Surface Modifications for Antifouling Membranes

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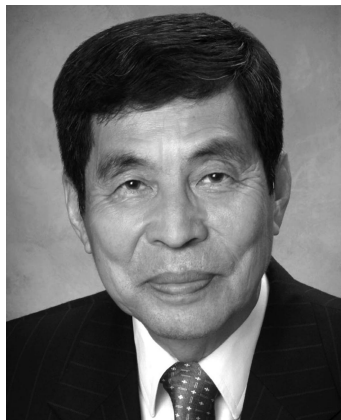
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1. Introduction

Fouling is the deposition of retained particles, colloids, macromolecules, salts, etc., at the membrane surface or inside the pore at the pore wall. Fouling reduces the membrane flux either temporarily or permanently. While the initial flux can be restored by washing the membrane or by applying back-pressures to the temporarily fouled membrane, it cannot be restored when the membrane becomes permanently fouled. The main focus of this Review is on the permanent flux decline. The fouling is caused by the interaction between the membrane surface and the foulants, which include inorganic, organic, and biological substances in many different forms. The foulants not only physically interact with the membrane surface but also chemically degrade the membrane material. For example, colloidal particles, such as natural organic matter (NOM), are considered as the main reason for membrane fouling, which could be controlled by the permeation hindrance and electric double layer repulsion. The formation of biofilms with extra-cellular polymeric substances (EPSs) and microbial cells matrix is the example of biofouling.[1] Biofilms are developed by the microbial cell adhesion and subsequent colonization on the membrane surfaces through EPS, which may account for 50−90% of total organic carbon. The biofouling could be minimized by periodical washing with chemicals such as sodium hypochlorite solution, but it will result in the simultaneous degradation of the membrane material's lifetime. It is a severe problem for membranes used in pressure-driven processes such as reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF) and also for other membrane processes, seriously hampering the applications of membrane processes. Hence, membrane fouling as well as its reduction has been a subject of many academic studies and industrial research and development efforts since the early 1960s when

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industrial membrane separation processes emerged. Selection of an appropriate membrane, pretreatment of the process fluid, adjustment of operating design, and conditions are all known to control fouling to some extent. On the other hand, development of absolutely nonfouling membranes seems extremely difficult, if not totally impossible. This Review surveys the latest efforts in which the reduction of irreversible fouling is attempted by the modification of the membrane surface. The separation process by membrane is essentially a surface phenomenon. More specifically, the skin layer or top surface layer plays the vital role. Therefore, it is a natural consequence to modify membrane surface for reducing the fouling.

It is generally accepted that an increase in hydrophilicity offers better fouling resistance because protein and many other foulants are hydrophobic in nature. Most nanofiltration membranes are electrically charged, which significantly reduces the scale-formation. During the past decade, the emergence of atomic force microscopy (AFM) enabled us to study the effect of the surface roughness in nanoscale on the membrane fouling. It is believed that the membrane fouling with particulate substance is enhanced by an increase in the surface roughness. It is shown in this Review that all of the above concepts, except for the membrane surface charge, are based on correlation of data, which are, at best, valid within a limited range of surface property parameters.

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Figure 1. Correlation between contact angle and relative flux. Reprinted with permission from ref 2. Copyright 1997 Elsevier.

Takeshi Matsuura received his B.Sc. (1961) and M.Sc. (1963) from the Department of Applied Chemistry at the Faculty of Engineering, University of Tokyo. He received his Doktor-Ingenieur at the Institute of Chemical Technology of the Technical University of Berlin (1965). He worked at the National Research Council of Canada from 1969 until he joined the University of Ottawa in 1992 as a professor and the chair holder of the British Gas/NSERC Industrial Research Chair. He served as a professor of the Department of Chemical Engineering (currently Chemical and Biological Engineering) and the director of the Industrial Membrane Research Institute (IMRI) until he retired in 2002. He was appointed to professor emeritus in 2003. He served also at the National University of Singapore (2003), University Technology Malaysia, Skudai, Malaysia (2007, 2009), at Myongji University, Yongjin, Korea (2008), as a visiting professor. He received the Research Award of International Desalination and Environmental Association in 1983. He is a fellow of the Chemical Institute of Canada and a member of the North American Membrane Society. He has published over 300 papers in refereed journals, authored and coauthored 4 books, and edited 6 books. A symposium of membrane gas separation was held at the Eighth Annual Meeting of the North American Membrane Society, May 18−22, 1996, Ottawa, to honor Dr. Matsuura together with Dr. S. Sourirajan. He received the George S. Glinski Award for Excellence in Research from the Faculty of Engineering of the University of Ottawa in 1998.

It is our intention to point out that, due to limited and delayed availability of research and development results from different sources, conclusions of the scientific community working in this area are not always firmly entrenched by experimental data obtained systematically.

It should be noted that many surface-sensitive techniques such as AFM, scanning tunneling microscopy (STM), secondary ion mass spectrometry (SIMS), X-ray photoelectron spectroscopy (XPS) (also known as electron spectroscopy for chemical analysis, ESCA), laser scanning confocal microscope (LSCS), electron spin resonance (ESR), neutron reflectivity (NR), and scanning electron microscope (SEM) have contributed to the study of membrane surface modification during the past decade or two. Each technique has its own merit and limitation. Fourier transform infrared (FTIR) with attenuated total reflection (FTIR-ATR), XPS with energy disperse X-ray (XPS-EDX), and SIMS could provide chemical and structural information of the membrane surface. XPS gives quantitative information of polymer, whereas SIMS provides qualitative information, although it can differentiate among polymers that have the same XPS data. AFM can measure porosity, pore size, and its distribution, nodule size, and aggregate size at the membrane surface. FTIR-ATR is not very surface sensitive due to a large penetration depth, although it is a common technique to analyze quickly the membrane surface.

Contact angle depends upon surface hydrophilicity (or hydrophobicity), roughness, porosity, pore size, and its distribution. If a membrane is highly porous, then the contact
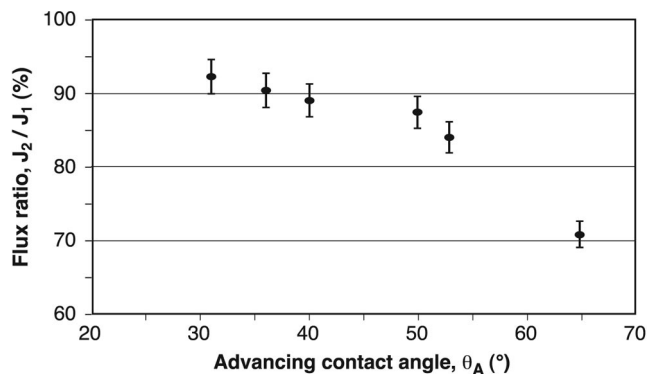
angle value may become very low, although the membrane is not necessarily hydrophilic. Similarly, the contact angle value of a membrane of higher surface roughness is higher as compared to the other membrane of lower surface roughness, although both membranes are of similar hydrophilic nature. The sizes of the pores obtained from the image analysis of AFM are often greater than those obtained from other methods, because it is believed that AFM measures the pore size size at the mouth of the funnel-shaped pores. Recently, AFM was used to investigate adhesion properties of membranes by force measurement. Using the colloidal probe technique, that is, by attaching micrometer-sized spheres to the cantilever, one can quantify the interaction force working between surface and probe. A membrane with a hydrophilic surface interacts strongly with a hydrophilic probe as indicated by a large phase shift, whereas the hydrophobic surface gives only a small phase shift.

1.1. Surface Hydrophilicity

It is usually assumed that fouling decreases with an increase in hydrophilicity of the polymeric material. The above assumption seems reasonable as with an increase in membrane surface hydrophobicity, hydrophobic organic molecules are driven more toward the surface, resulting in the enhancement of surface contamination. Nevertheless, there are only few works in which membrane fouling is directly correlated to the hydrophilicity/-phobicity of the membrane surface. One such correlation is given in Figure 1. The figure clearly shows that the flux ratio J/J_0 decreases, which means an increase in fouling, with an increase in contact angle (i.e., an increase in hydrophobicity). The correlation was, however, shown only in a limited range of contact angle, that is, from 30° to 65°. Given the fact that other membrane parameters also change together with the change in contact angle, the above correlation is of limited value. It would be dangerous to extrapolate the correlation in Figure 1 from extremely low (very hydrophilic) to extremely high (very hydrophobic) contact angle values, which is often exercised in the industry particularly when ultralow contact angle membranes are commercialized.

On the basis of the above assumption, many attempts have been made to increase the membrane surface hydrophilicity by surface modification, as summarized in Table 1. They are classified into several categories, and for each category there are advantages and disadvantages. For example, adsorption and coating could apply successfully, but the surface layer will also be easily removed after long usage of membranes. Surface chemical reactions are carried out often under strongly hazardous conditions. Although the

Table 1. Reduction of Fouling by Increasing the Surface Hydrophilicity

base material	treatment	function of the membrane
Adsorption		
PS	nonyl phenol polyethoxylates surfactants	UF, treatment of BSA solution; treated membranes were more uniform at the surface and more pronounced for the longer operating time period[11]
PS	MC, PVA, and PVP polymers	UF, treatment of BSA solution; the fouling was noticed due to the deposition of protein[12]
PS	various polymers and surfactants	UF, β-lactoglobulin solution; fouling due to protein adsorption on the surface was reduced[13]
polycarbonate and PS	poly(vinyl methyl ether) and MC	UF and MF, protein adsorption also occurred at the pore walls of the membranes; modified membrane showed a reduced pore size[14]
polystyrene divinyl benzene	high molar mass surfactants with sulfonate end groups	ED, flux reduction in humic acid, and DBS solutions; modification resulted in no fouling when DBS was used[15]
CA	poly(vinyl acetate-*co*-AA)	humic acid filtration; the most effective treatment was with an anionic polymer, which reduced adsorption of humic acid[16]
PS	Triton X-100 and pluronic F108 surfactants	NOM filtration; pluronic F108 surfactant reduced foulant adsorption significantly[17]
sulfonated PES	anionic SDS and cationic TTAB surfactants	NOM filtration; this enhanced NOM rejection but was accompanied by significant flux decline[18]
PES	PSSS surfactant	UF, flux reduction in PEGs and dextrans solutions; surface-modified membranes show better antifouling properties as compared to unmodified membranes[19]
PES	PVA and borax solution	UF, flux reduction in BSA solution; membrane surface modification by an adsorption-cross-linking process was dependent upon the adsorption-cross-linking cycles[20]
PP	Tween 20 surfactant	MF, treatment of synthetic wastewater in MBR; the monolayer state of surfactant showed higher remaining flux and stronger antifouling ability[21]
Coating		
cross-linked polystyrene embedded into PE	fluorinated long-chain pyridinium bromide	sodium humate filtration; the modified membranes were prepared by deposition of fluorinated amphiphilic compound in an oriented layer of the Langmuir–Blodgett type[22]
PS	PEI	UF, flux reduction in ovalbumin solution; hydrophilicity of the membrane appeared to be a more important factor in flux reduction than the charge[23]
PES	polyurea/PU	UF, flux reduction in BSA, PEG, dextran, and surfactant solutions; flux improvements are possible with the modifications[24]
reinforced PVC	polypyrrole	ED, flux reduction in organic foulants; modified membranes showed excellent antiorganic fouling properties in electrodialysis[25]
sulfonated PS and PES	quaternized poly(vinyl imidazole)	UF, flux reduction in BSA, and lysozyme solutions; significant improvement in protein adsorption was observed for modified membranes at low ionic strength[26]
zirconium oxide inorganic membrane	quaternized poly(vinyl imidazole)	UF, flux reduction in BSA, and lysozyme solutions; modified membranes are presumed to be negatively charged[27]
CTA, PES, and PVDF	phospholipid (MPC)	MF, flux reduction in BSA, yeast fermentation broth, beer, and orange juice; the coating increased the initial flux and decreased the rate of fouling;[28] the fouling was caused by mostly polysaccharide rather than protein
CA and PVDF	phospholipid	MF, flux reduction in BSA solution; phospholipid coating improved flux more in the PVDF membrane than in CA membrane[29]
regenerated cellulose	PVA layer spin-coated	BSA filtration; the thicker hydrogel coatings proved effective in preventing irreversible protein fouling[30]
PVDF	polyether-*b*-PA block copolymer	UF, oil–water emulsion from metal industry; composite membrane was found to perform similar to Amicom YM30 cellulose membrane, but with lower susceptibility to fouling in the UF of oil–water waste[31]
PE and PP	azlactone and PVA	MF, treatment of BSA solution; azlactone-modified membranes showed very low long-term fluxes and large decreases in permeate protein concentration[32]
PP	polysulfonamide	MF, polystyrene, and carboxylate-modified polystyrene latex spheres; coating significantly increases hydrophilicity and flux and reduces the fouling of the latex spheres[33]
aromatic PA	PVA	RO, treatment of water with a surfactant or a transition metal; the modified RO membranes exhibited no fouling and also high separation properties[34]
PES	bentonite, diatomite, iron oxide, kaolinite, titanium dioxide, zeolite, etc.	UF, treatment of surface water from Twente canal, lake, and reservoir (Delft, Netherlands); precoating results initially in higher fouling rate, which stabilizes after several filtration cycles[35]
PES	hydrophilic triblock copolymer, PEO-*b*-PPO-*b*-PEO, surfactant	pulp and paper effluent filtration; increasing the hydrophilic characteristics of the membrane before filtration could reduce the amount of organic foulants adsorbed to the membrane[36]

Table 1 Continued

base material	treatment	function of the membrane
PVC	MPC copolymer (PMB)	adhesion in PRP by fluorescence micrograph; the modified membrane exhibited significant reduction in biofouling[37]
PAN and sodium methallyl sulfonate	heparin	HF and flat-sheet, thrombin in blood dialysis; the modified dialyzer allows for significant reduction of systemic anticoagulant[38]
PS	pluronic triblock copolymer, PEO-b-PPO-b-PEO, surfactant	proteins adsorption, and adhesion in PRP; it was suggested that the bioinert property of PEO segments in the pluronic suppresses the adsorption of plasma proteins and platelets to the coated membranes[39]
PS	MPC and MPC copolymer (PMPU)	HF, reduction in cyctochrome C filtration, adsorption of rabbit blood and bovine serum protein; no adsorption or aggregation of blood cells and protein on the modified membrane was observed[40]
polylactide	MPC copolymer (PLA−PMPC) with various mol % MPC	adhesion in human PRP and leukocytes; it was found that the amount of absorbed protein and adherent blood cell on the polymer surface was decreased with the modification[41]
PVDF	PEO-b-PA 12 block copolymer	UF spiral-wound, motor oil−water emulsion filtration; the coated membranes exhibited significantly low-fouling properties[42]
PVDF	PVDF-g-POEM graft copolymer	oleic acid−triethanol amine−water filtration; modified membranes exhibit nanoscale size selectivity with good wetting properties[43]
NF and RO membranes from Sae-Han Corp. (Seoul, Korea)	PVA	NF and RO, treatment of humic acid solution, and dyeing wastewater effluent; the coated membrane significantly reduced fouling[44]
PVDF and acrylic copolymer	silicon-based surfactant and polyester	primary sewage effluent filtration; permeate flux reduction during filtration was most likely due to cake buildup, while internal fouling was limited[45]
poly(tetrafluoro ethylene)	sodium alginate−carrageenan blend film via cationic surfactant MTMA	OD, whole milk filtration; modified membrane reduced the fouling by the fat globule proteins of whole milk[46]
PVDF	hydrophilic polymer	UF, BSA and enzyme filtration, and adhesion in human PRP; as the antifouling properties were excellent, the membrane could be cleaned without using any cleaning agent[47]
PVDF	PVDF-g-POEM graft copolymer	NF, sodium alginate, BSA, and humic acid filtration; the modified membrane exhibited no irreversible fouling[48]
PP	PNIPAM−PC−C$_{18}$ copolymer	adhesion of human macrophage-coated films showed lower adherent cells than the control one;[49] however, long periods of storage can change the biocompatibility as well as surface structure
PVDF	poly(cyclooctene)-g-PEG graft copolymer	UF, soybean oil−water emulsion filtration; the stability and lifetime of the coated membrane against oil droplet were good, and after a long run the coated membrane flux crosses over the control[50]
PS and PVDF	coated with poly(cyclooctane-g-PEG)	oil−water emulsion; copolymers-coated films reduced fouling for water purification[51]
microporous PP	quaternization cross-linking hydrophilic and positively charged coating by p-xylylene dichloride and PDMAEM after plasma pretreatment	BSA and lysozyme filtration; the coated membranes exhibited moderately high resistance to BSA fouling at pH 3.0 and lysozyme at pH 7.4[52]
PAN-400 membranes from Sepro Membranes, Inc. (Oceanside, CA)	coated with hydrophilic PAN-g-PEG graft copolymer	BSA in phosphate buffered saline using dead-end filtration cell; the coated membranes showed a small decline (∼15% after 24 h) in flux in the protein solution as compared to commercial UF membrane[53]
terylene filter cloth	dip-coated with hydrophilic polyrotaxanes−TiO$_2$−PVA, dried, and cross-linked with glutaraldehyde	simulated wastewater containing dissolved milk powder; the coated membranes permanently improved antifouling property[54]
microporous PP	quaternization cross-linking hydrophilic and positively charged coating by poly(ethylene imine), cross-linked with p-xylylene dichloride and quaternized by iodomethane after plasma pretreatment	BSA and lysozyme filtration; the modified membranes resisted effectively protein fouling below the isoelectric point;[55] furthermore, MF characteristic has been unchanged
commercial TFC PA from GE Water and Process Technologies (Minnetonka, MN)	PEG-based hydrogels using PEGDA as the cross-linker agent and PEG acrylate or 2-hydroxyethyl acrylate or AA as comonomer	RO spiral-wound modules, model oil/water emulsions based on either a cationic DTAB or an anionic SDS surfactant with n-decane; the coated membranes exhibited less flux decline, and fouling resistance was independent of the pendant chain length of copolymer[56]
TFC PA from the Development Center of Water Treatment Tech. (Hangzhou, China)	multilayer formation of electrostatic self-assembly of poly(ethylene imine)	commercial low-pressure RO membrane, aqueous solution with a cationic DTAB surfactant; the coated membranes demonstrated improved fouling resistance[57]

Table 1 Continued

base material	treatment	function of the membrane
PES	PEGDA and trimethylolpropane trimethylacrylate via a thermal-induced surface cross-linking process	BSA filtration; modified membranes were less susceptible to fouling and had greater flux recoveries after cleaning as compared to control membrane[58]
PVDF	dip and surface flow coated chitosan	BSA adsorption and filtration; the higher flux recovery was observed for modified membranes;[59] membrane modified by combined dip and surface flow methods displayed the best antifouling properties
PVDF	coated with PVA and cross-linked by glutaraldehyde vapor	BSA filtration and natural water of Grand River (Kitchener, Canada); the reduction in BSA solution flux was lowered remarkably, and higher flux and slower fouling rate were noticed during natural water filtration by modified membrane[60]
TFC RO PA FT-30 from FilmTec Corp. (Edina, MN)	coated with comb or brush branched poly(alkylene oxide)	oil/soap (SDS and dodecane); the modified membranes exhibited less fouling[61]
Surface Chemical Reaction		
PAN	formaldehyde in presence of acid catalyst	protein filtration; the modified membranes exhibited excellent antifouling properties[62]
PE	hydrophilization by ethanol−water−inorganic acid	treatment of pure water for long time; the membrane hydrophilized by ethanol−water−inorganic acid showed much better initial flux and lower flux decline than membrane hydrophilized by ethanol alone[63]
PE	sulfonation by chlorosulfonic acid	treatment of pure water for long time; the modified membrane with low degree of sulfonation showed better flux as compared to the control ones[64]
PS	fluorination by fluorine gas	potato waste stream filtration; the modified membranes were provided in low fouling properties[65]
titania and zirconia inorganic membrane	phosphoric acid and alkyl phosphoric acid	UF, reduction in BSA rejection; increase in membrane hydrophobicity improves BSA rejection[66]
PA	glycidyl methacrylate treatment	UF, reduction in HAS; the grafting occurred at the outer surface of the membrane as well as at the pore walls in the interior membrane matrix;[67] polyaramid membrane was less sensitive to protein adsorption as compared to the mixed aliphatic-aromatic and the aliphatic membranes
PA	hydrofluoric acid and fluorosilicic acid	TFC, treatment of aqueous isopropyl alcohol solution; the short-term chemical treatment was useful approach for surface modification;[68] limited hydrolysis and fluorination were responsible for improvement of membrane performance
CA	sodium hydroxide in methanol and hydrogen peroxide solutions	humic acid filtration; the adsorption of humic acid was increased on the oxidized membranes[16]
PAN	sodium hydroxide treatment	BSA filtration; pore diameter is reduced with treatment, and hence salt rejection increases[69]
PVA	cross-linking with glutaraldehyde	hydrogel, treatment in creatinine, Fab, and IgC proteins; selectivity of creatinine over IgC increased as modification time increased;[70] selectivity of Fab over IgC initially increased but subsequently decreased with modification time
PS	CO_2 plasma treatment	BSA filtration; the modified membranes showed excellent protein transport kinetics[71]
PS	N_2 plasma treatment	BSA filtration; the N_2-plasma-modified membrane protected against fouling over a wide range of solution pH[72]
PES and sulfonated PS	UV treatment	NF, flux reduction in NOM solution; fouling of membrane was reduced significantly[73]
PS	O_2 plasma treatment	aqueous gelatin filtration; for oxygen plasma-treated membranes, the flow rates of pure water and gelation solution increased at all pH values[74]
PES	ion beam irradiation	large organics, and bacteria filtration; modification led to the reduction in charge of the membrane;[75] NOM fouling was reported to become reversible after modification
PP	CO_2 plasma treatment	MBR, activated sludge from wastewater treatment plant; modified membranes exhibited better flux recovery after cleaning than unmodified membranes[76]
PP	NH_3 plasma treatment	MBR, HF and flat-sheet membranes, synthetic wastewater; modified membranes by NH_3 plasma treatment exhibited better flux recovery after cleaning than control membranes[77]
PES	N_2, NH_3, Ar/NH_3, and O_2/NH_3 plasma	reduction in protein fouling; modified membranes showed increased water flux, reduced protein fouling, and greater flux recovery after gentle cleaning when compared to unmodified membranes[78]
PES	corona and O_2 plasma	MBR, wastewater treatment; short-term simulation results suggested that oxygen−plasma membrane had great potential[79]
CA NF (DS5DL) from Osmonics (Minnetonka, MN)	hydrofluoric acid	purification of industrial phosphoric acid; the modified membrane was useful for phosphoric acid, which contained ionic impurities;[80] increment of flux and improvement of rejection were also observed

Table 1 Continued

base material	treatment	function of the membrane
PS	potassium persulfate treatment	pulp mill effluent treatment; the surface-modified membrane significantly reduced adsorptive fouling, and water flux retention was increased[81]
PAN	organic bases (ethanolamine, triethylamine) and inorganic bases (NaOH, KOH) treatment	BSA filtration; the surface-modified membrane significantly increased the water flux and reduced the BSA rejection;[82] the reaction conditions (temperature and time) of the modification process were also optimized
PP	N_2 plasma treatment	MBR, activated sludge from wastewater treatment plant; after water cleaning, the flux recovery of the modified membrane was higher than the control[83]
poly(vinyl butyral)	hydrochloric acid treatment	UF, BSA filtration; the antifouling property improved due to the hydrolysis of butyral and acetate group;[84] the reaction time and concentration of hydrochloric acid are optimized
PAN	sodium hydroxide treatment	UF, Salvia Miltiorrhiza decoction filtration; the antifouling property increased as the hydrophilicity increased[85]
PAN, PES, and blended PAN−PES	contacting with aqueous NaOH solution at room temperature for 24 h	solutes containing dextran, PEG, and PSSS filtration; the modified membranes exhibited high flux recovery[86]
PP	CO_2, H_2O, NH_3, plasma treatment	BSA filtration; antifouling properties were improved for plasma-treated membranes[87]
	Surface Grafting	
PES, PS, and PVDF	grafting of cellulosic polymers containing hydroxyl or amine group	bovine albumin, dextran, and triton X-100 filtration; higher flux values on the order of 20−50% are observed against unmodified membranes[88]
PS	UV irradiation in the presence of block copolymer, dextran sulfate, and dimethyl-aminoethyl dexane	UF, flux reduction in BSA, lysozyme, and whey protein solutions; the flux of modified membrane increased by about 400% as compared to the control without sacrificing retention[89]
sulfonated PES	AA grafting by plasma polymerization	bleach effluent filtration; the grafted membrane exhibited lower fouling as compared to the control[90]
PS	grafting with PEG with acrylate functional groups at both ends	UF, flux reduction in olive oil−water emulsion; the modified membrane exhibited significantly less fouling[91]
PS	PEGs (various kD) grafting by γ-ray irradiation	UF-HF, reduction of adsorption of organic contaminants; the modified membranes showed better filtration performance as compared to the original ones[92]
poly(aryl sulfone)	grafting with various vinyl monomers	MF and UF, treatment of BSA solution; modified membranes exhibited low- or nonfouling properties[93]
PAN and PS	grafting with AA, MA, and HEMA	BSA filtration; modified membranes were found to have improved performance in protein ultrafiltration over control membranes[94]
PAN	UV assisted grafting AA, HEMA, PEGMA, and POEM with various kD	protein treatment by UF, fouling less than negatively or positively charged membrane, and adsorption in BSA; protein−polymer surface interactions were diminished by the surface modification[95]
PS	UV exposure, binding with gelatin, and collagen proteins	UF, treatment of BSA, and myoglobin solutions; the most promising modifier was found to be hydrolyzed gelatin[96]
PE	UV-assisted grafting AA	adhesion in PRP; platelet compatibility of the membranes was affected by the presence of various functional groups on the film surface[97]
PES	NVP, NVF, and N-vinyl caprolactam	BSA filtration; NVP-modified membrane showed the best performance in terms of low fouling and high flux[98]
PS	AA grafting	BSA filtration; the modified membranes contained short chains of PAA, which was of brush-like structure; the modified membrane showed better performance under basic environments[99]
PS	plasma-treated HEMA	lysozyme from hen egg-white filtration; the modified membrane exhibited lower adsorption of protein[100]
PES	UV-assisted grafting NVP	UF, flux reduction in BSA solution; combination of irradiation and NVP caused a more severe loss of observed rejection than irradiation alone[101]
PES	UV-assisted grafting NVP	NF, flux reduction in NOM solution; enlargement of pore structure was observed under long irradiation times[73]
PP	CO_2 plasma, grafting AA, and PDADMAC	MF, adsorption in HAS; the modified membrane layer of grafting by improved permeate flux, but the protein rejection did not change substantially[102]
PP	AA, DMAEM, and PEGMA	*Escherichia coli* bacterial, and carboxylate-modified latex particles suspensions; with backpulsing, considerable improvement was observed with surface-modified membranes[103]
PP	ozone-induced grafting, HEMA	MBR, treatment of synthetic wastewater; the modified membrane exhibited the highest flux[104]
PP	grafting γ-ray induced, HEMA	BSA filtration; the grafted membrane surface prevented the intimate contact of BSA proteins[105]
PP	plasma-treated AA and allyl amine	BSA filtration; excess power damaged the membrane surface; longer plasma treatment time resulted in more plasma coating and micropore blocking[106]
PP	grafting γ-ray induced, HEMA	BSA filtration; modified membrane showed better flux recovery after cleaning, higher solution flux, and lower BSA adsorption[107]

Table 1 Continued

base material	treatment	function of the membrane
PE	PDMS, PEG (1 kD), and perfluoroheptane	affinity in *Pseudomonas aeruginosa* bacteria; the membrane surface roughness and hydrophobicity increased as the biofilm was initiated by bacteria;[108] the biofouling was at a minimum value when the surface charge was minimized
PA	polyalkylene oxide, PEG diepoxide	surfactants (DTAB and SDS), and tannic acid filtration; the modified RO membranes improved resistance to fouling and offered excellent flux and separation performance[109]
aromatic PA	grafting by redox initiator, sulfopropylmethacrylate, and PEGMA	seawater/surface water filtration; modified membranes adsorbed less organic material and were more easily cleaned than unmodified membranes[110]
zirconia and carbon-supported zirconia inorganic membranes	grafting of NVP via silylation reaction	synthetic oil−water emulsion and microemulsion filtration; modified membranes maintained their hydraulic permeability even after many filtration runs as compared to unmodified membrane, which was fouled after just one run[111]
zirconia inorganic membrane	grafting of NVP	adsorption of lysozyme protein; maximum adsorption capacity of the membrane decreased by up to 76% due to surface modification[112]
PVDF	PEG (1 kD) and POEM	γ-globulin filtration, and protein adsorption; permeate flux decreased with increasing surface concentration of the grafted PEG polymer, while pore size remained unchanged[113]
PES	argon plasma treatment followed by AA	pure water flux of modified membranes was tremendously increased by plasma treatment; also, modified membranes were easier to clean and required little caustic to recover permeation flux[114]
PES	UV-assisted grafting NVP and 2-mercaptoethanol	UF, treatment of BSA solution; although protein rejection remained unchanged after modification, the permeability of the membranes decreased with increasing degree of grafting[115]
PES and PS	UV-assisted grafting NVP	UF, treatment of NOM solution; the modified membrane exhibited significantly lower propensity to fouling; the pore blockage/cake filtration model was established[116]
PS	argon plasma, grafting allyl amine	BSA filtration; the amphoteric character of the modified membrane by allyl amine protected against peptide sorption[117]
PP	AA, DMAEM, and POEM	UF-HF, flux reduction in NOM solution; the flux of graft-modified membranes depends on the solution chemistry of NOM feedwater[118]
PVC	adsorption of NVP and plasma treatment	UF, flux reduction in sludge solution; the specific resistance of the adsorbed cake layer decreased with the increase of grafting degree[119]
Nafion 117, functionalized PA-6, and PP	CO_2 plasma grafting AA, and multilayer assemblies by PAA, and polycation, PDADMAC, and branched poly(ethylene imine)	MF and PV, adsorption in HAS protein; permeate flux improved with the layer of grafting, but the protein rejection did not change substantially[120]
PES	argon plasma treatment followed by acrylamide	adsorption of BSA protein; grafting of the membranes made them less susceptible to BSA fouling and improved their flux recoveries[121]
PVC	adsorption of NVP and UV treatment	MF, flux reduction in sludge solution; the adhesive interactions between mixed liquor−suspended solid and modified hydrophilic surfaces were weaker than those of control membrane[122]
PP	N_2 plasma-induced grafting of sugar containing monomer	MF, flux reduction in BSA protein solution; membrane hydrophilicity increased significantly with grafting;[123] membrane grafting also resulted in higher flux and increase fouling resistance to BSA
PES	UV-assisted grafting of NVP	UF, flux reduction in BSA filtration; the surface-modified membrane effectively reduced irreversible fouling; the grafting protocol was also optimized[124]
PES	UV-assisted grafting of various vinyl monomers	UF, flux reduction in NOM solution; reversible fouling resulting from cake formation was only weakly dependent on membrane surface chemistry[125]
PVDF	POEM grafting by RAFT	MF, flux reduction in γ-globulin solution; the modified membranes displayed higher resistance to γ-globulin fouling as compared to control membranes[126]
PES and PVDF	grafting of AMPSA and quaternary DMAEM	affinity in *E. coli* bacteria; the modified membranes significantly reduced the biofouling[127,128]
PP	grafting of NVP and γ-stearyl-L-glutamate through bridging of siloxane oligomer	reduction in BSA filtration and adsorption, and adhesion of human PRP; amino groups were introduced onto the PP membranes by ammonia plasma treatment[129]
PS	grafting, various vinyl monomers	UF, reduction in BSA solution; NVP, AMPS, and AA monomer grafted membranes showed high protein retention, high solution flux, and low irreversible fouling[130]
PP and PS	UV grafting of AA	UF and MF, adsorption of BSA; the modified membrane exhibited lower adsorption of protein as compared to the control[131]
PET and PVDF	grafting of 4-vinyl pyridine	increase in antibacterial resistance; same efficiency in killing *E. coli* on contact was observed for the modified film after at 60 day storage in air[132]

Table 1 Continued

base material	treatment	function of the membrane
PVDF	grafting of inimer by ozone treatment and SSS grafting by ATRP	MF, γ-globulin adsorption; membrane exhibited substantially improved antifouling properties[133]
PES	oxygen plasma grating of AA, acetylene, diamino cyclohexane, and hexamethyl disiloxane	UF, PWP with long time; the hydrophilic membranes have higher initial flux as well as improved fouling resistance; the hydrophobic membranes displayed both lower flux and lower fouling resistance[134]
PS	AA, C_4 monomer, and DMAEM	treatment of BSA, and lysozyme solutions; the membrane modified with a positively charged monomer reduced adsorption of positively charged lysozyme;[135] similarly, the membrane modified with a negatively charged monomer reduced adsorption of negatively charged BSA
PES and PS	UV grafting, various vinyl monomers	UF, treatment of protein filtration; modified membranes exhibited low protein fouling but almost similar permeability and retention properties;[136] shorter grafted monomer chain lengths and high density grafted monomer resulted in low protein fouling
CA	PEG grafting through persulfate initiation	UF, treatment of textile auxiliaries; the modified membrane showed a decrease in the fouling tendency, and it acts as an antiwrinkle agent[137]
PP	adsorption of NVP and plasma treatment	MF, treatment of BSA solution; the preadsorption plasma approach was found to be facile and useful in improving the hemo-compatibility and antifouling property of the membrane[138]
PP	adsorption of GAMA and UV irradiation	MF, treatment of BSA solution; modified membranes were found to have higher water and protein solution fluxes, lower BSA adsorption, and better flux recovery after cleaning[139]
PES	electrophoresis-UV grafting of AA, AAG, HEMA, MA, and NVF	MF, treatment of surface water and NOM; the control of the membrane surface properties was performed by a combination of electrophoresis and UV grafting[140]
PA and polyester	plasma-grafted PEG	adhesion, treatment of *Listeria monocytogenes*; modified membrane inhibited significantly biofilm formation[141]
chitosan	plasma-grafted AA and vinyl sulfonic acid	adhesion, treatment of osteoblast-like cells; vinyl sulfonic acid grafted membrane substantially improved cell adhesion and proliferation as compared to the AA-grafted or control chitosan membrane[142]
PP	air-/O_2-/Ar-/CO_2-/H_2O-plasma grafting of PVP	MF-HF, activated sludge filtration; the surface-modified membrane had higher total surface free energy and had excellent antifouling properties[143]
PP	UV grafting of AA	MF-HF, activated sludge filtration; the surface-modified membrane had better filtration properties in the MBR[144]
PES and PVDF	UV grafting of AMPSA, quaternized DMAEM, and HEMA	MF, *E. coli* suspension filtration; the hydrophilic surface was less susceptible to fouling than the hydrophobic surface[145]
PES	argon plasma grafting of AA and HEMA	UF, wastewater treatment; the short-term-modified membrane exhibited better antifouling properties, but the long-term-modified membrane exhibited worse antifouling properties[79]
PP	UV grafting of HEMA	MF, BSA solution filtration, and BSA adsorption; the membrane showed good protein resistance as well as better hemocompatibility potential[146]
PES	UV grafting of PEGMA	UF, treatment of sugar cane juice; the optimized processes for low fouling conditions are relatively high monomer concentration and medium irradiation time[147]
PES	UV grafting of PEGMA and SPE with and without cross-linker MBAA	UF, treatment of myoglobin and humic acid solution; zwitterionic composite membrane showed better performance than the composite membrane made by PEGMA[148]
PES	corona-induced grafting of AA	UF, treatment of BSA solution; modified membranes showed significant antifouling properties, which are caused by the adhesion of proteins[149]
PP	UV grafting of acrylamide	MF-HF, synthetic wastewater filtration; the modified membrane exhibited much better regeneration and flux recovery[143]
PP	plasma-treated AA and amino-PEG	BSA and fibrinogen filtration; a 95% reduction in protein adsorption was exhibited for longer PEG chains[150]
PVDF	UV-treated MMA and PEG methyl ether methacrylate by reverse ATRP	MF, BSA filtration; PEG methyl ether methacrylate grafted membrane had more effective antifouling property than MMA grafted membrane, as observed in their hydrophilicity behavior[151]
PPESK	POEM grafted through ATRP	UF, BSA filtration; the antifouling ability of modified membrane significantly improved as compared to control ones due to the presence of brushes of comb-like polymers[152]
PS	methyl acrylate monomer grafted through UV irradiation	UF, BSA filtration; the antifouling property is improved due to methyl acrylate addition of the PS chain[153]
CA	grafting of PEG by sodium persulfate oxidizing agent	synthetic seawater filtration; higher flux values by 15−25% were observed against virgin UF membrane[154]

Table 1 Continued

base material	treatment	function of the membrane
PP	AA grafted through benzoyl peroxide initiator	BSA adsorption and filtration; antifouling property of the grafted membrane was dramatically improved[155]
PES, PVDF, and PES/PVDF blend	UV grafting of NVP	BSA adsorption and filtration; lower BSA adsorption and higher antifouling abilities were observed for grafted membranes;[156] an easy regeneration of modified membranes by an alkaline medium due to mostly reversible fouling was noticed
PVDF	UV grafting of AA, HEMA, MPD, and ethylene diamine	pasteurized and homogenized milk with 3.2% protein and 1.5% fat; antifouling properties (flux recovery, irreversible flux loss, total flux loss, and fouling resistance) of modified membranes were enhanced[157]
PVDF	grafted zwitterionic sulfobetaine methacrylate via ozone surface activation and ATRP	adsorption and filtration of BSA and γ-globulin; grafted membrane hardly adsorbed BSA, but slightly adsorbed γ-globulin;[158] filtration data of BSA showed perfect nonfouling characteristics; however, γ-globulin exhibited slightly higher membrane fouling
PS	UV grafting of MPDSAH inner salt	BSA filtration; antifouling properties of modified membranes were enhanced with increasing of grafting degree;[159] better antifouling property was exhibited in a wider pH range from 4.5 to 10.0
PVDF	grafting of PEGMA via ozone surface-initiated thermal-induced radical copolymerization, ATRP, and low-pressure plasma-induced polymerization	adsorption and filtration of BSA; grafting structure of brush-like PEGMA led to lower protein adsorption and better antifouling for BSA filtration than that of network-like PEGMA on membrane surface;[160] adsorbed proteins on the modified membranes were associated with the surface grafting structures
PP	UV-induced grafting of AA, acrylamide, and GAMA	synthetic wastewater filtration; antifouling was attributed to an increase in the grafting chain length[161]
	Incorporation of Hydrophilic Polymer	
PEI	PVP in casting solution	MF, treatment of BSA filtration; the PVP in the membrane matrix of the modified membrane prevented BSA adsorption, which was studied with ^{14}C-labeled BSA;[162] the heat-treated modified membrane offered nonfouling behavior, which was similar to CA membrane
PMMA	copolymers of methoxy PEGMA, MMA, and vinyl acetate (or AN) in casting solution	adhesion in PRP; the adhesion of platelets of the modified membrane was remarkably reduced both in flat sheet and in HF;[163] the modified membranes exhibited excellent antithrombotic property
PS	PVP in casting solution	UF-HF, less blood protein adsorption; modified membrane showed suppressed adsorption of platelets on the surface than PS and other surface modified membranes[164]
PES and PS	PEO/PPO-substituted ethylene diamine in casting solution	MF-HF, milk filtration; the modified membranes provided low fouling and could be easily cleaned by water;[165] the pore size of the membrane was 0.1−1 μm
PS	PVP in casting solution	BSA, and DL-histidine (DLH) protein filtration; BSA fouls the membrane externally, while DLH fouls it internally;[166] BSA fouls the non-PVP-modified membranes faster, while DLH fouls them more slowly
PS	PVP in casting solution	HF, reduction in cyctochrome C filtration, adsorption, of rabbit blood and bovine serum protein; the modified membrane showed a reduced adsorption of protein as compared to the control[40,41]
PES	PEO-b-PPO-b-PEO triblock copolymers in casting solution	UF, BSA filtration; the irreversible fouling was remarkably reduced, and modified membrane had an excellent flux recovery ratio[167]
cellulose	PEG in casting solution	UF-HF, flux reduction in the machine oil−water emulsion; modified membrane was not only resistant to fouling but also tolerant to a wide pH range[168]
PAN	zwitterionic, PAN-co-PDMMSA-co-PDMAEM in casting solution	UF, BSA filtration; by simple water flushing, the recovery of the flux ratio could reach 95% for the modified membrane[169]
PES	branched amphiphilic copolymers with PEG arms in casting solution	UF, BSA filtration; superior fouling resistant ability was exhibited; more specifically, irreversible fouling decreased remarkably in the modified membrane[170]
PPESK	amphiphilic, PPESK-g-PEG, graft copolymer in casting solution	UF, BSA filtration; the enrichment and arrangement of the PEG chains are responsible for antifouling properties;[171] the adsorption and deposition of protein molecules are decreased due to the repulsive forces that originate from the change of conformation and desolvation of the PEG chains
PVDF	amphiphilic hyperbranched-star of about 12 hydrophilic arms polymer in casting solution	BSA adsorption and filtration; by enrichment of hyperbranched star polymer, the antifouling properties improved substantially;[172] modification also resulted in higher water flux recovery
PVDF	PVDF-g-PEG graft copolymers	UF, treatment of sodium alginate, BSA, and humic acid filtration; the modified membranes completely resisted irreversible fouling;[173] the antifouling ability of the PEO brush is

Table 1 Continued

base material	treatment	function of the membrane
		due to the energetic barrier of the adsorption process that is brought about by the affinity of PEO to water and steric entropic barrier of PEO chains on the membrane surface
PES	sulfonated and pegylated PES in casting solution	UF, BSA filtration; the antifouling behavior of sulfonated PES blended membrane depended upon pH, whereas the presence of pegylated PES reduced fouling in a wider pH range[174]
PES	Pluronic F127, PEO–PPO–PEO triblock copolymer in the casting solution	UF, BSA and lysozyme filtration; the total fouling and irreversible fouling of the modified membrane remarkable decreased due to micelle formation and aggregation behavior of Pluronic F127[175]
PVDF	glucose-carrying methacrylate grafted via ATRP	adsorption, treatment of BSA solution; the modified membrane exhibited antiprotein-adsorption property[176]
PS	adding polyaniline nanofibers in casting solution	UF, BSA filtration; modified membrane showed lower flux decline rate and higher flux recovery as compared to the control PS membrane[177]
PES and TA-PES	adding PEG 2kD in casting solution	UF, BSA filtration; the TA-PES membranes displayed superior antiprotein-fouling resistant property due to the inherent zwitterionic characteristics of TA-PES[178]
PS	amphiphilic triblocks copolymers blended in casting solution	UF, BSA adsorption; the additive blended PS membranes displayed improved protein resistance as compared to unmodified PS membrane[179]
PES	adding poly(amide imide) (PAI) in casting solution	UF, milk–water filtration; the antifouling properties of the PAI blended PES membrane were improved, and the mechanical strength was also higher as compared to the control PES membrane[180]
PES	blending CTA in the casting solution	DNA solution filtration; the recovery rate was higher for modified membranes[181]
PPESK	blending amphiphilic PPESK-*g*-PEGMA in casting solution	UF, BSA; the antifouling properties of the blended membrane were significantly improved[182]
PES	adding polyoxyethylene sorbitan monolaurate (Tween-20) surfactant in casting solution	UF, BSA; irreversible and total fouling were decreased, and also excellent flux recovery was rendered for the modified membranes[183]
	Incorporation of Nanoparticles	
aromatic PA	TiO$_2$ nanoparticles deposition	*E. coli* filtration; a novel antibiofouling (antibacteria/-virus) composite membrane was developed by hybridized quantum size nanoparticles, which has excellent candidature for RO applications[8,9,184]
PAN, PES, and PVDF	TiO$_2$ nanoparticles deposition and entrap	activated sludge filtration; the TiO$_2$ entrapped membrane showed better antifouling properties than the control;[185] the preparation TiO$_2$ immobilized membrane was simple and is a useful method for fouling mitigation in MBR applications
PES	TiO$_2$ nanoparticles incorporated by self-assembly	treatment of pure water for a long time; composite membrane exhibited good separation performance and has antifouling potential[5]
sulfonated PES	TiO$_2$ nanoparticles self-assembled	MBR, activated sludge filtration; the TiO$_2$ self-assembled membrane showed better antifouling properties[186]
PVDF	Al$_2$O$_3$ nanoparticles and surfactant hexad-sodium phosphate incorporated	UF, treatment of oil–water waste from oil field; composite membranes exhibited significant differences in surface and intrinsic properties due to the addition of nanoparticles[6,187]
PPESK	TiO$_2$ nanoparticles entrapped	UF, flux reduction in BSA protein solution; the protein fouling in the modified membrane was reversible and easily cleaned[7]
PS	TiO$_2$ nanoparticles	UF, reduction in kerosene emulsified wastewater; the nanoparticle increased the oil–water interfacial tension so that both flux and deoiling rate increased;[188] the antifouling performance was exhibited at 1–2 wt % TiO$_2$ content
poly(L-lactic acid)	chitosan and dextran sulfate stabilized silver nanoparticles	adhesion, the modified membrane exhibited antibacterial activity against Methicilin-resistant *Staphylococus aureus*; the human endothelial cell attachment, proliferation, and viability depended on the layers of the coated assembly[189]
PES	TiO$_2$ nanoparticles entrap and UV irradiation	UF, treatment in nonskim milk; the initial PWP and milk water permeation of modified membrane were low as compared to the control[190]
PVDF	Al$_2$O$_3$ nanoparticles entrapped during the immersion precipitation technique	UF, oily wastewater filtration; the modified membrane exhibited superior fouling resistance quality[191]
poly(styrene-*alt*-maleic anhydride)/PVDF blend	blended membranes immersed into TiO$_2$ nanoparticles solution for a week	UF, BSA filtration and absorption; antifouling property significantly improved for the modified membranes[192]
PES/polyimide blend	blended membranes immersed into diethanolamine, TiO$_2$ nanoparticles suspension, and UV radiation	UF, BSA filtration; the flux recovery was increased for modified membranes containing diethanolamine and TiO$_2$ nanoparticles[193]

Table 1 Continued

base material	treatment	function of the membrane
PA TFC from Dow-Filmtec (Midland, MI)	coated with silver nanoparticles made from silver nitrate	spiral-wound modules (SW30HR-380), microbial adherence, and multiplication by red fluorescent (PKH-26) dye labeling; the growth of microbes was affected by silver-coated membranes;[194] simple silver redox reaction provided biofouling for seawater desalination
		Miscellaneous
TFC membrane: polyester support, PS inter, and PA active layers	deposition of colloidal elementary silver	disc-tube modules, surface pond water filtration; the higher permeate recovery was obtained in the modified membrane[195]
polysilicon	micromachining	albumin protein filtration; uniform pore sizes are possible, which improve permeability and selectivity[196]
PS	DNA blended	suppression of the human PRP adhesion; the biopolymer, DNA, contributes to hydrophilic and lower protein adsorption[197]
PS	sulfonated PEG acrylate diblock copolymers blended	adhesion of human PRP; the sulfonated diblock copolymer enhanced hydrophilicity and long-term stability more than the copolymer having no hydrophobic block[198]
PDMS	microdots of aluminum deposited	adhesion of fibroblast cells; a direct correlation between the micropatterned membrane and the adhesion of fibroblast cells is observed[199]

grafted surfaces are stable, chemicals used for grafting are sometimes environmentally unfriendly. Grafting by gamma ray and UV irradiation, or in the plasma chamber, is not easy to apply on a large industrial scale. It is known that poly(N-vinyl-2-pyrrolidone) (PVP), when blended to polysulfone, is concentrated at the membrane surface, rendering the surface more hydrophilic. However, PVP will eventually be leached out while being used in UF.[3]

There are many reports where it has been shown that hydrophobic surface favors less fouling. For example, we have shown experimentally that a smaller amount of humic acid is deposited onto a membrane modified by hydrophobic additive.[4] A fouling test with humic acid exhibited that the modified membranes suffered from reduction in permeation flux less than that of the control. It is also noted that the pore size and the MWCO of the modified membranes are lower than those of the control poly(ether sulfone) membrane.

There is a trend in the latest reports to blend nanoparticles in the polymeric material, by which an increase in hydrophilicity is expected. The higher hydrophilicity was indeed confirmed by the contact angle measurement.[5-7] Additional advantages of nanoparticle blended membranes are as follows: (1) the flux of the nanoparticle membranes is very high due to the larger effective surface area of the membrane caused by the nodular shapes having ridges and valleys as of atomic force microscope study;[8] and (2) when TiO_2 nanoparticles are blended, photocatalytic antibacteria/antivirus activities of TiO_2 occur simultaneously.[9] Most of the nanoparticles like TiO_2, Al_2O_3, etc., are metal oxides, which could easily be hydrolyzed to form hydroxides. TiO_2 magnifies the antibacterial activities of UV as TiO_2 generates peroxide redicals. In the absence of TiO_2, a higher dose of UV light is needed to neutralize bacterial activities.[10]

It should noted that we have cited literature on antifouling of surface-modified membranes, and not on antifouling polymer of the solid-modified surface such as Au, Si, Si/SiO_2, glass, mica, stainless steel, etc.

1.2. Surface Charge

It is also generally accepted that the repulsive forces working between the charged surface and the co-ions in the feed solution prevent the solute deposition on the membrane surface, thus reducing the fouling. However, no definite correlations, such as shown in Figure 1 for hydrophilicity, can be found between the surface charge density and the fouling. Nevertheless, there have been a number of attempts to reduce fouling by incorporating ionizable functional groups, as summarized in Table 2. In most cases, surfaces are negatively charged. It seems natural, considering that most of the colloidal particles such as NOMs that deposit on the membrane surface are negatively charged. However, attempts were made recently to develop positively charged membranes. Similar to the negatively charged surface membranes, the positively charged membrane surfaces behave as barriers for co-ions (cations) due to dominance of exclusion mechanism (Ulbricht et al.[203]); for example, the positively charged membrane surface exhibited repulsion against positively charged proteins (Kato et al.[218]). Recent experiments suggest that the addition of zwitterionic charged material can be more effective for antifouling surface as compared to the conventional positively or negatively charged membrane surface.[148]

1.3. Surface Roughness

The effect of surface roughness on the flux is also one of the unresolved issues. Elimelech et al.[219] found that the commercial TFC membrane they used fouled faster than the cellulose acetate membrane. They concluded that the observations occurred due to the greater surface roughness of the TFC membrane. Hirose et al.[220] showed a correlation between the membrane flux and the surface. The increase in flux with an increase in surface roughness was attributed to the increase of the area available for the membrane transport. Vrijenhoek et al.[221] on the other hand, showed a strong correlation between the fouling and the surface roughness for some RO and NF membranes, as shown in Figure 2.

This was attributed to the enhancement of interactions between colloidal particles with an increase in surface roughness (Elimelech et al.[219]); that is, colloidal particles preferentially accumulate at the valleys of the rough membrane surface. As a result, valleys become blocked and fouling becomes more severe for the rougher membrane surface (Vrijenhoek et al.[221]). However, contradictory results are also reported for organic particles (Riedl et al.;[222] Yan et al.[187]). Most probably, the adhesive forces (organic particles−membrane surface interaction, organic particles−

Table 2. Reduction of Fouling by Increasing Surface Charge

base polymer	treatment	function of the membrane
	Surface Negatively Charged	
sulfonated PS	sulfonic acid (surface negatively charged)	BSA, cytochrome, and myoglobin filtration; the myoglobin protein was rejected by the negatively charged membrane[200]
PS	propane sultone and propylene oxide	UF-HF, flux reduction in BSA and PEG-6 kDa solutions; the modified membrane showed better antiadsorption of solutes than did the control fibers[201]
CA	modified during casting	RO, treatment in wastewater, and brackish water; modified membranes showed better flux stability than conventional brackish water composite membranes[202]
PAN	grafting with carboxylic acid and sulfonic acid	protein filtration, fouling reduction more than positive charge; membrane surface with permanently anionic (sulfonic) and ionizable (carboxyl) group attachment membrane surface exhibited good protein antifouling properties[203]
PS	coated with sulfonated poly(phenylene oxide)	pulp and paper wastewater filtration; the fouling was lower in the modified membrane as compared to the control[204]
(1) PS (2) sulfonated PEES-b-PES block copolymer	increasing component 2 (surface negatively charged)	bacterial attachment; membrane flux was inversely correlated with PS content in contrast with cell attachment, which was directly correlated with PS content[205]
(1) PS (2) sulfonated PEEK	increasing component 2 (surface negatively charged)	humic acid filtration; blended membranes were found to have high porosity, charge, and pore size at the boundary of NF and UF membranes[206]
(1) PEI (2) sulfonated PEI	increasing component 2 (surface negatively charged)	BSA filtration, fouling reduction increase with increasing negative charge; the membrane fouling in the modified membrane was caused by reversible BSA adsorption[207]
(1) PAN (2) poly(AN-co-sulfopropyl acrylate potassium salt)	increasing component 2 (surface negatively charged)	BSA filtration, fouling reduction increase with increasing negative charge; as the concentration of component 2 increased, the membrane became more hydrophilic and the top layer of the asymmetric membrane increased in thickness[208]
(1) PEI (2) sulfonated PEEK	increasing component 2 (surface negatively charged)	increase in pure water permeation, and separation performance; modification improved the permeability and hydrophilic properties of the membrane[209]
(1) poly(AN-co-vinyl acetate) (2) poly(AN-co-vinyl acetate-co-sodium p-sulfophenyl methallyl ether)	increasing component 2 (surface negatively charged)	waste machine cutting fluid, oil–water filtration; the negatively charged membrane showed better antifouling properties than did the control[210]
cellophane	γ-radiation (surface negatively charged)	BSA filtration; the modified membrane decreased in salt permeability and increased in cationic selectivity[211]
(1) phenolphthalein PES (2) poly(AN-co-AMPSA)	increasing component 2 (surface negatively charged)	BSA filtration; the modified membrane was imparted the surface electrical properties and also resisted the protein fouling[212]
(1) PS and PVP (2) strongly acidic resin (DOWEX 50WX8 from Fluka)	increasing component 2 (surface negatively charged)	commercial emulsive oil, oil–water filtration; porous substructures densified when resin content increased[213]
(1) PVDF (2) sulfonated polycarbonate	increasing component 2 (surface negatively charged)	emulsified oily wastewater filtration; membrane fouling resistance was lowered with increasing surface negatively charged[214]
CA/chitosan blend	silver incorporated heparin modified CA/chitosan (surface negatively charged)	antiadhesion and antibacteria were performed with *E. coli* from an activated sludge bioreactor; modified membrane improved antibiofouling performance due to negatively charged surface and also introduced silver ions[215]
	Surface Positively Charged	
quaternary ammonium PS	amination (surface positively charged)	BSA, cytochrome, and myoglobin filtration; the cytochrome C protein was rejected by the positively charged membrane[200]
PAN	grafting monomers with trimethyl/ethyl ammonium	protein filtration, fouling reduction less than negative charge; positively charged membrane less fouled due to the dominance of electrostatic effects for protein adsorption[203]
regenerated cellulose	pyridine, thionyl chloride, and trimethyl amine treatment (surface positively charged)	BSA filtration; CTA, which is more hydrophobic than regenerated cellulose, shows stronger tendency to adsorb proteins and to fouling[216]
brominated poly(phenylene oxide)	amination (surface positively charged)	gelatin filtration, increase in pure water permeation; increased positive charge on the membrane surface enhanced the pure water flux and increased the antifouling properties[217]

organic particles interaction, etc.) are the important parameters for fouling (Huisman et al.[223]). Moreover, at the initial stage of fouling, particles are brought into direct contact with the membrane surface; hence the organic particles–membrane surface interaction governs the particle deposition. On the other hand, after the gel-layer formation, the interaction between organic particles and organic particles becomes more important. Thus, the effect of the surface roughness on the overall membrane performance is, at best, controversial. Moreover, little is known about the effect of surface roughness on the deposition of smaller organic molecules. Intuition somehow tells us that smoother surface shows less
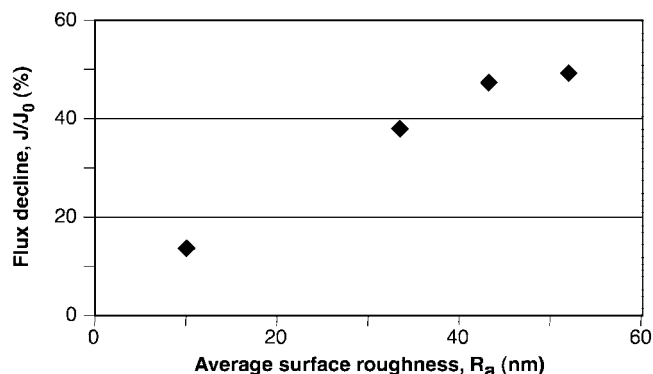
Figure 2. Relationship between relative flux and surface roughness of TFC RO membranes. Measuring conditions: 1500 ppm NaCl solution, 25 °C, 1.5 MPa. Plotted from data in Table 1 of Vrijenhoek et al.[221]

adsorption to the organic molecules, and in general a rougher and heterogeneous surface is better for adsorption, because of the larger suface area, than a smoother surface. The effect of surface roughness on salt precipitation is another unre-solved issue. Interested readers may consult the articles on the surface roughness effect on colloidal particles found in

ref 224. Overall, efforts are currently focused on the reduction of surface roughness. Some pertinent works on the effect of the surface roughness are listed in Table 3.

1.4. Biomimetic Surface

When polymeric membranes are used in biomedical applications, such as hemodialysis, the adsorption of proteins usually stimulates the attachment of fibrous and antibiotic moieties, leading to subsequent biological responses such as thrombus formation and immuno-responses. The extent of protein adsorption is mainly determined by the delicate balance of the interaction between the protein molecules and the membrane surface. Considering the membrane surface alone, the factors that affect the protein adsorption include hydrophilicity/-phobicity balance, surface charge, and surface roughness. These factors are similar to those that have been already discussed in terms of the surface modification for the reduction of membrane fouling. The heterogeneous surfaces that exhibit a microdomain structure for a multi-component system also influence the protein adsorption.

Another approach that has been developed only recently is to mimic the cell membrane. Mimicking the biological cell membrane functions is considered to result in less adhesion for biomacromolecules. It is well-known that the

Table 3. Reduction of Fouling by Decreasing Surface Roughness

base polymer	treatment	function of the membrane
	Better Results with Rough Surface	
PA TFC	change in solvent in in situ polymerization	increase in water permeation rate with rough surface; the increase in flux with an increase in surface roughness was attributed to the increase of the area available for the membrane transport properties[220]
(1) PS and nylon	(1) smooth surface	apple juice, dense surface fouling layer for smooth surface; surface roughness strongly influenced surface fouling layer
(2) PES and PVDF	(2) rough surface	morphology;[222] smooth surface produced dense fouling layer, while rough surface produced a more open fouling layer
PA TFC	change in solvent in in situ polymerization	Iincrease in water permeation rate with rough surface; the modified membrane exhibited lower fouling properties[225]
PVDF	addition of nanosized Al_2O_3 particle	α-amylase solution, less fouling for rough surface; the modified membrane exhibited antifouling properties due to the nanometer particles addition[187]
PPESK	various roughness, addition of POEM brushes	UF, BSA filtration; rougher surface of modified membrane significantly improved the antifouling ability as compared to the smoother surface control[152]
	Better Results with Smooth Surface	
(1) PA TFC	(1) smooth surface	colloidal suspensions, less fouling for CA membrane; the modified membrane showed a strong correlation between the
(2) CA asymmetric	(2) rough surface	fouling and surface roughness[219,226]
PE	various roughness	*P. aeruginosa* bacteria; the modified smoother membranes contributed better resistance to biofouling[108]
PES	various roughness by molecularly imprinted polymer	MF, treatment of cAMP solution; a good correlation between flux reduction versus roughness of the modified imprinted membrane was observed[227]
(1) PES	(1) smooth surface	feed streams of pulp and paper industry, more fouling for rough surface; the control membrane showed the greater adhesion and also greater fouling as compared to the modified membrane[228]
(2) PES and polyacrylate blend	(2) rough surface	
(1) PES	(1) smooth surface	BSA filtration, large flux drop for rough surface; a strong correlation between membrane surface roughness and flux loss was reported;[229] as the surface roughness increased, the adhesive force between membrane surface and proteins also increased
(2) PES and polyacrylate blend	(2) rough surface	
(1) PES	(1) smooth surface	UF, treatment of spent sulphite liquor, large flux drop for rough surface; PS membranes exhibited much larger flux decline than did PES membranes[230]
(2) PS	(2) rough surface	
PA-polyurea TFC	various roughness	RO, lake water, and simulated aqueous solutions of silica, humic acid, and $CaCO_3$; the modified smoother membranes contributed better resistance to fouling[231]
PES	various roughness by rigid star polymer via PVA bridging	NF, organic contaminants surrogate water-soluble dye (Rhodamine WT), and arsenic, As(III); by molecular deposition of 6-arm star oligomers, antifouling membrane of smooth surface was achieved[232]

Table 4. Reduction of Fouling by Biomimetic Surface

base polymer	treatment	function of the membrane
	Attachment through Covalent Bond	
CA	chemical, protease enzyme	UF, treatment of hemoglobin (or BSA) solutions; necessary to optimize the immobilized enzyme layer by changing the conditions or even the methods of enzyme immobilization[233]
PE	UV-plasma, poly(tetramethyl glycol) through PAA and chemical, phosphorylcholine	reduction in the amount of adhered platelets; the modified membrane surface had a lower adsorption of proteins;[97] the length of the lipophilic spacer is an important parameter for the protein adsorption
PES	chemical, sulfo ammonium zwitter ion (PDMMSA)	adhesion of human PRP; grafted membranes showed good antithrombogenic properties[238]
PAN	copolymerization, α-allyl glucoside	BSA adsorption, and adhesion of human PRP and macrophage cell; membrane modification revealed that the adsorption of BSA, platelet, as well as attachment of macrophage on the membrane surface could be suppressed[235,239]
poly(AN-co-maleic acid)	chemical, PEGs (various kD)	BSA filtration and adhesion of human PRP; antifouling and biocompatibility of modified membrane improved[240]
silicone rubber and PVC	UV irradiation and chemical, O-butyrylchitosan through bridging of 4-azidobenzoic acid	adhesion of human PRP; modified membranes exhibited less platelet adhesion than did unmodified membranes[241]
PU	chemical, DMMSA through PAA copolymer	adhesion of human PRP; both platelet and protein adsorption were significantly reduced on modified membranes[242]
PVDF	plasma, heparin through bridging with PAA	adhesion of human PRP; heparin immobilization on the membrane surface resulted in the inhibition of platelet adhesion on the membranes[243]
PP	UV-plasma, α-allyl glucoside, sugar, and phospholipid analogous	BSA adsorption, and adhesion of human PRP; protein and platelet adhesion was reduced for modified membranes;[244] activity and stability of immobilized enzyme were improved; the modified exhibited an excellent blood compatible surface
PAN and poly(AN-co-HEMA)	chemical, phospholipid moieties	BSA adsorption and adhesion of human PRP; results showed that the biocompatibility of AN-based copolymer membranes could be improved significantly with the introduction of phospholipid moieties[245]
PP	N₂ plasma, α-allyl glucoside	SMBR, flux reduction in wastewater filtration; after continuous operation of 160 h, the flux recovery, the reduction of flux, and the relative flux ratio for the modified membranes were 17.8% higher, 5.1% lower, and 140% higher than those of the original membrane, respectively[246]
poly(AN-co-maleic acid)	chemical, PEGs (various kD)	BSA filtration, adsorption, and adhesion of human PRP; membrane porosity increased as the PEG content of the casting solution increased[247]
poly(AN-co-maleic acid)	chemical, chitosan, and/or gelatin	BSA filtration; dual layer biomimetic membranes were prepared;[248] it was found that after the immobilization of chitosan and/or gelatin, the surface hydrophilicity increased and platelet adhesion decreased significantly
PVC	chemical, PEG through bridging of ethylene diamine and HDI	adhesion of human PRP; the finished product possessed antifouling properties due to the PEG chemistry[249]
poly(AN-co-maleic acid)	chemical, heparin, and/or insulin through bridging of ethylene diamine	adhesion of human PRP; modified membranes exhibited greater surface hydrophilicity as compared to nascent ones;[250] heparin immobilized membrane showed the best hemo-compatibility among all membranes studied
poly(AN-co-HEMA)	chemical, phospholipid moieties	BSA filtration; the surface modification improved flux recovery and also resulted in higher water and protein solution fluxes;[251] biocompatibility is also enhanced as observed by cell adhesion studies
silicone rubber	tetraether phospholipid, caldarchaeol, through aminopropyl trimethoxysilane	adhesion of bacteria, *S. aureus* and *S. epidermidis*; the biomimetic coating and specific funtionalization prevented the biofouling effectively[252]
PEEK and PU	immobilization of arginine−glycine−aspartic acid	adhesion, the immobilized membrane exhibited better performance in terms of protein secretion and biotransformation of human hepatocytes[253]
PVC	chemical, PEGMA through ozonization	MF, irreversible fouling in filtration of PRP solution due to adsorption of human plasma solution of albumin, globulin, and fibrinogen[254]
PAN	chemical, GAMA by UV grafting	BSA UF; adsorption of BSA was dominated with the increase of grafting degree;[255] the glycosylated membrane showed high flux recovery
PEI and PS	chemical, hydrophilic polymer brushes of PEGMA and HEMA via ATRP	adsorption of BSA; hydrophilic PEGMA and HEMA brushes exhibited significant resistance to protein adsorption[256]
PP	chemical, α-allyl glucoside by nitrogen plasma-induced immobilization	BSA filtration; flux recoveries were higher for the modified membranes due to the hydrophilic α-allyl glucoside[257]
	Attachment through Noncovalent Bond	
CA	adsorption, and cross-linking with glutaraldehyde, protease enzyme	UF, treatment of hemoglobin (or BSA) solutions; the modified membrane observed lower BSA proteins adsorption[233]

Table 4 Continued

base polymer	treatment	function of the membrane
PS	blending, PMPC and MPC copolymer (PMB and PMD)	adhesion in human PRP, and adsorption of albumin, γ-globulin, fibrinogen; modified membranes significantly reduced plasma protein adsorption[234]
PDMS	coating, phosphorylcholine-substituted methacrylate	adhesion in various cells and bacteria, and human plasma; the novel biofouling-resistant opto-sensors are generated by surface modification[258]
PE	blending, MPC	adhesion of human PRP; modified membranes significantly reduced proteins adsorption[259]
PU	adsorption, sulfobetaine grafting through HDI spacer	adhesion of human PRP; The platelet adhesion decreased significantly as compared to unmodified membranes after 1 and 3 h of filtration[260]
PU	semi-IPN, 6-O-carboxymethyl chitosan	antibacterial test with *E. coli*, and adhesion of human PRP; modified surface exhibited reduce protein adsorption[261]
PMMA	adsorption, MPC copolymer (PMB)	adhesion of human blood serum protein and rabbit plasma, and BSA adsorption; this is the unique method of a one-step procedure for the preparation of stable electroosmotic flow in the polymeric microchips[262]
PS	coating, PMMA-co-POEM	alginate, BSA, and microbial cell-lysate filtration; higher flux recovery, at least after a five cycle filtration−washing process, was observed[263]
PET	blending, MPC copolymer (PMBL)	adhesion in human hepatocellular liver carcinoma cell; the carbohydrate-immobilized phosphocholine polymer surfaces were suitable for biorecognition as well as preservation of proteins and cells[264]
PES	blending, tertiary amine-modified PES	UF, the modified membranes displayed superior protein-fouling-resistant property due to the antipolyelectrolyte effect; tertiary amine-modified PES behaves like zwitterionic polymer[265]
PES	MPC copolymer composed of *n*-butyl methacrylate and MPC in casting solution	UF, BSA filtration; the flux recovery of the modified MPC blended PES membrane was remarkably increased;[266] the degree of irreversible fouling was decreased from 0.46 to 0.09 as compared to control PES versus modified MPC blended PES membrane
Cellulose ester, nylon, PP, and PVDF	lysozyme, lytic transgrycosylase coated	*Micrococcus lysodeikticus* bacterium suspension; turbidity and absorbance of cell suspension were decreased;[267] coated membranes exhibited biofouling due to the lysing property of enzyme

cell membrane is mainly composed of various types of phospholipids, proteins, and carbohydrates. Proteins in the bloodstream do not adsorb irreversibly onto the surface of the cell, suggesting that their surfaces are biocompatible. On the basis of this concept, the attention of some groups is focused on the development of membranes with biomimetic surface layer.[97,233−236] Especially the layers containing phospholipid analogues, polypeptide backbones, and sugar moieties were found very effective at reducing protein adsorption. This method can be potentially applied for the biofouling reduction in bioseparation, artificial organs, and also water treatment. Other biomimetic materials include chitosan, dextran, gelatin, heparin, and insulin.

Most surface modifications are performed by the formation of a covalent bond by grafting biomimetic polymers onto the membrane surface using chemical reaction, plasma, UV irradiation, etc. (Table 4). Although these methods look promising in the laboratory scale, their industrial applications seem more challenging due to the limitation of areas that can be covered in UV and plasma applications. Ishihara et al.[234] proposed blending of phospholipids polymer into polysulfone to make the host polymer more biocompatible. They have synthesized copolymers of 2-methacryloyloxyethyl phosphorylcholine with various methacrylates.[234,237] The copolymers were blended with polysulfone in the membrane casting solution. It was found that the content of phospholipids at the membrane surface became higher than in the bulk due to the surface migration of the copolymer. It was

also found that human plasma fibrinogen adsorption on the membrane surface was reduced by blending copolymers. This approach is similar to the blending of surface modifying macromolecules (SMMs) discussed later more in detail.

1.5. Surface Thin-Film-Layer

A thin selective layer is deposited as a coating of thin-film-layer (TFL) on top of the membrane surface by interfacial in situ polymerization reaction: for example, if a top surface of membrane came in contact with an aqueous solution of diamine and subsequently came in contact with diacid chloride (or triacid chloride) solution in hexane, and then at the interface forms the polyamide, which is basically a polycondensation reaction of diacid chloride (or triacid chloride) and diamine. Monomers should have two (or more than two) reactive groups for polycondensation reaction. TFC membranes have been successfully prepared from various polymers.

It is well established that thin film composite (TFC) RO membranes are prepared by the in situ polymerization technique applied to porous MF/UF/NF membranes. According to this technique, a porous membrane is immersed in an aqueous diamine solution, and then the top surface of the membrane is brought into contact with diacid chloride (or triacid achloride) solution in hexane. Thus, a thin-film-layer (TFL) is formed on top of the porous substrate as a result of polycondensation. A variety of TFLs can be formed

Table 5. Reduction of Fouling by Surface Thin-Film-Layer

base polymer	treatment	function of the membrane
TFC membrane: PES support, PEG (6 kD), and poly(ethylene imine) in water phase and TDI in hexane phase	coating of cross-linked polyurea or aromatic PA to prepare TFC	treatment of fluid reactants; the modified RO membranes exhibited excellent antifouling properties;[273] the membranes were used both in the plate-and-frame and in the spiral-wound configurations
PS	thin-film protective coating, plasma-treated acetonitrile	fruit juice filtration; the flux rate dropped only slightly for surface-modified membrane, whereas virgin membrane flux rate dropped significantly[274]
PVA	dynamically prepared TFC	treatment of pepsin protein filtration; the modified membranes showed high antifouling properties as compared to unmodified membranes[275]
PP	multilayer polyelectrolyte, PAA and PDADMAC	MF, adsorption in HAS; the modified membrane observed significantly reduced the adsorption of protein[102]
aromatic PA	TiO$_2$ nanoparticles deposition	treatment of *E. coli* filtration; a novel TFC membrane was developed by self-assembled quantum size nanoparticles and polyamide[8,7,184]
Nafion 117, functionalized PA-6, and PP	multilayer polyelectrolyte, PAA, PDADMAC, and branched poly(ethylene imine)	MF and PV, adsorption in HAS protein; the permeate flux improved with the layer of grafting of the modified membrane[120]
CA	dynamically prepared lignin sulfate layers	treatment of model solution containing dyes−surfactants−salts filtration; the dynamic compact polymer layer, which was made by lignin sulfate, acted as a TFC in the filtration process[276]
interpolymer of PE and styrene-*co*-divinyl benzene having sulfonic acid or quaternary ammonium group	coating and curing of urethane acrylate to prepare TFC	IEM, ionic transport properties of various sodium carboxylate electrolytes; the antifouling properties were achieved without sacrificing the electrochemical transport properties[277]
PVDF	poly(ethylene imine) and TDI using IFP	affinity in *E. coli* bacteria; modified membranes were found to have strong bactericide effects toward *E. coli*[128]
ceramic-supported PES	PVA and PA TFC by IFP	Oil−water microemulsion filtration; modified membranes were more hydrophilic, and hence there was less fouling in the oil and water separation[278]
TFC membrane: PET mat support, PVA nanofibrous middle, cross-linking with glutaraldehyde active layers	polyether-*b*-PA block copolymer or cross-linked PVA hydrogel with surface-oxidized MWNTs	soybean oil−nonionic surfactant−water emulsion filtration; the modified membrane exhibited significantly reduced irreversible fouling[167]
TFC membrane: PES support membrane	coated through in situ polymerization of various acrylates	UF/NF/RO, treatment of reactive dye effluents; the modified membrane evaluated for application of brackish water desalination[279]
TFC membrane: PET mat primed with chitosan, PAN nanofibrous	soaked with NaOH solution and coated with chitosan	UF/NF, flux reduction in vegetable oil−surfactant−water emulsion; modified membranes exhibited higher flux rates than did conventional membranes[280]
TFC membrane: PS support, MPD in water and ICIC in the hydrocarbon oil	coating of PA-polyurea	RO, simulated aqueous solutions of silica, humic acid, and CaCO$_3$, and lake water; the modified membrane contributed better resistance to fouling than did control[231]
TFC membrane: PET mat support, PVA nanofibrous middle, cross-linking with glutaraldehyde active layers	PEO-*b*-PA 12 block copolymer coating layer	soybean oil−nonionic surfactant−water emulsion filtration; the modified membrane exhibited a flux rate (>130 L/m^2·h), which is significantly higher than any commercial membrane and also observed antifouling properties[281]
TFC membrane: PS support, MPEG and MPD in water and TMC in hexane phase	coating of aromatic PA with PEG to prepare TFC	treatment of tannic acid aqueous solution and cationic surfactant−water emulsion filtration; an envisioned idea of attachment of PEG layers in TFC membrane using chemical coupling reaction was achieved[282]
TFC membrane: PES support, TMC and silver (Ag) nanoparticle in dichlorofluoroethane phase and MPD in aqueous phase	cross-linked aromatic PA with entrapped Ag nanoparticles	treatment of *Pseudomonas putida* adhesion; the Ag particle played effectively the bactericidal effect to prevent biofouling;[283] the surface roughness increased with addition of Ag particles
TFC membrane: PS support, TMC in hexane phase, and piperazine in aqueous phase, afterward, soaked with aminopropyltrimethoxysilane in hexane, and with titanium isopropoxide in hexane	aromatic PA bonded covalently with SiO$_2$ layer and TiO$_2$ layer of materials	PEG filtration; the TiO$_2$ particle prevented the fouling; the SiO$_2$ layer stayed as intermediate layer between PA and TiO$_2$ layer materials;[284] the TiO$_2$ materials played a crucial role in antifouling as the end groups

Table 5 Continued

base polymer	treatment	function of the membrane
TFC membrane: PS support, TMC and zeolite nanoparticles in hexane phase, and MPD in aqueous phase	aromatic PA entrapped with sodium zeolite, NaA, and silver exchanged zeolite, AgX, nanoparticles	adhesion of live bacteria cell, *Pseudomonas putida*; the nanoparticle prevented the biofouling;[285] the modified membranes increased in the hydrophilicity and increased in the surface roughness
TFC membrane: PS support	branched PEO brush, made by both ATRP and free radical polymerization, coating layer	RO, treatment of sodium lauryl sulfate and dodecane filtration; the PEO brush-modified membrane retained much higher flux than did commercial membranes[286]
TFC membrane	PA-polyurea formation during in situ polymerization	RO, treatment of the lake water, and the simulated aqueous solutions of silica, humic acid, and $CaCO_3$; the fouling resistance depends on the electrical conductivity of the membrane[287]
TFC membrane: poly(phthalazinone ether amide) support, piperazine in aqueous phase and TMC in *n*-hexane phase	coating of cross-linked aromatic-cyclic PA	treatment of acid chrome blue K (ACBK) dye and NaCl salt mixed solution; insignificant decline in flux was observed due to deposition of dye ACBK molecules on coated NF membrane[288]
TFC membrane: PES support, terephthaloyl chloride in benzene phase and 1 wt % MPD with PVA (or PEG 2 kD or chitosan) in aqueous phase	cross-linked with PA and PVA, PEG, and chitosan	static adsorption of β-lactoglobulin; coated hydrophilic membranes demonstrated 30−35% reduction of protein adsorption due to PVA, PEG, and chitosan[289]
TFC membrane: PS support, 2 wt % MPD in water phase and 0.5 wt % TMC in *n*-hexane phase	grafted with 3-monomethylol-5,5-dimethylhydantoin	diluted sodium hypochlorite solution and adsorption of *Escherichia coli*; modified membranes improved both chlorine resistance and antibiofouling properties[290]

from different polymers including polyamide-hydrazide, polyamide-ether, polyamide-urea, polyurea, polybenzimidazole, and similar polymers. Utilizing this concept, formation of thin-film-layer (TFL) on membrane surface is currently one of the projects undertaken to prevent fouling (Table 5).[268−272] The TFLs can also be formed through noncovalent or van der Waals bond by using hydrophilic or negatively charged materials. In general, TFL is applied to the MF/UF membranes so that the surface becomes smoother, hydrophilic, and negatively charged, which is expected to reduce the fouling. Recently, MF/UF/NF membranes were fabricated from surface-modified nanofibrous membranes, and RO membranes were developed with nanoparticles entrapped in the top surface layer of the thin-film-nanocomposite (TFN) membranes. It is expected to prevent fouling in many systems.

2. Surface-Modifying Macromolecules

When two macromolecules are blended, one of the macromolecular components tends to migrate toward the surface to reduce the surface energy. By controlling the amount of the migrating component, the surface can be modified by blending only a small quantity of macromolecules. The process is one step unlike coating, grafting, and other processes that require an extra surface-modifying process. Several works based on this principle are summarized in Table 6.

On the basis of the above concept, surface-modifying macromolecules (SMMs) have been developed by our group. SMMs migrate and reorganize themselves at the membrane surface so that the tail groups are oriented perpendicular to the air surface.[302−305] The SMMs often show micellar formation analogous to that of surfactants. The concept of the SMM-blended membrane is schemati-

cally illustrated in Figure 3. The hypothesis is based on AFM, LSCS, SEM, XPS, etc. The high elemental composition of SMM is present at the active membrane surface, while the concentration of the base material is low. With an increase in distance from the top surface to the depth direction, the SMM concentration decreases as observed by XPS elemental analysis. SMM-rich microdomains are often embedded in the active surface. A gradual depletion of the number of microdomains was observed throughout the bulk matrix from the top membrane surface down to a depth of 12 μm by LSCS. These observations from AFM and SEM further noticed that SMM-blended membranes are often rougher than the base polymeric membrane.

Mayes' group used methyl methacrylate and PEO-based Comb polymer as the macromolecular additive to increase the surface hydrophilicity of poly(vinylidene fluoride) membrane.[173,293,294] SMMs prepared by our group are, on the other hand, based on polyurethane prepolymers.[296,304,305] When both ends of a prepolymer are capped by hydrophilic chains, hydrophilic SMMs are formed. Their blending to the host polymer renders the surface more hydrophilic. On the other hand, when end-capping is made by fluorohydrocarbon chains, the SMMs become hydrophobic, and the blending of these SMMs renders the surface hydrophobic. Interestingly, blending both hydrophilic and hydrophobic SMMs can reduce fouling. The fouling reduction by hydrophobic SMMs is attributed to the accumulation of fluorohydrocarbon at the surface, which acts as a surface lubricant. This shows that an increase in surface hydrophilicity alone does not provide a necessary condition for fouling reduction.

Table 6. Surface-Modifying Macromolecules for Membrane Fouling

base polymer	treatment	function of the membrane
	Increase in Surface Hydrophilicity	
poly(MMA-*co*-glycidyl methacrylate	blending, heptadecafluorodecyl acrylate	treatment of oil−water filtration; the modified membrane exhibited antifouling properties[291]
PP	blending, hydroxylated PP	dextrans, and PEGs filtration; modified membrane offered good selectivity, flux, and antifouling property[292]
PVDF	blending, PVDF-*g*-POEM graft copolymer	BSA adsorption; surface enrichment of hydrophilic comb polymer imparts significant resistance to the adsorption of BSA;[293] the modified membrane displays significant resistance to BSA fouling as compared to pure PVDF
PVDF	blending, comb having methacrylate backbone and PEG end groups	BSA filtration; separation surface porosities for comb-modified membranes are up to an order of magnitude higher than PVDF controls[294]
CA	blending, MPC copolymer (PMB)	albumin, γ-globulin, and fibrinogen protein filtration; the modified membrane showed good permeability and low membrane fouling property[295]
PES	blending, hydrophilic PEG ends capped PU	river water filtration; blended membrane showed better long-term stability on flux, and the TOC removals were higher as compared to the literature value[296]
PS	blending, hydrophilic PS-*g*-PEG graft copolymer	BSA filtration; membranes prepared were hydrophilic but water insoluble;[297] modified membrane delivers enhanced wettability, porosity, and protein resistance
PES	blending, amphiphilic sulfobetaine copolymer (PBMA-*co*-PDMMSA)	BSA filtration; irreversible fouling was considerably reduced, and a flux recovery rate of 82.8% was achieved[298]
PES	blending, neutral SPC	UF-HF BSA proteins filtration; adsorption of BSA proteins decreased for blended membrane[299]
PS	in-situ formation of hydrophilic PEG ends capped PU	RO/NF, seawater desalination; the modified membrane exhibited improved flux stability due to the bulky hydrophilic PEG ends capped PU as compared to the control TFC membrane[300]
PVDF	blending, PMMA-*r*-PEGMA random copolymer	BSA adsorption; adsorption of BSA was decreased with increasing content of PMMA-*r*-PEGMA;[301] the modified HF membrane displayed reversible BSA fouling and improved PWP
	Increase in Surface Hydrophobicity	
PES	blending, hydrophobic fluorotelomer end-capped PU	oil−water emulsion filtration; the gel-layer resistance of the blended membrane decreased with an increase of additive[302]
PES	blending, hydrophobic fluorotelomer end-capped PU	humic acid filtration; high correlation exists for deposited humic acid and reduced flux[4]
PES	blending, hydrophobic fluorotelomer end-capped PU	river water filtration; the modified membrane proved to have a positive effect in terms of membrane performance as compared to the control;[303] the low-permeate flux membrane exhibited higher removals and lower fouling

3. Conclusions

After surveying the latest works on the surface modification aimed at the reduction of membrane fouling, the following conclusions can be drawn.

Regarding the effect of hydrophilicity on the fouling, the surface contact angle is used as a measure of the surface hydrophilicity. The contact angle is, however, affected by the surface morphology such as surface roughness and membrane pore size. Currently, there is no experimental method to measure hydrophilicity as a unique property of the membrane chemistry. The fouling reduction by membranes of very high hydrophobicity suggests that there are chemistry parameters other than hydrophilicity that affect the membrane fouling.

Regarding the effect of surface charge, the experimental method and the surface electrochemistry seem far better established than other parameters such as hydrophilicity and surface roughness.

Emergence of AFM, which allows the measurement of surface roughness in nanoscale, enabled the correlation of the surface roughness and the fouling, which means fouling increases with an increase in surface roughness. The correlation has been established but only for a limited range of RO and NF membranes. It is not known if the correlation can be extrapolated to a wider range of membrane roughness. It is also not known if the same theory is applicable to the fouling resulted from the deposition of small organic molecules and inorganic salts. The mechanism of hydrodynamics on the rough surface of nanoscale is also unknown. It should also be noted that there is a different view regarding the contribution of surface roughness to the membrane performance. Some researchers believe that an increase in surface roughness favorably affects the membrane performance, because enhanced surface roughness may result in an increase in the effective surface area, and consequently an increase in the permeate flux. The effect of membrane surface area on the membrane performance is a controversial research topic even now.

The simultaneous changes in the other parameters, when one of the parameters is changed, make the study of membrane fouling even more difficult.
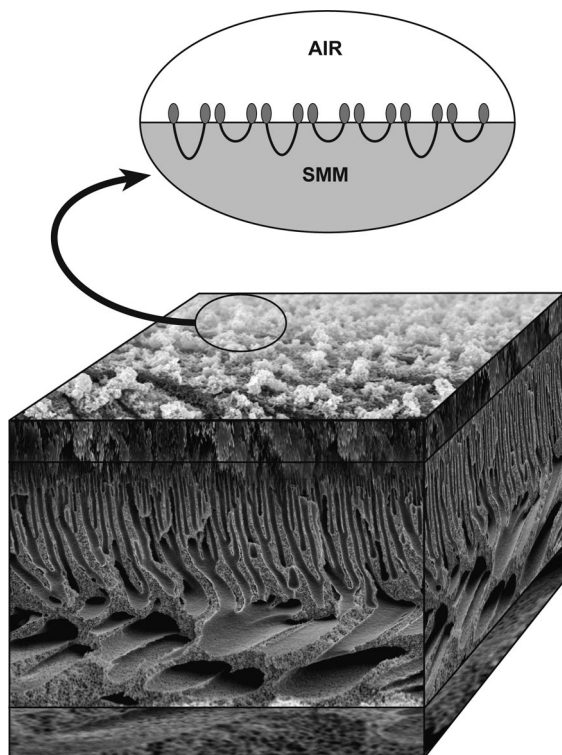
Figure 3. An art diagram illustrating SMM-blended membranes: dumbell-shaped tail contains functional end groups, and the bent line contains polymer chain.

Regarding the contribution of biomimetic surface, immobilization of biomacromolecules plays a significant role to prevent the biofouling. This is one of the latest attempts to reduce the membrane fouling as the references that relate to this approach indicate. Formation of biomimetic surface is considered to be the most effective way to prevent the biofouling. Although its applications are rather limited at the current stage of development, wider applications are expected in the future. Similarly, the formation of thin-film-layer based on the well-established technique to fabricate TFC membranes for RO is a new attempt for fouling reduction. In this approach, the formation of selective thin layer is not necessary. Rather, the top thin film is formed only to increase hydrophilicity of the membrane surface. A thin layer of self-assembled polyelectrolytes may also serve to increase the surface hydrophilicity.

Last, but not least, the most important unanswered question is how much the surface modification can contribute to the fouling reduction. Some researchers have experienced that membrane fouling can indeed be reduced by modifying the membrane surface only when the solution is dilute or only in the initial stage of the separation experiment. Once the deposition of foulants has taken place, the surface modification is no longer effective to prevent fouling. This is understandable because the effect of solute/membrane interaction is severely reduced once a layer of deposited foulants is formed. The surface properties can no longer play a role in further deposition of foulants.

This implies that there are no membranes that are free from fouling under any circumstances. To maximize the effectiveness of the modified surface, incorporation of other devices to prevent the foulant deposition onto the membrane surface, for example, membrane module design, membrane cleaning, etc., is necessary.

4. List of Abbreviations

AA	acrylic acid
AAG	2-acrylamidoglycolic acid
cAMP	adenosine 3′:5″-cyclic monophosphate
AMPSA	2-acrylamido-2-methyl-1-propanesulfonic acid
AN	acrylonitrile
ATRP	atom transfer radical polymerization
BSA	bovine serum albumin
CA	cellulose acetate
CTA	cellulose triacetate
DBS	dodecyl benzene sulfonate
DMAEM	2-(dimethylamino)ethyl methacrylate
DMMSA	N,N'-dimethyl-N-methacryloyloxyethyl-N-(3-sulfo-propyl) ammonium
DTAB	dodecyl trimethyl ammonium bromide
ED	electro-dialysis
GAMA	D-gluconamidoethyl methacrylate
HAS	human albumin serum
HDI	hexamethylene diisocyanate
HEMA	2-hydroxyethyl methacrylate
HF	hollow-fiber
ICIC	5-isocyanato-isophthaloyl chloride
IEM	ion-exchange membrane
IFP	interfacial polymerization
IPN	interpenetrating polymer network
MBAA	N,N-methylene bisacrylamide
MBR	membrane bioreactor
MC	methyl cellulose
MMA	methyl methacrylate
MPC	2-methacryloyloxyethyl phosphorylcholine
MPD	m-phenylene diamine
MPDSAH	[3-(methacryloylamino)propyl]-dimethyl(3-sulfo-propyl) ammonium hydroxide
MPEG	monoamino-monomethoxy PEG
MTMA	myristyltrimethylammonium bromide
MWNT	multiwalled carbon nanotube
NVF	N-vinylformamide
NVP	N-vinyl-2-pyrrolidone
OD	osmotic distillation
PA	polyamide
PAA	poly(AA)
PAN	poly(AN)
PAN-co-PDMMSA-co-PDMAEM	poly(AN-co-DMMSA-co-DMAEM)
PBMA-co-PDMMSA	poly(n-butyl methacrylate-co-DMMSA)
PDADMAC	poly(diallyldimethyl ammonium chloride)
PDMAEM	poly(DMAEM)
PDMMSA	poly(DMMSA)
PDMS	poly(dimethyl siloxane)
PE	polyethylene
PEEK	poly(ether ether ketone)
PEES	poly(ether ether sulfone)
PEG	poly(ethylene glycol)
PEGDA	PEG diacrylate
PEGMA	PEG monomethacrylate
PEI	poly(ether imide)
PEO	poly(ethylene oxide)
PES	poly(ether sulfone)
PET	poly(ethylene terephthalate)
PLA-PMPC	poly(lactide-co-MPC)
PMB	poly(MPC-co-n-butyl methacrylate) 30 mol % MPC
PMBL	poly(MPC-co-n-butyl methacrylate-co-2-lactobion-amidoethyl methacrylate)
PMD	poly(MPC-co-n-dodecyl methacrylate) 30 mol % MPC
PMMA	poly(MMA)
PMPC	poly(MPC)

PMPU	poly(MPC-*co*-2-methacryloyloxyethyl phenylurethane) 30 mol % MPC
PNIPAM-PC-C$_{18}$	poly[*N*-isopropylacrylamide-*N*-(phosphorylcholine)-*N*′-(ethylenedioxy)bis(ethyl)acrylamide-*N*-(*n*-octyldecyl)acryl-amide]
POEM	poly(oxyethylene methacrylate)
PP	polypropylene
PPESK	poly(phthalazinone ether sulfone ketone)
PPO	poly(propylene oxide)
PRP	platelet-rich plasma
PS	polysulfone
PSSS	poly(SSS)
PU	polyurethane
PV	pervaporation
PVA	poly(vinyl alcohol)
PVC	poly(vinyl chloride)
PVDF	poly(vinylidene fluoride)
PVP	poly(NVP)
PWP	pure water permeation
RAFT	reversible addition−fragmentation chain transfer
SDS	sodium dodecyl sulfate
SPC	soybean phosphatidylcholine
SPE	*N,N*-dimethyl-*N*-2-methacryloyloxyethyl-*N*-(3-sulfopropyl) ammonium betaine of molar mass 279 g/mol
SSS	sodium 4-styrene sulfonate
TA-PES	tertiary amine-modified PES
TDI	toluene diisocyanate
TMC	1,3,5-benzenetricarbonyl trichloride
TOC	total organic carbon
TTAB	tetradecyl trimethyl ammonium bromide

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