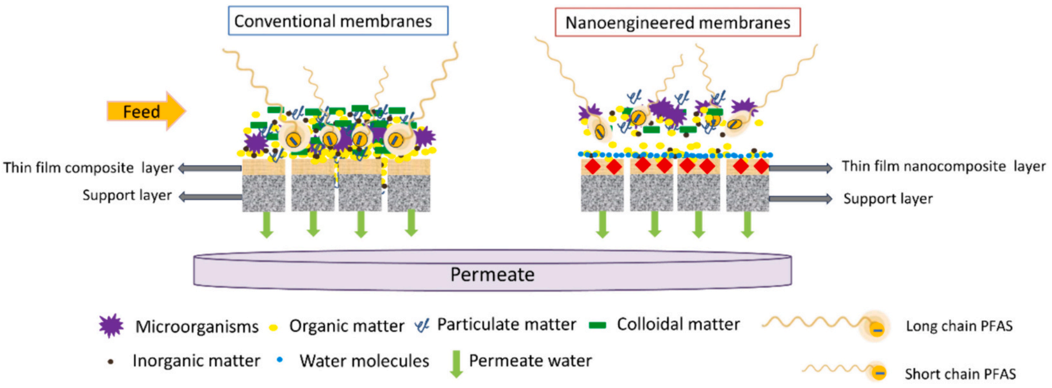

Fig. 6. Membrane fouling mechanism for PFAS removal in nanoengineered membranes.

surface fouling and found that upon the introduction of Ca^{2+} into the solution, the Ca^{2+} ions engaged in complexation/bridging interactions, resulting in the aggregation of one PFOA molecule and one HA molecule to produce greater flocs [58].

The abovementioned studies investigated the fouling characteristics on a small scale when PFAS are present. Nevertheless, further research is crucial to analyze the potential impacts of other contaminants on the removal of PFAS. One effective approach to reduce PFAS-associated membrane fouling is to enhance the negative charge density of the membrane [121,122]. This can be achieved by introducing carboxyl groups attached to amino groups on the polyamide through amide bonding. The negative charge from these groups acts as a charge shield, preventing PFAS molecules from coming into contact with the membrane surface due to the electrostatic repulsion between the negatively charged PFAS and the negatively charged membrane. Additionally, the utilization of gas-phase reactants for surface modification presents a compelling method to adjust the surface characteristics of the membrane with minimal fouling consequences [123].

6. Regeneration studies

Although adsorptive membranes are commonly used for PFAS removal, the primary limitation of their large-scale use is the need to regenerate spent membranes once their adsorption capacity has been depleted [11]. Typically, the regeneration stage involves applying a washing media to the exhausted membrane. Different research utilized different rinsing reagents, and ethanol and water were proven to be less efficient than methanol [56,61]. For example, the methanol rinsing method recovered 112.4 ± 11.8 % of PFOS and enabled the regenerated nanofibrous membrane to possess a high removal efficiency of 85 % for PFOS even after 20 regeneration cycles [124]. A recent study also showed that the pH-swing approach is highly successful for regenerating the membrane during methanol regeneration. The expansion of molecular brushes with a positive charge can offer numerous and easily reachable locations for electrostatic and hydrophobic interactions. Additionally, the higher concentration of OH- ions can shield the positive charge of the molecule, causing a transition from electrostatic attraction to repulsion (Fig. 7). Once the repulsion forces exceed the hydrophobic interactions, the adsorbed PFOA is released, leading to the regeneration of the membrane [122]. In general, it is necessary to conduct additional research on the regeneration of adsorptive membranes, which is frequently overlooked, using various solvents and innovative approaches to enhance the reusability of the membranes.

7. Challenges and future perspectives

Despite the advancements made in using nanoengineered membranes for PFAS removal, some obstacles must be overcome to expedite the real-world use of nanoengineered membrane technology. So far, nanocomposite membranes have been applied in filtration and adsorption modes, but the degradation study has not been reported. Accordingly, the development of catalytic/photocatalytic nanocomposite membranes for PFAS degradation is an important step for growing the membrane market. However, some challenges are observed, such as the activation of photocatalysts, light sources, and the leaching of catalysts from the membrane surface. Ideally, membranes should be photoreactive under sunlight irradiation with good quantum efficiencies and proper mass transfers. Additionally, the design of reactors with correctly illuminated submerged photocatalytic membranes is required. Achieving a uniform distribution of nanoparticles on the surface of polymeric membranes is a highly contentious issue in the development of nanocomposite membranes. Advanced oxidization has been revealed to be ineffective in fully degrading PFAS, producing shorter-chain PFAS that are more mobile. Membrane fouling is a prominent issue in

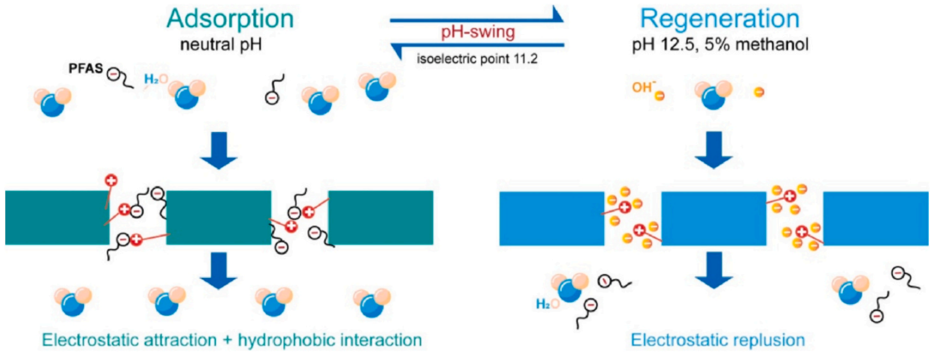

Fig. 7. Schematic diagram of pH-swing-based membrane regeneration. Reprinted from Ref. [122], Elsevier, 2024.

membrane processes, and it may have a detrimental effect on the efficacy of PFAS removal. Moreover, the performance of membranes for PFAS removal is reduced drastically in the presence of coexisting ions during the membrane filtration. Cleaning the membrane properly would be highly appropriate for reducing the fouling scale. Electrospinning has been used to produce precisely defined nanofiber membranes characterized by regulated orientation and size. However, the control and consistent manufacturing of these distinctive nanofibers remain a formidable challenge. The leaching of nanomaterials into aquatic ecosystems may transpire over the extended operation of nanoengineered membranes, during the production process of the membranes, and as a result of improper disposal of used membranes. The nanomaterials that are discharged may have the ability to undergo environmental transformations, leading to their uptake by different aquatic species. Consequently, a potential danger exists to both human health and environmental systems.

Considering the aforementioned issues, many potential future endeavors are suggested. Efforts to mitigate the leaching of nanomaterials would include using more resilient techniques for immobilizing the nanoparticles onto the membrane matrix, such as chemical grafting. There is a need to explore advanced fabrication solutions that are both scalable and cost-effective, in which additive (3D) manufacturing shows potential. Furthermore, it is essential to appropriately integrate operating parameters, PFAS features, and membrane properties with efficient methods for nanoengineered membrane systems. Enhancing electrospun nanofiber membranes' mechanical stability is critical for use in a high-pressure filtration system targeting PFAS. In this case, biodegradable polymers are not a good option. Using these membranes might potentially lead to the failure of the separation process at any given moment. Techno-economic analysis is crucial for using nanoengineered membrane technology on an industrial scale. The transfer from lab-scale to real-scale must be initiated to realize possible advantages, such as sustainability and energy usage. Therefore, these factors would be crucial in facilitating the advancement of nanoengineered membrane technology for PFAS removal.

8. Conclusions

This study critically evaluates the advancements made in the field of nanoengineered membrane applications for the removal of PFAS. Nanocomposite membranes have been shown to improve the effectiveness of PFAS removal through adsorption and filtration. On the other hand, electrospun nanofiber membranes have demonstrated superior removal efficiency of PFAS through a combination of adsorption, degradation, and filtering mechanisms. Various properties of nanoengineered membranes significantly influence the removal performance of PFAS. These properties encompass dimensions (size and shape), surface characteristics, composition, polymer type, and operational parameters such as PFAS concentration, applied pressure, co-existing anions, and water metrics. The investigation of the removal process indicated that many factors play a significant role, including electrostatic repulsion, attraction, hydrophobic contact, size exclusion, and the presence of reactive radicals. The long-term stability and performance of membranes that have been developed at the nanoscale need to be further investigated. Furthermore, comprehensive research on nanoengineered membranes' fouling scenarios and techno-economic analysis has been lacking. Future endeavors should be directed toward next-generation nanoengineered membranes that exhibit enhanced stability, superior performance, and scalability for remediating PFAS.

CRediT authorship contribution statement

Md. Nahid Pervez: Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Tao Jiang:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization. **Yanna Liang:** Writing – review & editing, Writing – original draft, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data sharing is not applicable as no new data were generated.

Acknowledgments

The authors acknowledge financial support from the US National Science Foundation (Award number CBET 2225596) and FuzeHub Jeff Lawrence Innovation Fund (Project number 2022-IC-0000000076).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jwpe.2024.105471.

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