Efficacy of Electrospun Nanofiber Membranes on Fouling Mitigation: A Review

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ABSTRACT: Despite the advantages of high contaminant removal, operational flexibility, and technical advancements offered, the undesirable fouling property of membranes limits their durability, thus posing restrictions on their usage. An enormous struggle is underway to conquer this major challenge. Most of the earlier reviews include the basic concepts of fouling and antifouling, with respect to particular separation processes such as ultrafiltration, nanofiltration, reverse osmosis and membrane bioreactors, graphene-based membranes, zwitterionic membranes, and so on. As per our knowledge, the importance of nanofiber membranes in challenging the fouling process has not been included in any record to date. Nanofibers with the ability to be embedded in any medium with a high surface to volume ratio play a key role in mitigating the fouling of membranes, and it is important for these studies to be critically analyzed and reported. Our Review hence intends to focus on nanofiber membranes developed with enhanced antifouling and biofouling properties with a brief introduction on fabrication processes and surface and chemical modifications. A summary on surface modifications of preformed nanofibers is given along with different nanofiller combinations used and blend fabrication with efficacy in wastewater treatment and antifouling abilities. In addition, future prospects and advancements are discussed.
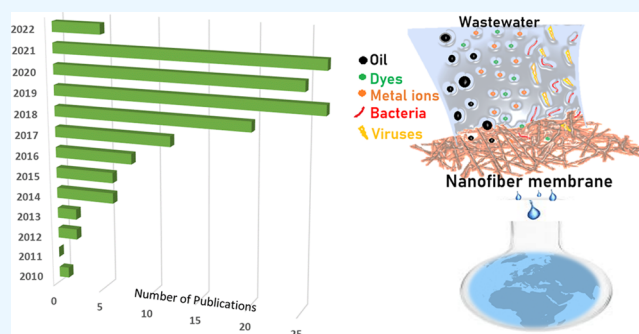
1. BRIEF INTRODUCTION TO THE FOULING OF MEMBRANES

A severe growth in population coupled with industrialization has amplified water contamination (by the release of dyes, heavy metals, and cyanides). In the last few decades, researchers have adopted membrane-based separation techniques to address the issues with respect to water and food processing applications.[1] Membrane filtration integrated with nanotechnology plays a key role in lowering water pollution either by removal of contaminants or by concentration of them. Compared to conventional treatment methods, it possesses significant advantages such as selective separation, simplified operation procedure, low space requirement, low production cost, and nonrequirement of chemicals for purification.[2] However, membrane technology suffers from one of the major challenges, membrane fouling, whereby the sludge particles or foulants become accumulated on the surface (cake formation) or pores of membranes.[3,4] Consequently, it adversely affects the productivity, selectivity, durability, and even life span of the membranes.[5]

Reliant on the type and strength of foulants on the membrane, fouling has been categorized into reversible (removable) and irreversible (irremovable) fouling.[6] Reversible membrane fouling arises when there is a weak interaction between the foulant and the membrane. On the other hand, irreversible fouling occurs when there is a strong attachment of the foulant on the membrane.[7] Reversible fouling can be removed by physical washing such as sponges, backwashing, or hydrodynamic scouring (surface washing), while irreversible fouling requires chemical treatment involving acid and base.

According to the nature of the foulants, membrane fouling is classified into three categories: biofouling, organic fouling, and inorganic fouling.[8] The first one, biofouling or microbial fouling, arises when bacteria/microorganisms accumulate on the surface or pores of the membrane. The soluble microbial product (SMP) or extracellular polymeric substances produced by accumulated bacteria strongly resist antibodies, thereby decreasing the membrane performance.[9] Biofouling can be either reversible or irreversible. Reversible biofouling originates

when microbes are loosely adhered to the membrane surface. Irreversible biofouling is observed when a biofilm has a strong affinity to the membrane. As stated earlier, reversible biofouling can be removed by physical or chemical means. However, irreversible biofouling is difficult to control owing to the formation of protective biofilm (EPS) on the surface of the membrane.[10] Researchers have adopted several strategies to curb biofouling and to improve the overall performance of the membrane.[11] The manipulation of bacterial communication systems or quorum sensing (QS) systems is reported to be one of the effective ways to reduce biofilm formation.[12]

The next category, organic fouling, refers to the deposition of organic matter on the surface or pore of a membrane by physical or chemical means and is considered to be the primary reason for chronic biofouling. The properties of organic matter such as size, molecular mass, polarity, functional group, etc. are found to play a prominent role in organic fouling.[13,14] It has been previously reported that the surface properties of membranes such as charge, roughness, and hydrophilicity play a crucial role in controlling organic fouling. The irreversible nature makes this type of fouling a difficult one to control and thus often requires severe chemical treatment methods.[15] This type of fouling resistance as found in NF (nanofiltration) membranes could be improved by the use of nanomaterials such as graphene oxide (GO), silica, carbon nanotubes (CNTs), titania, etc.[16] Abdikheibari et al.[17] improved the fouling resistance of the poly(piperazine amide) (PPA) membrane by loading it with amine-functionalized boron nitride $BN(NH_2)$ nanosheets. The fouling resistance of the PPA–BN membrane was evaluated by using cross-flow filtration experiments and contact angle and atomic force microscopy (AFM) analysis. Membranes with smooth surfaces tend to restrict the accumulation of foulant on the membrane surface, while repulsive interaction between the foulant and the membrane restricts the deposition of foulant on the surface of the membrane. The hydrophilic and smooth morphology of the PPA–BN membrane is reflected in AFM and contact angle measurements. Moreover, it possesses high negative charge density, which is desirable to repel the organic matter from the surface of the membrane. The repulsive interaction between the PPA–BN and NOM (natural organic matter) prevents the deposition of organic matter onto the membrane surface and reduces the organic fouling propensity. Ca^{2+} ion is reported to promote organic fouling by forming a network structure with alginate, an organic foulant. So, in their study, Li and his team[18] introduced a complexing agent, EDTA, to remove Ca^{2+} from the fouling layer. The complexation of EDTA with the Ca^{2+} ion leads to the destruction of alginate–Ca^{2+} gel networks, and subsequently the organic fouling of the FO membrane get reduced. Shao et. al[19] investigated the oil-in-water emulsion in the presence/absence of surfactants and organic foulants with three membranes. A superhydrophilic membrane may be fouled by surfactant and organic foulants. Sulaiman and co-workers developed a PVA/chitosan/TEOS hybrid membrane for the treatment of wastewater containing copper ions.[20] These membranes are resistant toward humic acid, an organic foulant. The hybrid composite possesses good physical flexibility and is used for the treatment of wastewater containing heavy metals.[19−22]

Inorganic fouling, the last category, describes the deposition or agglomeration of inorganic cations (Ca^{2+}, Al^{3+}, Mg^{2+}), anions (PO_4^{3-}, OH^-, SO_4^{2-}, CO_3^{2-}), salts ($CaCO_3$, $K_2NH_4PO_4$, $CaSO_4$), and metal hydroxides ($Ca(OH)_2$, $Mg(OH)_2$) on the surface of membranes, leading to inorganic fouling. Inorganic

fouling occurs by two main pathways, namely, crystallization and particulate fouling. The crystallization process refers to the process by which nuclei or crystals get deposited on the active site of membranes. Particulate fouling, on the other hand, refers to the deposition of colloidal particles. The factors such as ionic strength, pressure, pH, temperature, etc. are found to play an important role in inorganic fouling. Owing to the strong cohesive force between inorganic species and membranes, inorganic fouling is irreversible in nature, and hence it is difficult to remove by physical methods.[23] Shahid et al.[24] explored the scale inhibiting property of CO_2 on RO membranes by assessing the data obtained from the Ryznar stability index (RSI), salt rejection, mass balance, and morphological analysis. RSI is related to the scaling potential: an RSI value <6.5 indicates a greater probability for scale formation, while RSI values of 7 imply no probability of scale formation. The RSI value of RO membrane purged with CO_2 falls to around 7, indicating that it is effective in reducing scale formation on RO membranes. Salt rejection and mass balance data were complementary to RSI analysis and suggest that CO_2 is far superior to other anticipants. The absence of any scale deposition or crystal formation on the surface of CO_2-treated RO membrane as observed from AFM analysis further confirms the aforesaid result. In a recent study, Wan et al.[25] employed both acidic ($AlCl_3$) and basic coagulants ($NaAlO_2$) to remove calcium phosphate salt, which is mainly responsible for pressure-retarded osmosis (PRO) membrane fouling. Significant improvement in the water flux was observed for the membrane treated with coagulant. This is attributed to the removal of phosphate by the coagulation action of $AlCl_3$ and $NaAlO_2$. Their studies thus show that acid and caustic coagulants control the fouling on the PRO membrane. Mustafa et al.[26] investigated the antifouling ability of native and methyl-grafted NF membranes. The experimental result suggests that grafted membranes possess strong antifouling efficiency and diminish the fouling caused by organic (alginate) as well as inorganic foulants (iron and manganese salts). PRO is one of the advanced technologies that is used for water treatment purposes. Seawater desalination brine (SWBr) and wastewater brine (WWBr) are common feeds for PRO membranes. Han and co-workers[27] employed pH modification as well as antiscalants (EDTA and HEDP) to mitigate membrane fouling in PRO processes. The experimental results suggest that pH modification and antiscalant pretreatment suppress inorganic fouling by forming stable complexes with inorganic foulants such as Ca^{2+} and Mg^{2+}.

Traditional membranes and nonwoven membranes have limited pilot scale applications because of fouling. The major cause of fouling is rather surface roughness along with large and wide pore size.[28] Nonetheless, experimenting with nanofibers has shown to have a positive effect. In addition to low production cost, acceptable selectivity and great permeability are other foremost interesting factors of nanofibers.[29] The ability of nanofibers to be embedded in other mediums and their high surface to volume ratio are added advantages in fouling reduction.[30,31] A survey of the literature reveals the availability of review articles on the basics of membrane fouling,[4] fouling in membrane bioreactors,[32] fouling with respect to membrane distillation, fouling in ultrafiltration, nanofiltration, and reverse osmosis membranes,[33] and much more. A search of data on the Web of Science on May 20, 2022, with the keywords electrospun membrane and antifouling, provided an increased number of publications as well as citations with respect to the subject area (provided in Figure 1). However, fouling mitigation with a focus
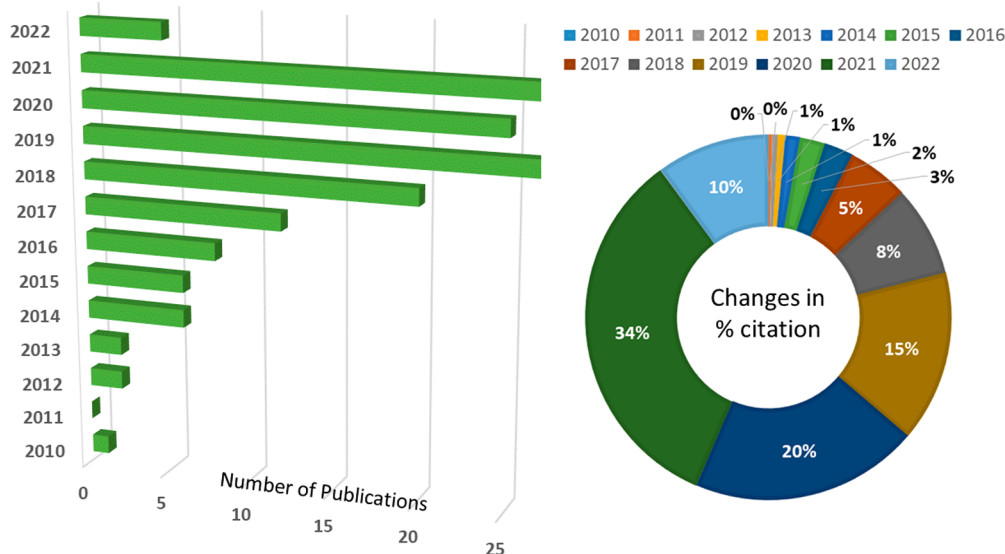
Figure 1. Web of Science search data results with the keywords "electrospun membrane" and "antifouling" on May 20, 2022, indicating an increased number of publications and percent citations from the year 2010.

on electrospun nanofibers is not unearthed. Herein, this Review briefs on the development of nanofiber membranes and elaborates on the strategies used in nanofiber membranes for fouling mitigation.

2. CONCISE BACKGROUND OF ELECTROSPUN NANOFIBER MEMBRANE FABRICATION

Electrospinning is a superior process to develop uniform fibers on the nano- and microscale. The process involves the stretching of a polymer material in a single direction in the presence of an electric field from a feeding jet, leading to the formation of stable, uniform, and continuous electrospun nanofibers. Typically, the electrospinning process requires an electric field induced by a high-voltage power supply between a polymer solution and a collector, and in the presence of a high field, the electrical force overcomes the surface tension of the polymer solution and contorts the drop at the spinneret tip into a "Taylor cone", followed by the expulsion of a thin charged jet.[34] The jet first undergoes a stable stretching followed by random whipping and bending, with an additional stretching contributed by charge repulsion and solvent evaporation. This unstable stretching leads to a very large spin draw ratio, where the terminal jet speed attains a portion of the sound speed, giving solidified fibers on the collector with a submicrometer diameter and a randomly oriented nonwoven structure. The features of the nanofibers produced by electrospinning, such as texture, morphology, shape, and diameter, depend on the inherent polymer properties, such as molecular weight and its distribution, viscosity, conformation of the polymer chain, conductivity, surface tension, pH, and solvent vapor pressure, and also operational conditions, such as power of the electric field, geometry of the electrode, distance between the spinneret and collector, rotating speed of the collector, and feeding rate of the polymer solution additionally surrounding the environment.[22,35]

The basic electrospinning instrument consists of four main parts, namely, glass syringe, metallic needle, power supply to generate the electric field, and metallic collector. The polymer solution is loaded into the syringe, which is attached to a needle to generate a jet. Electric voltage is applied between the needle and the collector to start the spinning process (when electric

charges migrate into the polymer solution through the needle). This induces instability in the polymer solution as a result of the introduction of charges on the polymer droplet. Simultaneously, the mutual repulsion of charges generates a force that resists the surface tension, leading to the flow of polymer solution in the direction of the electric field as shown in Figure 2. Further
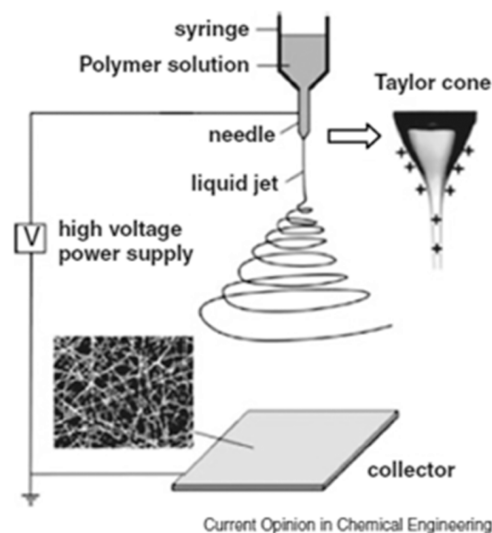

Figure 2. Schematic illustration of the electrospinning process. Reprinted with permission from ref 37. Copyright 2004 John Wiley and Sons.

enhancement in the electric field leads to distortion of the spherical droplet, giving it a conical shape (Taylor cone). Subsequently, the formation of ultrafine nanofibers takes place, and they are collected on the metallic collector. A stable charge jet can be produced when the polymer solution has adequate cohesive force, and during the process the inside and outside forces (of charge) cause the whipping (combined effect of solvent evaporation and charge repulsion) of the liquid jet toward the collector. This whipping motion makes the polymer chain stretch and slide, resulting in the generation of fibers with smaller diameters (nanofibers).[36]
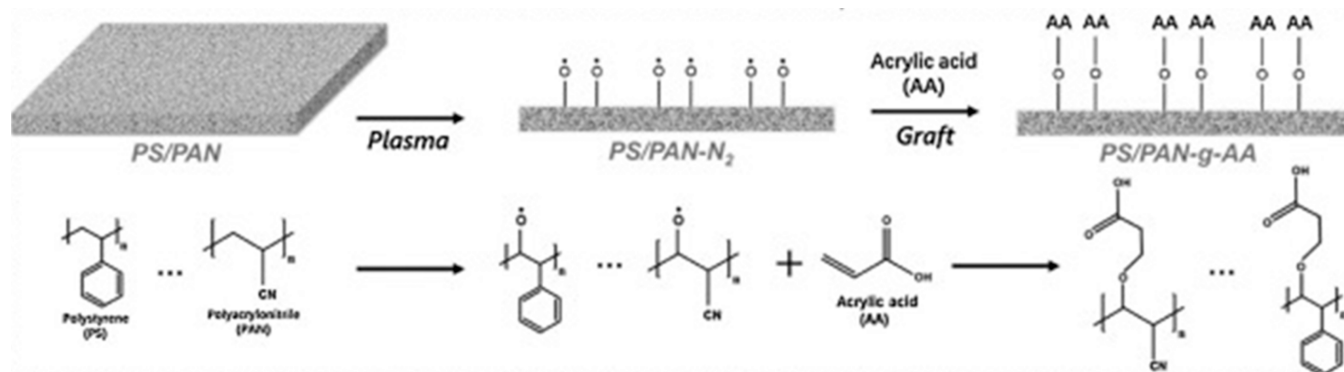
Figure 3. Schematic illustrating the reaction process of PS/PAN-*g*-AA nanofibrous membranes. Reprinted with permission from ref 48. Copyright 2019 Elsevier.
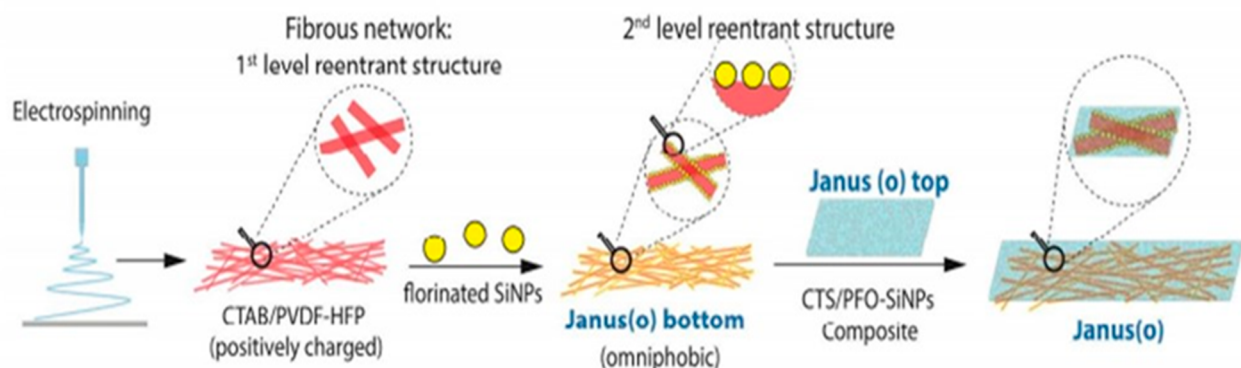

Figure 4. Fabrication procedure of the Janus(o) membrane. Reprinted with permission from ref 55. Copyright 2017 American Chemical Society.
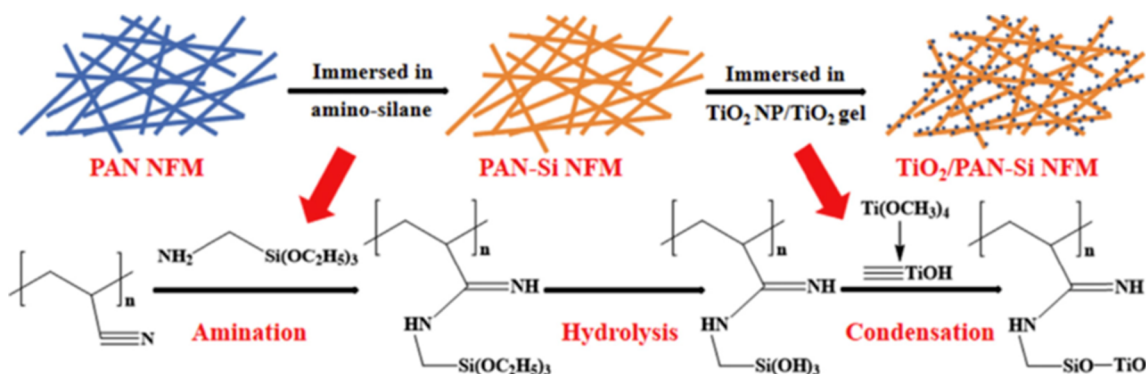

Figure 5. Schematic illustration of the preparation process and reaction principle of the TiO_2/PAN−Si NF membrane. Reprinted with permission from ref 62. Copyright 2019 Elsevier.

Even though the electrospinning method seems to be a simple process with easy adaptability, the major challenge lies in various processing conditions—optimization to produce nanofibers. The main processing conditions are applied voltage, concentration of the polymer, solution viscosity, solution flow rate, distance between the collector and the needle, needle diameter, humidity, temperature, and solvent and solution conductivity. Every polymer has its own critical value with respect to various experimental conditions mentioned above, and any deviation from its critical value adversely affects the formation of nanofibers. For example, an increase in the applied electric field or a flow rate beyond the critical value will lead to the formation of beads or beaded nanofibers.[38] The distance between the metallic needle and the collector differs with polymer solution,[39] and the distance influences the morphology because it is controlled by deposition time, rate of evaporation, and whipping or instability interval.[40] The spinning depends on the uniaxial stretching of the charged jet, which depends on the concentration of the polymer solution. An increase in the concentration of the solution above its critical point (optimum concentration at which beadless uniform nanofibers generate) results in defective or beaded nanofibers.[41] Solution conductivity helps in the formation of the Taylor cone and controlled diameter nanofibers. The lower conductivity solution will not have sufficient charge to form the Taylor cone, resulting in no electrospinning, and a solution with higher conductivity beyond the critical value will hinder Taylor cone formation and electrospinning. The solution with optimum (critical value)
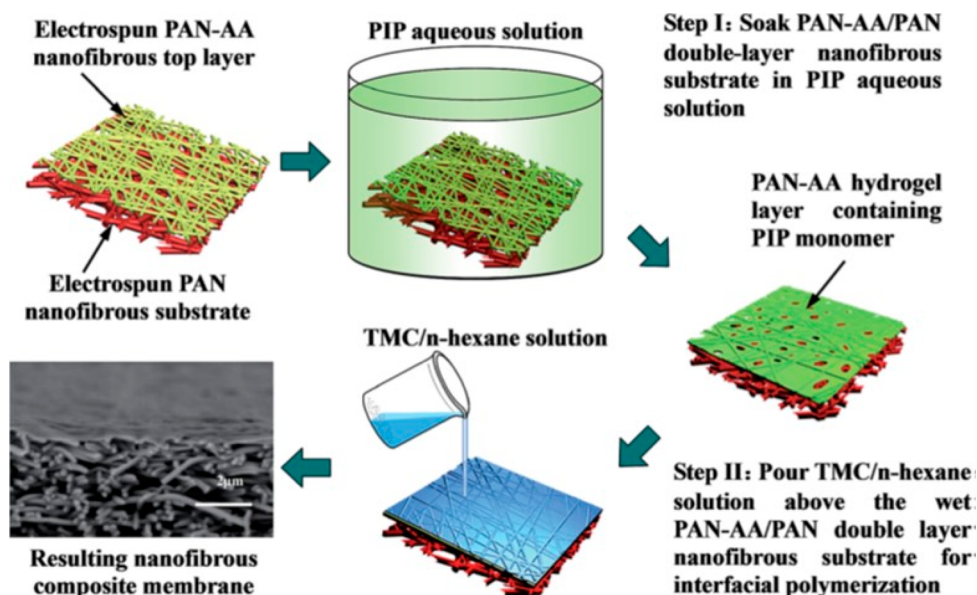
Figure 6. Schematic representation for the fabrication of the PPA−PAN−AA/PAN nanofibrous composite membrane. Reproduced from ref65 with permission from the Royal Society of Chemistry.
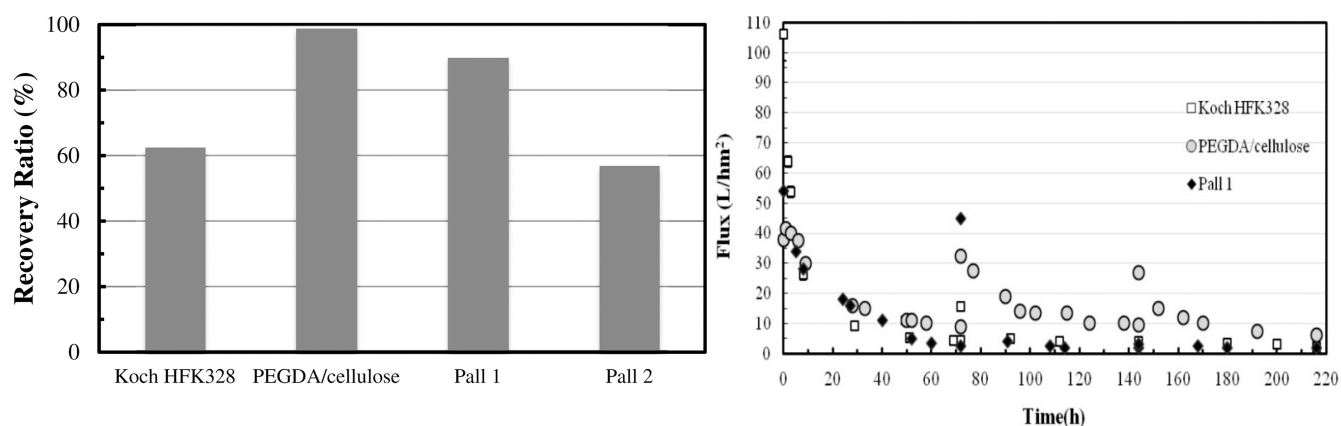
Figure 7. Flux recovery ratio and flux change for short- and long-term fouling tests, respectively. Reprinted with permission from ref 70. Copyright 2014 Elsevier.
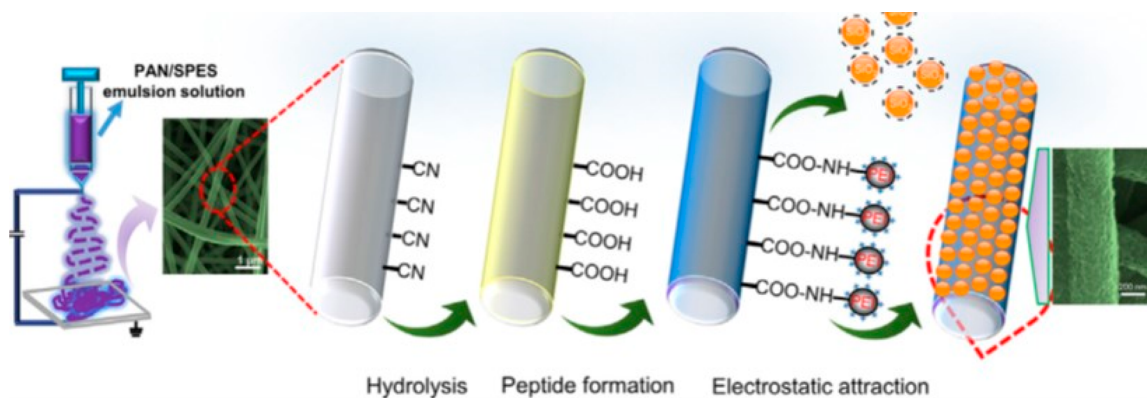
Figure 8. Schematic for the fabrication of the PAN/SPES/SiO$_2$ nanofibrous membrane. Reprinted with permission from ref 77. Copyright 2018 Elsevier.

conductivity will enhance the charge on the surface of the droplet to form a Taylor cone and in addition decrease the fiber diameter.[42] The solvent plays an important role in the formation of beadless smooth nanofibers. The two main characteristics of a

solvent are the following: (a) it should dissolve the polymer completely, and (b) the solvent should have a moderate boiling point, which gives an idea about the volatility of the solvent. Commonly volatile solvents are preferred because their fast

Table 1. Surface-Modified Electrospun Membranes with Antifouling Performances

membrane	preparation method	antifouling study	ref
poly(ethylene terephthalate) dimethyl 5-sodium sulfoisophthalate)/poly(ethylene terephthalate)	three-layered electrospinning	Estrol Turquoise Blue N-G dye rejection ≈58%	81
		FRR = 9%; Estrol Turquoise Blue N-G dye rejection = 95%	
		FRR = 15.3%	
cellulose−poly(2-methacryloyloxyethyl phosphorylcholine)/polydopamine over cellulose substrate	sequential and codeposited/ electrospinning	BSA FRR = 25%	82
		biofouling: 6.1 ± 0.5% and 6.3 ± 0.4% for *S. aureus* and *E. coli*, respectively; biofouling:1.6 ± 0.3% and 1.0 ± 0.1%) for *S. aureus* and *E. coli*, respectively	
		BSA FRR = 95%	
PVDF−(poly(hydroxyl ethyl methacrylate)/CS (chitosan)	in situ polymerization/ electrospinning/surface costing	BSA FRR = 88%	83
PVDF/nylon-6,6/chitosan	PVDF casting over electrospun nylon-6,6/chitosan nanofibrous membrane	BSA rejection: 93%	84
		RF: 0.054	
		IF: 0.071	
(poly(arylene ether nitrile)/hallosite nanotubes/graphene oxide/polydopamine)	electrospinning followed by surface coating via vacuum-assisted filtration	water/hexane emulsion rejection: >99%	85
		FRR after 10 cycles: >95%	
PEN/GO−PDA poly(arylene ether nitrile)/graphene oxide/polydopamine	electrospinning/surface coating via vacuum suction method/coating	Direct Blue 14 rejection: 92.6%	86
		FRR after 3 cycles: 95%	
graphene oxide/aminated polyacrylonitrile	electrospinning/surface dip coating	oil/water emulsion rejection: ≥98%	87
		FRR: 71.5	
calcium alginate/poly(hydroxybutyrate)/carbon nanotubes	electrospinning followed by surface coating/cross-linking	BSA rejection: >99%	88
		FRR after 3 cycles: 91%	
chitosan-coated bacterial cellulose−gcMWCNTs	electrospinning followed by surface coating	BSA rejection = 99.74%	89
		FRR after 3 cycles: 96%	
poly(acrylic acid)−poly(vinyl alcohol)/polysulfone	electrospinningof PAA/PVA over polysulfone substrate	BSA rejection = 95.5 ± 0.6%	90
		FRR: 78.3 ± 0.3%	
		biofouling: 90% reduction in *S. aureus* growth	
chitosan−glutaraldehyde−terephthaloyl chloride	electrospinning of PVDF followed by surface coating of chitosan	BSA rejection = 98.9 ± 0.2	91
		FRR: 93.04%	
poly(vinyl alcohol)/glutaraldehyde	electrospinning of PVA followed by cross-linked GA/PVA coating	soybean oil/water emulsion rejection >99.5% with a negligible decline in flux for 24 h cycle	92

evaporation rates support easy evaporation of the solvent from nanofibers, but highly volatile solvents are generally avoided because the high evaporation rate may cause drying of the jet at the needle tip.[43] Environmental aspects such as temperature and humidity also affect the characteristics of nanofibers such as morphology and diameter. Humidity controls the solidification of the charged jet, which influences the diameter of the nanofiber and also plays an important role in the creation of pores in nanofibers. This phenomenon mainly depends on the chemical nature of the polymer.[44] Temperature also affects the diameter of nanofibers. The change in temperature influences the rate of evaporation of the solvent and the viscosity of the solution, which lead to a decrease in the mean diameter of the fiber.

3. STRATEGIES FOR FOULING MITIGATION AND IMPROVEMENT OF MEMBRANE PERFORMANCE

In order to enhance the properties of electrospun membranes against fouling, it is very important to functionalize the membranes so that the fouling particulates do not adhere to the membrane surface. In this regard, several techniques are being documented in the literature and can be broadly classified into three main categories, namely, surface modification, blend modification, and composite synthesis. These processes provide specific features (such as charge) to the membrane which aid in a better antifouling nature. However, specifically when it comes to the separation of oil and its products, modifications by providing only charge will not be efficient, and properties such superhydrophilicity or superhydrophobicity are needed. Here, in either case, when the membrane possesses superhydrophilicity, it will allow only the water molecules to pass through it, whereas it will allowing only oil passage when superhydrophobic in
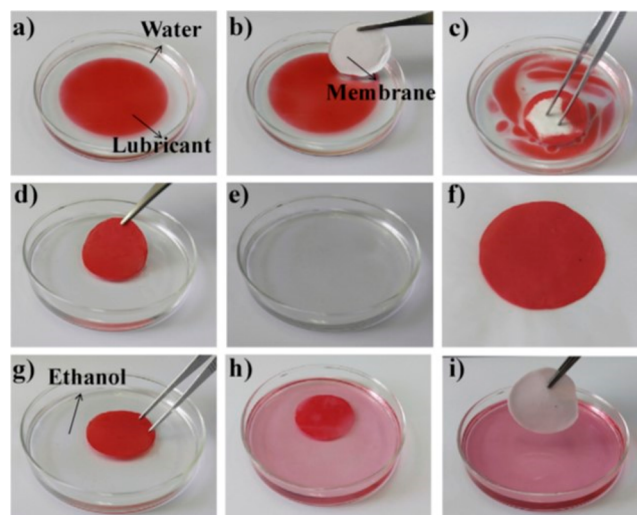
Figure 9. Separation process of lubricant (dyed with Oil Red) from the water surface and the recycling process in ethanol: (a) dyed lubricant in water; (b) immersion of the membrane in the lubricant water system; (c) lubricant allowed to decorate on the membrane; (d) lubricated membrane leaving water; (e) water after lubricating the membrane; (f) lubricated membrane; (g) immersion of the lubricated membrane in ethanol; (h) recovery of the lubricant in ethanol; (i) recovered membrane and lubricant in ethanol. Reprinted with permission from ref 105 . Copyright 2016 Elsevier.

nature. However, each method has its advantages and disadvantages. Thus, herein different approaches to fabricate improved antifouling electrospun membranes and the advancements to date are being elaborated upon.
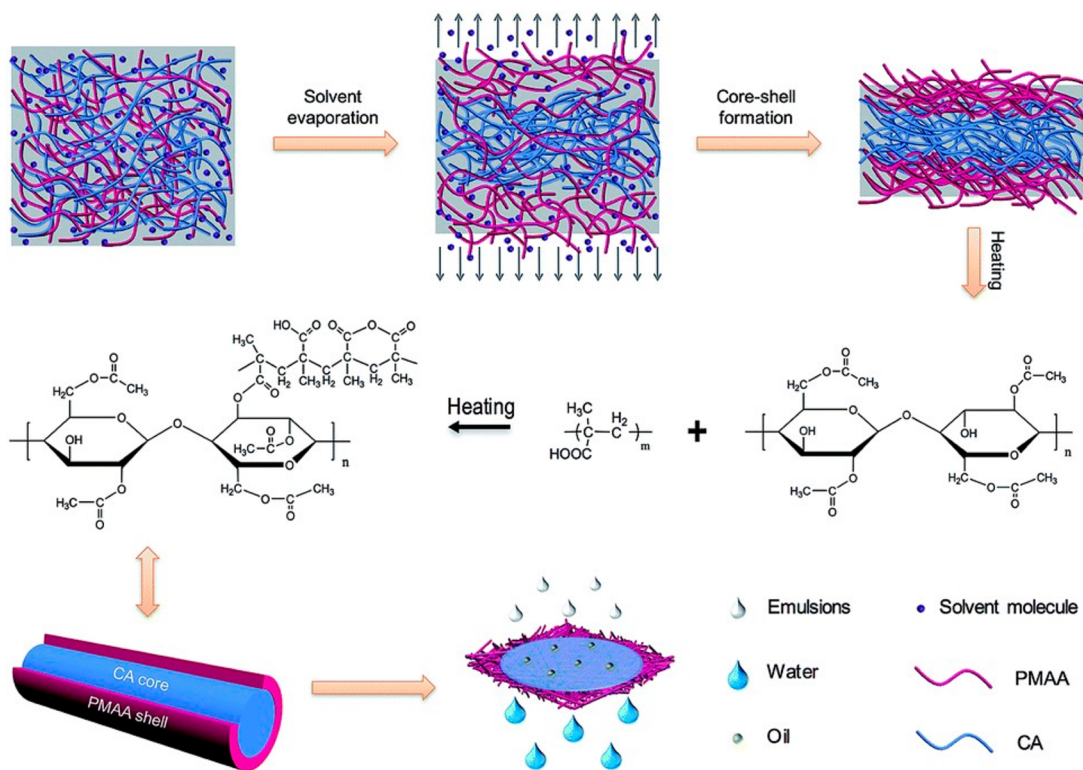
Figure 10. Schematic of the uniaxial electrospinning process. Reprinted with permission from ref 116. Copyright 2013 Royal Society of Chemistry.
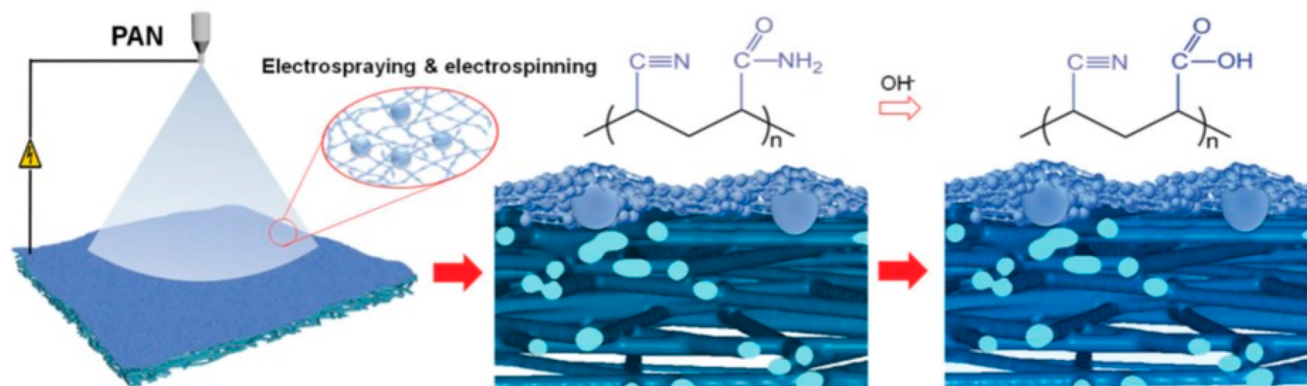
Figure 11. Schematic representation for the formation of the nanofibrous skin layers. Reprinted with permission from ref 119 . Copyright 2018 John Wiley and Sons.

Figure 12. Schematic diagram of cross-linked PLGA—SH/8cPEGa nanofibrous mesh including (a) catechol—thiol cross-linking, (b) catechol—catechol conjugation, and (c) surface-exposed PEG chains. Reprinted with permission from ref 129. Copyright 2013 Royal Society of Chemistry.
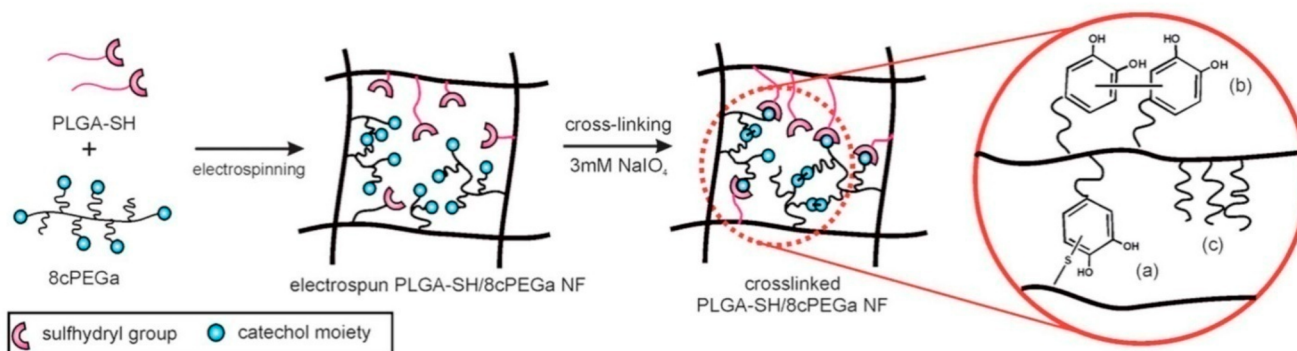
Table 2. Composite and Blend Electrospun Membranes with Antifouling Performances

membrane	antifouling study	ref
GO/PVDF	BSA FRR = 87%	130
	total fouling R_t = 20%	
	(reversible) R_r = 7%	
	(irreversible) R_r = 13%	
	biofouling:	
	Log reduction value (*E. coli*): 5.69	
	Log reduction value (*S. aureus*): 5.32–5.76	
PVDF−HFP/Cu^{2+}	FRR for polystyrene microspheres: 12.2%	131
	FRR for polystyrene microspheres: 98.10%	
S-PVDF/PVDF	oil rejection: 100%	132
	FRR for oil separation: 54%	
	R_t = 93%	
	R_r = 48%	
	R_r = 45%	
S-PVDF/PVDF/GO	oil rejection: 100%	
	FRR for oil separation: 59%	
	R_t = 92%	
	R_r = 51%	
	R_r = 41%	
PES/hydrous manganese dioxide (HMO)	% oil rejection: 94.04	133
	FRR: 71.10%	
modified PAN/polyaniline−nylon core shell	% BSA rejection: 89.11	134
	FRR: 91.85%	
chitosan/PVA over nonwoven PET	FRR for PEG: 82%	135
chitosan/PVA	FRR for Direct Red 80: 87–92%	136
chitosan/PVA/SiO$_2$	% rejection Direct Red 23:98	137
	FRR: ≈81	
montmorillonite (Mt)/chitosan/PVA	% rejection Basic Blue 41:95	138
	FRR: ≈88	
poly(phenylsulfone)/PEG	BSA FRR = 83%	139
PVDF-Ag-GO	BSA FRR = 93.8 ± 2.6%	140
	R_{ir} = 6%, R_r = 47%	
PAN−*para*-aminobenzoatealumoxane NPs	activated sludge FRR= 95.94%	141
	R_{ir} = 4.05%, R_r = 68.58%	
fumarate−alumoxane/PAN	activated sludge FRR = 96%	142
	R_{ir} = 4%, R_r = 72%	
PES/Ag NPs	activated sludge FRR = 99%	143
	R_{ir} = 1%, R_r = 4%	
polyurethane PU/PVP	olive oil rejection: 91.7%	144
	gingelly oil rejection = 99.68%	
	FRR = 84.1 ± 3%	
PAN/GO	FRR of oil/water emulsion: 50%	145
hydrolyzed-PAN/GO	FRR of oil/water emulsion: 99%	

3.1. Surface Modification.

Surface modification deals with the surface treatment of electrospun membranes and requires post-treatment techniques such as spray coating, dip coating, plasma treatment, and sputtering and also methods like thin film formation, cross-linking, etc.

3.1.1. Surface Grafting. Grafting is a technique that deals with the incorporation of functional groups onto the surface of the membrane by either a chemical reaction or plasma treatment in order to tune the hydrophilicity or hydrophobicity of the membrane surface as required. This, in turn, enhances the antifouling nature by not allowing the particles to settle on the

membrane surface. Mei et al.[45] functionalized electrospun polyacrylonitrile (PAN) membranes by simply reducing the nitrile groups to amine and further coupling it with glycerol diglycidyl ether (GDGE) and poly(ethylene glycol) diglycidyl ether (PEGDGE), which act as spacers to give flexibility. Finally, to give an antibacterial effect polyhexamethylene guanidine hydrochloride (PHGH) was functionalized onto the surface giving PAN−NH$_2$−GDGE−PHGH and PAN−NH$_2$−PEGDGE−PHGH membranes, respectively. Biofouling was carried out for *Escherichia coli* and *Staphylococcus aureus* via the culture method, and an antibacterial efficiency of 99.996 and 99.988% for *S. aureus* and 99.866 and 99.922% for *E. coli* was observed for PAN−NH$_2$−GDGE−PHGH and PAN−NH$_2$−PEGDGE−PHGH, respectively, whereas near negligible activity was observed for the control membrane. This was maintained and observed to be nearly constant after three cycles. To validate the filtration ability of the membranes, bacterial solutions were filtered through a dead-end filtration unit giving superior water flux, and the relative flux recovery was in the order of 53.7 > 37.6 < 95.0 < 99.9% for PAN, PAN−NH$_2$, PAN−NH$_2$−GDGE−PHGH, and PAN−NH$_2$−PEGDGE−PHGH membranes, respectively. Obaid et al.[46] followed a novel process to functionalize the surface of electrospun polysulfone membrane by immersion in NaOH solution at various temperatures and times, for incorporating the OH functional group. The optimum membrane showed an increase in hydrophilicity and increased underwater superoleophobicity (oil contact angle of 158° ± 8), while the pristine membrane showed an oil contact angle of 0°. This membrane also exhibited an outstanding rejection efficiency of 99.99% for the oil−water system and a water flux recovery ratio of 94 and 96.8% after the fifth filtration cycle of soybean oil and hexane−water mixtures, respectively. Xin et al.[47] followed the co-electrospinning method to fabricate zwitterionic polyurethane/poly(BMA-*co*-DMAEMA) composite mats. First, the sulfobetaine precursor of poly(BMA-*co*-DMAEMA) copolymer was used, wherein functionalization by sulfobetainization led to the increased hydrophilicity of membranes. These membranes showed an improved antifouling property against bovine serum albumin (BSA) with a 40% reduction in protein adsorption with an increasing composition of zwitterionic copolymer.

3.1.1.1. Grafting Based on Plasma Activation. Activating the membrane surface by plasma treatment is vital to keep up the overall performance of the membrane. Prefabricated electrospun polystyrene/polyacrylonitrile (PS/PAN) composite films were plasma treated in a nitrogen environment to generate free radicals and then allowed to react with acrylic acid via dip coating as depicted in Figure 3.[48] These surface-grafted membranes showed improved superhydrophilic and underwater superoleophobic surfaces with separation efficiency of >99.8% (paraffin oil) and >99.5% (kerosene) for the PAN-*g*-AA and PS/PAN-3-*g*-AA membranes, respectively. The efficiency was maintained after 5−10 cycles, showing good antifouling activity against oil.

Owing to the effective grafting efficiency, many groups have reported to have used plasma treatment for graft functionalization. Sun et al.[49] and Yalcinkaya et al.[30] fabricated PVDF−HFP and PVDF/PAN electrospun nanofibrous membranes using intermediate plasma activation followed by poly(ethylene glycol) methyl ether methacrylate (PEGMA) and TiO$_2$ grafting, respectively. Upon surface grafting, both membranes show an increase in hydrophilicity with the water contact angle (WCA) dropping to 0. The PVDF−HF/PEGMA membrane showed an

improved resistance toward BSA protein adsorption, whereas the PVDF/PAN/TiO$_2$ was able to filter oily wastewater with an antifouling property and allow it to be used for 10 cycles unlike the control PVDF/PAN membrane. It also self-cleaned itself from BSA adsorbed protein under UV light (with 95% efficiency). On the other hand, Zhao et al.[50] used electrospun polypropylene fibril (PPF) for grafting poly(acrylic acid) instead of PVDF-based membranes. The grafting of poly(acrylic acid) increases the wetting behavior with a decease in WCA from 132 to 90°, and it finally gets completely absorbed within 8 s. This led to an increased water flux and reduced BSA adsorption fouling by 67% as compared to the virgin PPF electrospun membrane.

3.1.2. Electrospraying. Electrospraying is well-known as electro-hydrodynamic atomization and is a comparatively new nanoencapsulation technique very similar to electrospinning, although in the place of nanofibers, nanoparticles are formed. Versatile factors such as surface tension, flow and viscosity of the liquid stream, applied current and voltage, and size of the capillary nozzle influence the nature and size of the particle.[51] For example, PAN substrate with a hierarchical structure consisting of PAN and SiO$_2$ NPs was fabricated using an integrated electrospinning and spraying method.[52] In this, the SiO$_2$ surface coating provides superhydrophilic and underwater superoleophobic properties. This resulted in an antifouling ability that was evaluated for an *n*-hexane-based surfactant-stabilized emulsion for a set of 10 cycles and resulted in no significant change in the separation efficiency and flux. With the same PAN base polymer, a polypropylene electrospun membrane was fabricated with electrosprayed oxidized multi-wall carbon nanotube (OMWCNT) layers.[53] The incorporation of OMWCNTs resulted in a hierarchical composite membrane on the PAN/polypropylene electrospun membrane with an electrosprayed OMWCNT layer. The optimized composite membrane provided 3.11 times higher percent rejection with a compromise of 17.30% flux over the PAN/PP control membrane. On the other hand, Zhu et al.[54] followed a sequential electrospinning and electrospraying route to fabricate asymmetrically superwettable Janus skin (F-SiO$_2$@PVDF−HFP/PS and SiO$_2$@PAN) over an electrospun LiCl/PVDF hydrophobic nanofibrous membrane (NFM). Huang et al.[55] followed a multistep technique, i.e., the adsorption of fluorinated SiNPs, fluorination by chemical vapor deposition, and SiNPs−CTS/PFO spraying, to provide an electrospun CTAB/PVDF−HFP membrane with surface hydrophilicity and omniphobic properties as presented in Figure 4. Thus, the fabricated membrane was fortified with an antiwetting omniphobic substrate and an antifouling skin layer, proving antifouling toward saline oil-in-water emulsion (35 g/L NaCl and 1000 ppm crude oil) for a membrane distillation operation of 10 h without compromise on its flux and rejection. However, the hydrophobic PVDF−HFP membrane and the omniphobic membrane were fouled and experienced a loss in flux by less than 20% of its initial flux.

3.1.3. Dip Coating. Dip coating is a known, low-cost method for the fabrication of functional layers. This is also known as slurry or vacuum slurry dip coating, which has the advantage of layer thickness adjustment but the disadvantage of slowness.[56] The phenomenon involves the force of inertia, gravitational force, viscous drag, and surface tension.[57] Ao et al.[58] demonstrated a simple dip coating of an electrospun cellulose nanofibrous (CNF) membrane in aqueous graphene oxide solution to achieve a superhydrophilic GO@CNF membrane, having a surface underwater−oil contact angle (OCA) ≈ 155° as compared to 149° for pristine CNF membrane. Also they

achieved an antifouling capacity with high separation efficiency (>99%) for the hexane/water system after 10 cycles, maintaining a flux of 0.96 m^3 h^{-1} m^{-2}. Similarly Lv[59] and his team also prepared underwater oleophobic PVDF electrospun membranes with a hydrophilic surface, by coating polyamide 6 (PA6). The PA6 solution penetrated into the PVDF nanofibers and furnished an interconnected pore network. This provided an enhanced mechanical strength (eight times), superior hydrophilicity (WCA: 132° to ≈0°), and superoleophobicity (OCA: 20.5°−148°) for pristine PVDF- and PVDF/PA6-coated membranes, respectively. The antifouling capacity also had increased, maintaining the oil/water emulsion separation efficiency >99% with a flux of above 850 LMH for three cycles.

Unlike the above studies, Liu and Yuan[60] fabricated a biodegradable superhydrophobic membrane instead of hydrophilic, consisting of an electrospun nanofibrous poly(lactic acid) substrate coated with polydopamine (PDA) and Ag NPs for oil separation. A hierarchical surface was obtained with superhydrophobicity (WCA ≈ 158.6°) and superoleophilicity (OCA = 0°). This property aided in achieving a permeation flux of 2664.3 ± 48.2 LMH and water-in-oil emulsion separation efficiency of 98.4 ± 1.0%. In addition, the separation efficiency was maintained for 20 cycles with a marginal drop in WCA, which still remained above 140°. The membrane could also show biofouling against *E. coli* and *S. aureus* with efficiency reaching 98.2 ± 0.4% and 99.0 ± 0.4%, respectively. In a similar manner, a superior hydrophobic membrane with WCA of 162° and OCA ≈ 0° and with a flux of 3106.2 ± 100 LMH was reported by Ma et al.[61] Researchers have also tried a dual-coating procedure as represented in Figure 5 and prepared a TiO$_2$/PAN−Si composite membrane.[62] The membrane was able to perform several separation cycles for petroleum ether, pump oil, and soybean oil with a rejection efficiency of 99%. Also, owing to the self-cleaning ability of the TiO$_2$ NPs, the membrane regained its initial flux upon treatment with UV light.

3.1.4. TFC Fabrication. Thin film composite (TFC) membranes are widely applied in PRO applications because of their promising permeability, excellent acceptance of a wide range of pH, and long durability.[63] Despite their intolerance toward chlorine and oxidants attacks, researchers have fabricated TFCs onto electrospun NFs to enhance the antifouling ability. The layered constructions of TFCs offer several combinations to improve the performance and durability. Wang et al.[64] fabricated a three-layered TFC membrane consisting of thin films (polyether-*b*-polyamide copolymer (Pebax)/MWNTs or PVA/MWNTs) fabricated over the electrospun PVA/GA membrane support of a nonwoven microfibrous substrate. The presence of the TFC coating in the presence of MWNTs provides higher hydrophilic properties owing to the formation of effective hydrophilic nanochannels which provide better water passage. In a similar manner, Yang et al.[65] also prepared a three layered thin-film nanofibrous composite (TFNC) membrane but on a dual-supported electrospun PAN−AA/PAN substrate as illustrated in Figure 6. The optimum (PPA0.05−PAN−AA/PA) TFNC membrane showed nearly 1.5− 2 times better rejection for different salts as compared to the control membrane, and the rejection order decreased in the order Na$_2$SO$_4$ (>98.5%) ≈ MgSO$_4$ (>98.5%) > MgCl$_2$ (88.8%) > CaCl$_2$ (82.6%) > NaCl (25.3) with a flux rate of 64.4 LMH. The membrane also withstood a long-time antifouling study (60 h) against MgSO$_4$ solution, maintaining the flux rate, with a marginal decline (≈ 9%) and rejection of over 98%. This performance and stability

were attributed to the enhanced hydrophilicity and interconnection between the PPA barrier layer and the nanofibrous PAN layer. Besides, the PAN−AA sublayer was able to gather more aqueous PIP monomers beneficial for the PPA barrier layer. Yang et al. proved that the surface charge on the TFC influences the antifouling property of the CA NF.[66] An oxidized CA TFC NF membrane with negatively charged materials coating on it offered a strong electrostatic repulsion with the foulants and thereby increased the self-cleaning ability.

3.1.5. Sputtering and Cross-linking. As per the literature survey, for sputtering there was only one study done by Kassa et al.[67] to prepare a surface-modified electrospun PAN membrane by coating it with thin film metallic glass (TFMG) ($Zr_{53}Cu_{26}Al_{16}Ni_5$) using magnetron sputtering. This coating not only provided an increase in hydrophobicity (WCA from 24° to 136°) but also gave enhanced chemical and thermal stability. The superior hydrophobicity paved the way for oil/water emulsion separation achieving a maximum separation efficiency of 100% and with reusability confirmed with an antifouling study, where the adsorbed oil (WCA 145°) was easily removed with acetone wash and the WCA recurred (136°).

Thus, from the various methods described above, it can be observed that the basic idea is to increase or decrease the hydrophilicity of the membrane surface particularly in the case of the oil separation experiments. However, for protein or bacterial studies, charge and antibacterial components take the primary role. Several other articles describing the role of different modifying agents are also deliberated upon. Conventional PVDF NF membranes were blended with poly(methyl methacrylate) block and poly [*N,N*-2-(dimethylamino)ethyl methacrylate) block (PMMA-*b*-PDMAEMA) to give antibacterial and hydrophilic characteristics.[68] Further, PVA coating and cross-linking in methanol made the membrane superhydrophilic, and based on the alkyl chain length, the water flux and WCA (0−20°) varied, giving maximum flux with the longest alkyl chain. The total fouling against BSA protein was the lowest at 49.99 of which 45.5% was reversible as compared to the PVDF/GO@PVA membrane. Besides, it showed biofouling against *E. coli* (4.2×10^5 CFU/mg) and *S. aureus* (6.1×10^5 CFU/mg) by damaging the cytoplasmic membrane and inhibiting cell growth.

Cellulose acetate (CA) NF membranes were fabricated over aluminum plates and impregnated with chitin nanocrystals via a Buchner funnel filtration setup to give it a superhydrophilic nature.[69] This led to the H-bonding between the chitin and cellulose molecules forming a weblike network at the junction of cellulose acetate fibers and gave the electrospun membrane a thin coating. This coating not only increased the hydrophilic nature with a contact angle of 0° but also rendered superior biofouling toward *E. coli* (CECT 516) cells.

The UV curing method was applied to cross-link cellulose nanofibers (CN) and PEG over the electrospun PAN/PET NF membrane to fabricate a hydrophilic membrane.[70] Different ethylene glycol monomers, namely di(ethylene glycol) 15 diacrylate (DEGDA), tetra(ethylene glycol) diacrylate (TEGDA) 16, and poly(ethylene glycol) diacrylate (PEGDA), were used for cross-linking with CN. This coating led to improved hydrophilicity with WCA of ≈14 for all the prepared membranes, but PEDDA/PAN/PET outperformed all the membranes in terms of flux and rejection (PEG and BSA). This is because of the longer monomer chain, which forms intermolecular cross-linking (rather than intramolecular cross-

linking), leading to a homogeneous network by the conversion of double bonds unlike intramolecular cross-linking, which promotes cyclization. PEDDA/PAN/PET also outperformed some of the commercial membranes such as Pall Life Sciences omega membranes and Koch HFK 328 14 membranes by showing a flux recovery ratio of almost 100% and 75% after a short- and long-term BSA antifouling study, respectively (depicted in Figure 7).

An interesting antifouling study was carried out by measuring the underwater dynamic oil−membrane interaction force on the membrane surface giving its nature toward oil droplets, where the negative force denotes hydrophilicity and positive denotes hydrophobicity.[71] Direct cross-linking of electrospun poly(ether imide) (PEI) nanofibers with ethanediamine (EDA) over a commercially available hydrophobic PVDF membrane was carried out to increase its hydrophilicity (WCA of $38.5 \pm 1.6°$) and underwater oleophobicity (OCA of $145.3 \pm 0.8°$) compared to the PVDF membrane [WCA ($127.2 \pm 0.5°$) and underwater OCA ($40.5 \pm 0.7°$)]. The PVDF/PEI−EDA membrane exhibited strong resistance to crude oil showing negative force. This effect was applied in the oil/saline membrane distillation process where the flux recovery ratio for the PVDF membrane was 15% whereas the PVDF/PEI−EDA membrane had a near 100% flux recovery ratio.

A combination of embedded silanized silica nanoparticles (f-SiO_2 NPs) within PVDF nanofibers with silver nanoparticles and carboxylated multiwalled carbon nanotubes (AgNPs/f-MWCNTs) coating was successfully fabricated via diffusion-induced phase separation for biofouling activity as well as biocidal properties.[14] Likewise, Yuan et al.[72] coated a PVDF substrate with PVDF/graphene solution to get a dual-layer composite membrane, and it showcased robust properties with a flux of $150 \text{ kg/m}^{-2} \text{ h}^{-1}$ and an oil purity of 99.90% after 10 cycles of separation. Another combination with silica-based nanofiber (SNF) membranes (PAN coated) having SiO_2 NPs[73] and $NiFe_2O_4$ NPs[74] gave an excellent antifouling property with their honeycomb-like NF layer (HCNFM).[75] These coatings provided superhydrophilic properties and also showed antifouling behavior against oil/water emulsions for 10 separation cycles. PAN NFs and PVA NFs are interesting ones offering superhydrophilic surface coatings with zero water contact angle.[76] A blend of PAN and sulfonated poly(ether sulfone) NF membranes followed by amination of PAN and subsequent binding of negatively charged SiO_2 NPs, as depicted in Figure 8, provided a lotus-leaf structure having superhydrophilicity and underwater superoleophobicity with WCA of 0° and OCA of 161°, respectively.[77] Guo et al.[78] and Kang[79] et al. performed oil/water separation fouling experiments with poly(hydroxy butyrate)−calcium alginate/carboxyl multiwalled carbon nanotubes (PHB−CaAlg/CMWCNT) and *β*-cyclodextrins−polydopaminepoly(L-lactic acid) (*β*-CD−PDA@PLA) electrospun composite membranes, respectively. The flux for the oil emulsion and Brilliant blue solution was 84.01% and 89.73%, respectively, with respect to pure water flux for the PHB−CaAlg/CMWCNT membrane, displaying an excellent antifouling property. On the other hand, the *β*-CD−PDA@PLA membrane was utilized for separation of oil emulsions and MB with a separation efficiency of 99.5% and dye adsorbability of over 95%. The membrane was able to perform for 30 continuous cycles to decontaminate the toluene emulsion and methylene blue wastewater. Likewise, other different surface-modified electrospun NF membranes are illustrated in Table 1 with their antifouling performances for different feed solutions.

In advanced membrane research, apart from chemical, physical, and biological control, surface grafting is employed to increase the superhydrophobic nature of the membrane surface which acts as a self-cleaning material and hence avoids the biofouling. The other method of biofouling mitigation via surface modification is the grafting of antibacterial agents and minimizing the bacterial adhesion. Shi et al. designed a high-strength and antibiofouling poly(amidoxime) HA-PAO nanofiber membrane via supramolecular cross-linking.[80] The membranes contained the antibacterial ions immobilized on the framework via cross-links, leading to a larger amount of adsorption ligands on the surface.

3.2. Blend and Composite Nanofiber Membranes.

Composite membranes are another class of membranes, in which instead of special modifications done over the surface of electrospun membranes, fillers such as nanomaterials, clay, etc. are premixed with the polymer solution before electrospinning. These fillers that possess charge give a specific charge throughout the membrane and also a synergetic effect arising from properties of both the polymers and filler materials. This charge aids in antifouling behavior by not letting the particulate matter settle on the membrane surface. However, NPs tend to agglomerate in high compositions.[93] Blend membranes are a mixture of two different polymers or polymer/fillers that are miscible with each other, providing a homogeneous matrix. Thus, it gives a uniform charge without the worry of agglomeration.[94] This gaze allowed researchers to utilize different nanomaterials in the fabrication of composite NFs; especially, TiO_2 NPs are being extensively studied owing to their nontoxicity, stability, economic feasibility, and self-cleaning property.[95] Daels et al.[96] and Karimi et al.[97] synthesized PA-6 (polycaprolactam)/TiO_2 and PVA/TiO_2 membranes, respectively. The charge and hydrophilicity provided by Ti offered biofouling capabilities to the PA-6/TiO_2 membrane with 100% degradation of *S. aureus* bacteria under UV radiation. PVA/TiO_2 membranes were able to reject the oil/water emulsion with a minimum flux decline ratio (FDR) of 32.8 as compared to the neat PVA membrane with a FDR of 81.34. Similarly, Zhang et al.[98] used TiO_2 NPs in the fabrication of PVP pine-branch-like TiO_2 NFs, Bode-Aluko et al.[99] fabricated photocatalytically active PAN/TiO_2 NFs, and Wang et al.[100] fabricated TiO_2/PVDF NFs with a beads-on-string structure. Membranes showed antifouling and self-cleaning performance resulting from the photocatalytic property of TiO_2 along with interchangeable superhydrophilicity/superhydrophobicity.

Three-dimensional woven filters with Ag NPs were synthesized wherein weft yarns were wrapped by PAN electrospun nanofibers.[101] The filtration experiments were carried out for activated sludge for an operation time of six days, cleaned, and used again for nearly seven cycles. Incorporation of Ag NPs gives it the required hydrophilicity for high flux, antimicrobial resistance, and durability. Correspondingly, Pant et al.[102] fabricated polyurethane-based nanofibers, incorporated with silver-doped fly ash. This furnished the membrane surface with a spider-web-like network giving it more surface area and charge. Thus, this provided superior MB adsorption, arsenic removal (~75%), and biofouling against *E. coli* as compared to pristine PU membrane. Independently, Xavier et al.[103] manufactured polystyrene/poly(vinyl(methyl ether))/Ag NPs electrospun membrane and studied its biofouling activity against *E. coli* bacteria via direct contact method. Incorporation of Ag NPS led to increased hydrophobicity. However, the antibacterial activity of PS fibers containing Ag increased owing to the microgrooved structure and easy release of Ag ions.

On the other hand, Hammami et al.[104] demonstrated the use of organosilica nanoparticles as a filler material, and Liu et al.[105] experimented with ZnO NPs in the PVDF substrate for oil/water separation. The incorporation of NPs resulted in the superhydrophobic surface being able to shift from being hydrophobic to superhydrophobic or from superoleophilic to superamphiphobic by the simple addition of ammonia or 1*H*,1*H*,2*H*,2*H*-perfluorodecyltriethoxysilane. This gave the membrane superior antifouling and oil separation properties (Figure 9), and the membrane was able to be reused for 20 cycles without losing any efficiency.

Different filler materials are being explored on different polymers to find the best suited for wastewater treatment. PVDF has been explored to quite an extent especially in oil/water separations owing to its superior hydrophobic, stability, and chemical resistance characteristics.[106] Either the simple PVDF form is being utilized, or its modified forms are being used.[107] Blending of PVDF with PMMA gives it a hydrophilic touch, achieving a WCA of <10°. The membrane showed remarkable antifouling tendency against secondary effluent where the filtration is carried for 16 days without a cleaning operation. A removal efficiency of 100% for suspended solids and 48% of chemical oxygen demand were achieved. Notably, Chen et al.[108] fabricated poly(*p*-phenyleneterephthalamide) (f-PPTA)/PVDF electrospun NF membrane with superhydrophilic properties owing to the multiarms of f-PPTA which act as water channels in the nanofibrous membrane. With an advancement in the integration of hydrophilicity and charge (either positive or negative) by the incorporation of fillers; zwitterionic membranes have gained importance in giving better target contaminant removal efficiency.[109] Vasantha et al.[110] utilized polysulfobetaine and polysulfabetaines (PSBs) to create an antibacterial NF membrane with zwitterionic and hydrophilic character for antibacterial adhesion. Ozcan et al.[111] synthesized super-hydrophobic poly(trifluoroethyl methacrylate-random-sulfobetaine methacrylate) copolymer to fabricate a self-cleaning PTFEMA-r-SBMA electrospun membrane that shows a decrease in 80% BSA fouling as compared to the pristine PVDF membrane.

Amphiphilic NFs were fabricated using ter polymer, polystyrene-*b*-poly(ethylene-r-butylene)-*b*-polyisoprene (KB), and poly(lactic acid) (PLA), which led to an increase in hydrophilicity of the NFs thus decreasing the WCA.[112] An assembled electrospun nanofibrous cellulose and polysulfone on a poly(ethersulfone) ultrafiltration substrate in a layered fashion gave excellent antifouling ability.[113] Interestingly, Jalvo et al.[114] fabricated core−shell nanofibers with poly(lactic acid) at the core surrounded by PAN/cellulose nanocrystal (CNC) or PAN/chitin nanocrystal (ChNC) shell following a coaxial electrospinning technique. PAN/CNC and PAN/ChNC membranes exhibited enhanced hydrophilicity and super-hydrophilicty, respectively, with ChNC loading instigating a 240% increase in flux rate. The PAN/CNC membranes were negatively charged whereas the PAN/ChNC membranes showed neutral charge or slightly positively charge, which behaved better against the *E. coli* bacterial adhesion. This is attributed to the enhanced positive charge and superior hydrophilicity achieved working together against the negatively charged *E. coli* bacteria. On the basis of the size exclusion principle, both the membranes succeeded in giving considerable retention of *E. coli* and *Aspergillus niger* spores.

Similarly, Kao et al.[115] also used a coaxial technique to prepare core/shell polyacrylonitrile (PAN)—polybenzoxazine (PBA) fibrous membrane, with PBA forming the outer shell. The membranes showed superhydrophobicity with WCA > 150° and BSA adsorption of just 10.63 mg g^{-1} as compared to 36.38 mg g^{-1} for PAN. However, it was noticed that the hydrophobicity was not the only criterion for the antifouling behavior, but the surface free energy of the membranes also played a crucial role. It was observed that the PAN/BA membrane showed more BSA adsorption even after inheriting high WCA. This can be explained as follows. Core—shell formation in PAN/PBA membranes due to curing at high temperature provides a lower surface free energy and smoother surface, unlike PAN/BA membranes which possessed high surface energy and roughness without curing. CA/methylacrylic acid (MAA) core—shell nanofibrous hydrogel membrane (NHM) fabricated using an uniaxial electrospinning process (depicted in Figure 10)[116] was pH-responsive, superhydrophilic, and underwater in character. These membranes demonstrated high separation efficiency (99%) under gravity for oil/water emulsions at all pH ranges with an antifouling ability for n-hexane/water emulsion for 10 consecutive cycles without any loss in flux or rejection. A chitosan/nylon-6 composite solution was coated over a glass fiber filter giving better antimicrobial activity.[117] PAN was made composite with SiO$_2$, PET, and PVDF to fabricate fluffy NFs with larger pore size and spacing between the adjacent nanofibers aided by SiO$_2$.[118] The hydrophobic PVDF NF layer constructed at the side close to the breathing part of the membrane demonstrated good stability and permeability due to which the segregation of particulate matter was avoided.

Ding and co-workers have reported some distinguished work on the separation of oil/water emulsions. They were successful in fabricating a lotus-leaf-like hierarchical structured membrane with a porous skin layer via elaborate tuning of the transient state of the electrospraying and electrospinning and hydrolysis of PAN (Figure 11).[119] The membrane surface showed exceptional separation capabilities for different oils with negligible oil fouling (for hexane/water emulsion) and recovery by simple water washing. In a subsequent work, PAN was blended with PEG to form the nanofibrous substrate over which poly(ethylene glycol) diacrylate nanofibers were cross-linked.[120] The cross-linking provided the membrane with superhydrophilcity, oleophobicity, and a high flux rate with an ability to separate oil/water mixture over a long period cycle, and the membrane was reused for 10 consecutive cycles without any decay in efficiency. In the next approach, they modified silica nanofibrous membrane by coating Al$_2$O$_3$ NPs via cross-linking with 3-(3-(trifluoromethyl)-phenyl)-2H-benzoxazine-6-carbaldehyde (BAF-CHO).[121] Continuing the work, fibrous isotropically bonded elastic reconstructed (FIBER) aerogels were fabricated with 3D superelasticity and superhydrophobicity with varying SiO$_2$ composition, having high antifouling performance for the petroleum ether/water emulsion system for 11 separation cycles.[122]

Instead of making thin film nanocomposites, Jang et al. fabricated PAN nanofiber membranes with GO and silver NPs, which gave a better flux than TFN membranes.[123] In the synthesis procedure in which Ag$^+$ reduced to Ag NPs, GO is reduced to rGO simultaneously. This hydrophobic membrane exhibited contact inhibition mechanisms for amplified sterilization for E. coli and S. aureus. rGO-g-poly(amidoxime)/Ag NPs/PAN nanofiber membranes also exhibited the same kind of interactions and even better dye removal efficiency.[124] The surface plasmon resonance effect and bactericidal effect of Ag$^+$ ions increased the light adsorption and hence the electron transfer from Ag to the rGO-g-poly(amidoxime) conduction band, thereby generating hydroxyl and superoxide radicals. Researchers have proved that the biofouling of the membranes can be reduced by the carbon or GO or quantum dot-based membranes as they exhibit oxidative stress on the metabolic activities of the bacteria and destruct their cell wall. Along with this, the inclusion of NPs induces hydrophilicity to the membranes and provides surface charge, improved water permeability, and biofilm antiadhesion.[125]

CA nanofibrous coating on PTFE substrate gave the required hydrophilicity to the hydrophobic substrate layer, and upon the addition of Si NPs, the authors were able to study oil fouling up to 30 h. An interaction between hydroxyl groups of CA and Si NPs with water molecules provides a hydration layer that prevents oil from adhering to the membrane surface.[126] N-Halamine and poly(vinyl alcohol-co-ethylene) (PVA-co-PE) NFs, with the property of chlorination, were able to kill bacteria up to 99.99% with a direct contact.[127] Benzyl triethylammonium chloride (BTEAC)-functionalized poly(vinyl alcohol) (PVA) NFs coated over polycarbonate were studied against Klebsiella pneumonia.[128] Another group, Kim et al.,[129] cross-linked catechol-conjugated 8-arm PEG (8cPEGa) and thiolated PLGA (PLGA—SH) (depicted in Figure 12) to form the blend NF membranes. Cross-linking of 8cPEGa improved the hydrophilicity of the membranes, giving it antibacterial and antifouling properties. Overall, it could be concluded that the addition of NPs and antifouling agents to NFs efficiently delayed the fouling process. The role of zwitterions and amphiphilic functional groups appears enormous in the current scenario.

4. FUTURE DIRECTION AND CONCLUSIONS

Antifouling and biofouling are multifaceted complications disturbing the extensive range of ongoing separations in industries. Most of the previous records on antifouling membranes have concentrated on performance flux, mechanism of fouling and biofouling, and conventional fouling mitigation utilizing antifouling agents and fabricating zwitterionic membranes. However, unveiling the addition of targeted antifouling and biofouling agents, new surface coats, and multifunctional agents in blending and composites making is still in demand. Traditional methods of biofouling mitigation are introducing the biocides to the membranes, restricting the use of biodegradable dissolved organic carbon, distracting the quorum sensing of the bacteria, and using electrical fields to control biofouling. However, advanced research involves the surface modification of membranes. The modifications pertain to the achievement of superhydrophobic membranes which would act as self-cleaning ones and also to the introduction of antimicrobial functional groups on the membranes so that microbial adhesion is avoided.

Nanofibers which undergo electrospinning provide a compatible coating with an essential antifouling property. The engineering of nanofibers with an appropriate incorporation of antifouling agents is an added advantage. The Review has given an overview of plentiful membranes which could offer good antifouling and biofouling properties. Although reduced antifouling and biofouling are observed in nanofiber membranes, more insight into the mechanism of antifouling properties in the nanofiber membrane matrix, modifications to the optimization of the process, investigations on mechanical properties, and improvement of membrane performance is still

needed. A study on the effect of fibrous nature on the fouling mitigation of NF membranes is still needed. The surface topography of nanofibers delays the fouling process by controlling the local interactions with foulants. Such nanofiber mats are opening up new opportunities in the fields of medical, environmental, and several other applications.

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Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

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