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Improvements in Electrospun Nanofibrous Membranes and Their Applications in Water Treatments

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ABSTRACT

Nanofibrous structures/buildings offer many interesting features due to their large spacious surface area. This makes them promising through various applications, especially water management. This new generation of large-membered/poous membranes shows great hope that it can be used in a variety of partition systems due to its outstanding features such as surprisingly high porosity (≥ 90%) and connected 3D pore structure, as compared to standard techniques. This article reviews updates of electrospun nanofibrous layers (ENMs) with some highlights of recent achievements, issues, and future ideas for water treatment. To begin with, the basic principles of electrospinning were discussed. The use of ENMs in thin-film composite membranes (TFC) in pressure-driven procedures for water treatment is introduced. The new use of ENMs in the membrane distillation (MD) process is hotly discussed. Deductions, conclusions and opinions are expressed in terms of recent developments.

Keywords: Nanofibers, water treatment, electrospinning, electrospun nanofibrous layers, thin-film composite membranes, membrane distillation

1.0 INTRODUCTION

The water crisis is undoubtedly a global challenge. The problem stems from water pollution, declining water availability, climate change, global population growth and the demand for water-intensive industries [1, 2]. One solution to this challenge is to filter out available or polluted water sources to remove pollutants and make drinking water [2, 3, 4]. Industrial wastewater from the metallurgy, oil and gas industries, mines or chemical industries may be rich in heavy metals, organic and inorganic compounds and chlorine products have become the world's most common pollutants. Therefore, the separation of oil-rich wastewater, especially oil / water mixtures, is a worldwide challenge due to the large amount of wastewater that is produced in many industrial systems and in everyday life. These oil-based pollutants from the industry should be carefully treated before incorporating into any body of water receive due to environmental and health needs [5, 6].

Many oil-splitting techniques have recently been offered that deal great promise, but most come with some restrictions. Conventional methods, such as rotation/flotation, gravity separation and scaling are all effective when using free mixes of oil and water mixtures (oil droplet> 150 mm, with distributed oil sizes varying between 20 and 150 mm); however, they do not work when the size of oil (<20 mm) i.e., oil emulsions is small [5, 6]. Table

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1 below shows the physical classification of these droplets according to their size [7, 8]. Inefficiencies, high operating costs and secondary waste processing also hinder things with common strategies. There is consequently an urgent need for the introduction of advanced strategies that can separate oil and water.

Table 1 Physical classification of oil and grease droplets [7, 8]

Physical classes	Diameter range (μm)	Description
Free oils	Above 150	Droplets that quickly come up to the surface under calm conditions because of the discrepancy created by different densities of the oils and water.
Dispersed oils	20-150	Droplets that is stable due to inter-particle forces such as electric charge.
Emulsified oils	Below 20	Droplets those are stable because of the chemical action of active agents on the surface.
Dissolved oils	<5	Droplets that are either dissolved in oil or evenly distributed.
Oil-wet solids	-	The surface of any solid elements that have oil sticking them.

The membrane filtration/ categorisation technology has recently established itself as a good way to separate oil and oil-based mixtures. Therefore, filtering of membranes is widely accepted in the use of food, fuel cell, insect repellents (desalination) and pharmaceuticals [9, 10]. Compared with conventional technology, membrane separation technology provides greater efficiency in oil removal, resulting in even higher quality of energy costs. These benefits make membrane filtration as a very useful way to separate oil and water mixtures in various industrial valleys/ effluents [11, 12]. Among the various forms of nanofiber production, electrospinning is the most promising and effective in many ways, and new experiences for development and development are being investigated.

There is no doubt that polymer fibers in the nanometric grade get more attention in its use as a filtration media when compared to other applications. This is because of their unique filtering properties for liquids and air. With the flexible reactivation of the function and content of the dope solution, a nanofibrous membrane with a nanofibrous structure can be developed [13, 14]. The small size of the pore and its small distribution, as well as the very high porosity, enable electrospun nanofiber membranes (ENMs) to effectively distinguish water pollution and wastewater treatment [15, 16]. According to statistical figures published by the ISI Web of Science, between 2010 and 2019, there was a significant increase in research articles related to oil / water separation (Figure 1). About ~1100 papers were published between 2010 and 2019, and more than half of these papers were published between 2017 and 2019, reflecting the geometric progression of field value. The general purpose of this review is to highlight recent progress in the development of ENMs with a particular focus on the latest achievements, issues, and perspective ideas in water treatment programs.

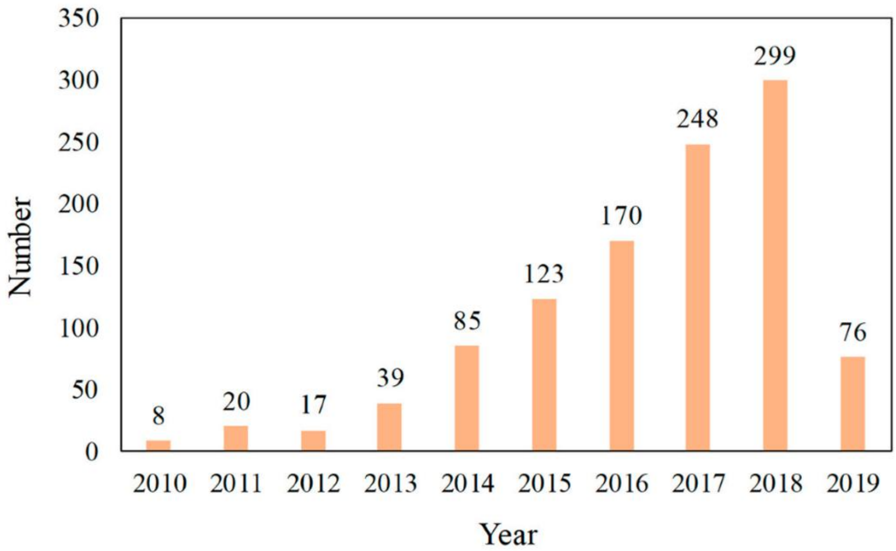


Figure 1 ISI articles published between 2010 and 2019 that refer to oil/water separation

2.0 POLYMERIC NANOFIBERS MEMBRANE PRODUCTION TECHNIQUES

Determining a technique for the fabrication of polymeric nanofibers membranes is dependent on both the selection of the polymer and the preferred structure of the membrane. There are some important processing procedures for preparing polymer nanofibers. They are drawing, electrospinning, isolation of phases, self-assemblage, and templates preparation. Drawing is a technique that is performed similarly to dry spinning in the fiber industries. As a result, extremely longer single nanofiber is formed one at a time. However, Choong [10]; Homaeigohar [17]; Xing, Wang [18] and Nain, Amon [19] assert that there is problem with the processing technique of drawing because only viscoelastic materials, which can endure strong deformations in the pulling process, are produced as nanofibers. In the technique of templates preparation, the membranes that are nanoporous can be used as templates to create solid or hollow nanofibers from many different raw materials. These materials include carbons, electronically conducting polymers, metals, and semiconductors. Even then it is not possible to produce continuous nanofibers one at a time using this method [17, 19, 20].

Phase separation is another method that involves dissolving, gelation and extracting by freezing, drying or using varied solvents. Choong [10]; Shao, Chen [21]; Homaeigohar [17]; Ma and Zhang [22] believe a nanoporous foams formed although it is time consuming to convert the solid polymer. Finally, self-assembly is a where randomly distributed pre-existing organize themselves into a pattern. The organization is caused by particular, local interfaces among the constituents themselves. This technique too is time consuming like phase separation [23, 24]. Therefore, Huang, Zhang [38] conclude that electrospinning appears to be the most appropriate process for producing nanofibers continuously from different polymers. The electrospinning process produces ultrafine fiber continuously (diameter of 10 μm to 10 nm) by using an electrical driving force to impel a polymeric melt, or solutions through the spinneret. The principal advantages of this technique lie in its relative ease.

It is easy to setup. It operates at high speed, little costs, and high versatility. This controls fiber diameter, microstructure and organization, apart from allowing a vast material selection [25-27]. That is why electrospun nanofibrous membranes have attracted more attention for water treatment purposes through different membrane separation processes.

2.1 Electrospinning Technique

A basic electrospinning setup consists of three elements: a nozzle, a high-voltage power supply, and a grounded conducting collector [28]. A simple diagram of the electrospinning set up is shown in Figure 2.

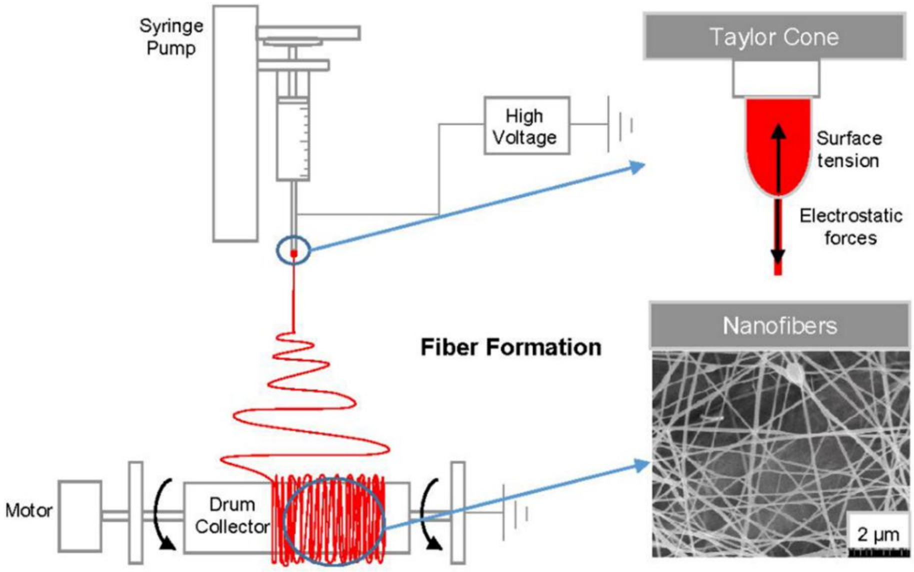


Figure 2 Schematic diagram of electrospinning set-up [28]

There are two principal variations in the organization of the upward electrospinning (UWEs) and the downward electrospinning (DWEs). They are different in amount of beads as well as the fiber orientation. According to Alghoraibi and Alomari [29], upward electrospinning (UWEs), the fiber is collected uniformly as opposed to the random orientation produced by using the downward method. By contrast, fewer beads are formed during the upward process when compared to those formed during the downward set-up [30]. In electrospinning, the downward setup is most favorable for small-scale laboratory use, because it is possible to monitor and produce optimally. Alghoraibi and Alomari [37] and Abdel-Hady *et al*. [30] assert that the upward process, on the other hand, is more suitable for industrial production scales because it is challenging to mass produce nanofibers using a single conventional needle. Figure 3 displays the schematically the conventional vertical and horizontal electrospinning setup.

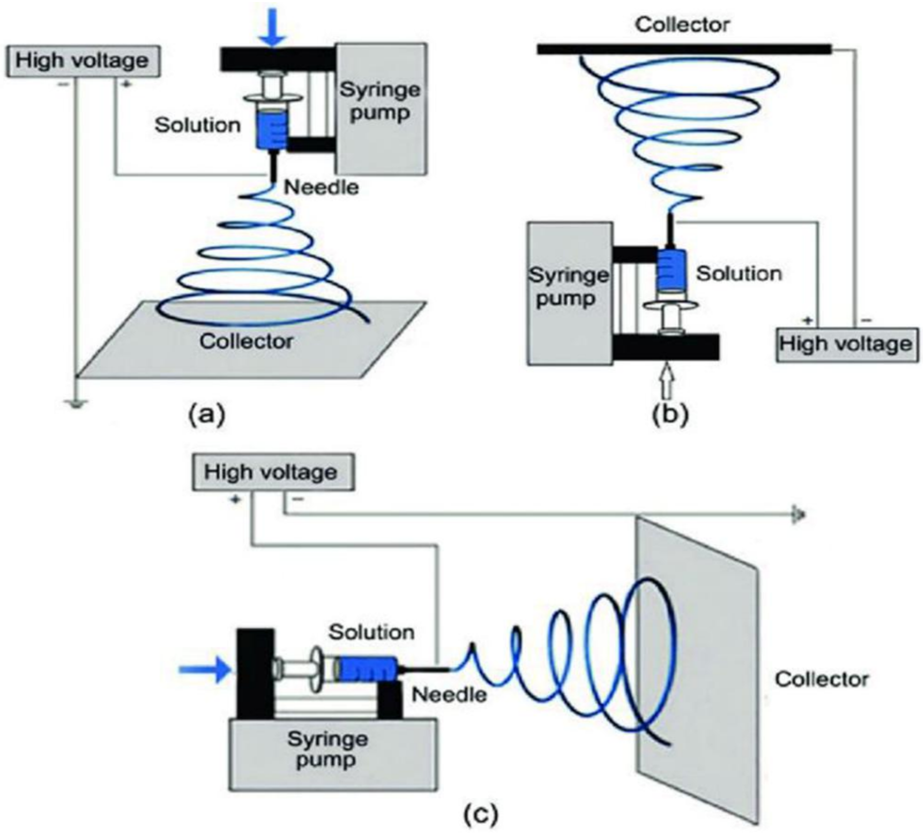


Figure 3 A schematic diagram of the electrospinning set-up from the (a) downward (b) upward and (c) horizontal viewpoint [29, 30, 31]

2.2 Principle of Electrospinning Technique

A high voltage source charges the used polymer solution, creating a jet. The polymer solution is subjected to two forces: a capillary force, due to solution properties, and an electrostatic force, induced by the electric field. The capillary force wants to minimize the surface tension by breaking up of the jet into drops [32]. The electrostatic force is caused by the Coulomb repulsive forces between equally charged ions. In equilibrium, both capillary and electrostatic forces are equal to each other. While increasing the electric field, the electrostatic field strength reaches a critical limit and overcomes capillary forces. In case of a polymer solution with a low viscosity, a drop will disintegrate in smaller drops. By increasing the viscosity, a polymer fiber is established, created by a number of entanglements between polymer chains [33]. The jet is stretched longitudinally, forming a thin fiber with an average cross section at nanometer scale.

2.3 Electrospinning Parameter Investigation

The electrospinning mechanism is influenced by solution and process parameters, and ambient conditions. These parameters presented in Table 2 can determine the morphology of ENMs.

Table 2 Different factors in electrospinning [34]

Parameters	Description
Solution traits	Viscosity
	Polymeric concentrations
	Molecular weights of polymers
	Electrical conductivities
	Surface tensions
	Solvent ratios
Process states	Applied voltages
	Tip to collector distances
	Flow rates
	Needle diameters
Ambient condition	Temperatures
	Humidity
	Atmospheric pressures

The first class of parameters are the solution parameters, with the most important being the polymer concentration, molecular weight and conductivity (Table 2). The concentration is in relation with viscosity, having a major effect on fiber morphology, such as the possible appearance of beads at low viscosities [11]. Thicker fibers may occur at increased viscosity, as there are more entanglements between the different molecules. The conductivity of a polymer solution contributes to the elongation level of the jet. Therefore, solutions with a different conductivity, but electrospun at a similar electric field strength, can induce higher elongations, resulting into thinner fibers [34]. A second class of parameters are process parameters. To induce charges in the solution, a high voltage is applied, creating an electrostatic field [35, 36]. The applied voltage may cause changes in fiber diameter. Thicker fibers may appear when the voltage is increased, because the repulsion force between charges increases as well, resulting into a higher mass flow [11]. However, an increased voltage can also decrease the fiber diameter, when charges in the jet repel each other, causing the jet to elongate and thin. In most cases, the type of polymer determines an increasing or decreasing fiber diameter. The distance between the tip of the nozzle and the collector may influence fiber yields, evaporation rates and instability intervals [11, 36]. The class of ambient parameters, such as temperature, atmosphere pressure and humidity can change the morphology of the fibers. The humidity can influence the fiber diameter, where the fiber diameter increased with decreasing humidity [34].

2.4 Characterization of ENMs

Having an ENMs in the right structure is not enough for the purpose of water treatment. In fact, in-depth knowledge of the polymer used and the additives, morphology, and specifications are complete in water treatment applications [37]. A variety of marking methods can be used directly to test the effectiveness of the membrane in water treatment. The results of this step can be used directly to adjust the electrospinning parameters to improve the new membrane with improved performance. Separation methods can be divided into two main groups that include multiple strategies [38, 39]. These methods have been widely used to measure the characteristics of nanofibrous layers such as pore size and distribution, surface size, nanofiber width, surface strength/energy (hydrophobicity or hydrophilicity), elemental structure, chemical composition, and -membrane fouling potential [40-43]. Among all the features of ENM, the most critical for water treatment purposes are pore size, surface morphology, and higher strength [43]. The general structure (both morphology and topography) of polyether-sulfone (PES) ENMs is shown in Figure 4. The techniques for differentiation and the results obtained for ENM analysis are discussed in detail in the literature [16].

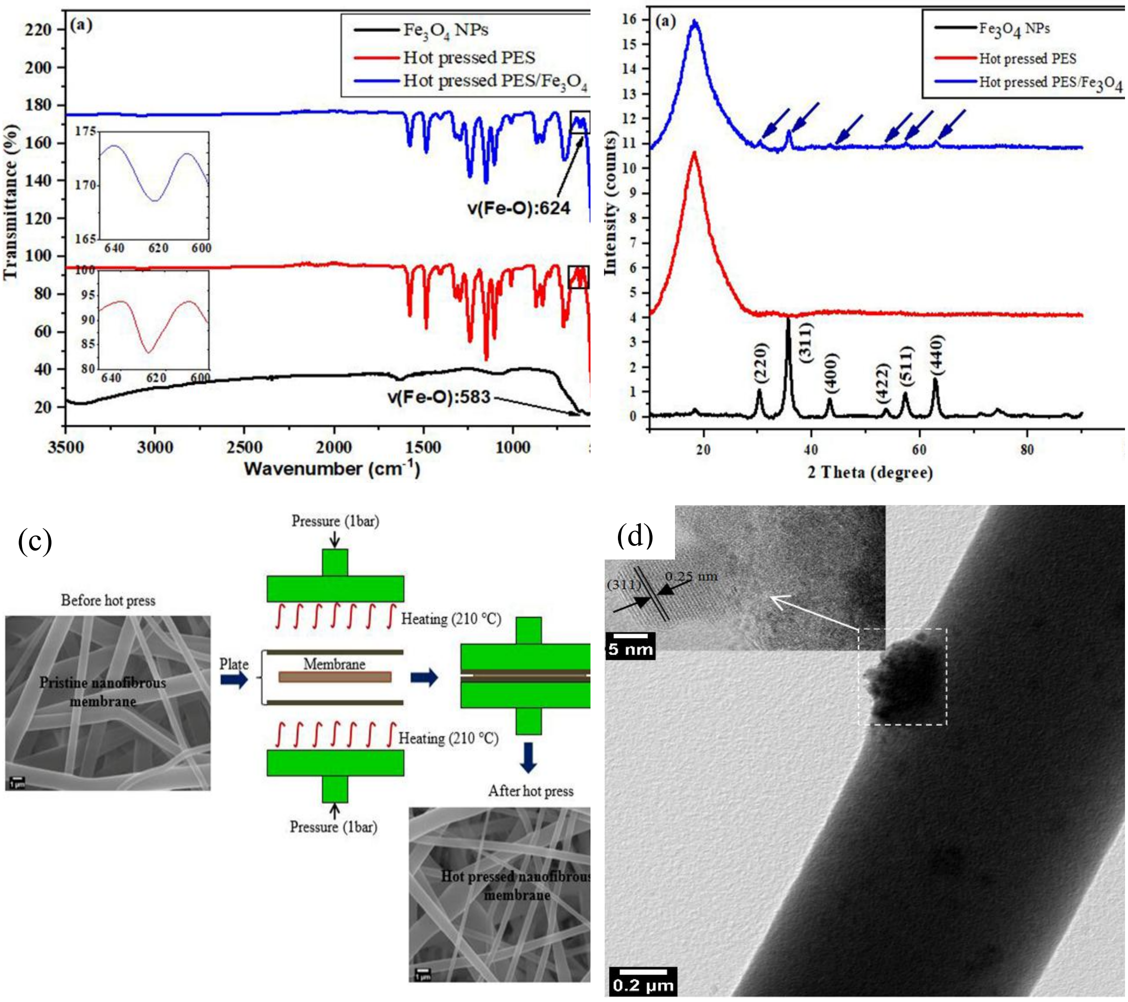


Figure 4 Characterization techniques and obtained results for analysing the PES- ENMs (a) FTIR (b) XRD (c) FE-SEM and (d) TEM analysis [16]

3.0 APPLICATIONS OF ENMS IN WATER TREATMENT

Several methods have been recommended to effectively treat different types of industrial wastewater, including oily wastewater. It is possible to divide these processes into two groups, based on their pore size. The first group contains isothermal membrane procedures, like the membranes driven by the hydrostatic pressure (for example microfiltration (MF), nanofiltration (NF), ultra-filtration (UF), Reverse osmosis (RO), the electric potential-driven process electrodialysis (ED), and the forward osmosis (FO)). According to Ahmed, Lalia [44]; Choong [10]; Lalia, Kochkodan [45], the second group includes non-isothermal membrane procedures like membrane distillation (MD), thermo-osmosis (TO) which can utilize waste heat and renewable energy sources. Two examples of renewable energy are geothermal and solar. MF and UF are both long established techniques for treating water, and RO is commonly employed in not only desalinating, but also purifying water. The MD is an innovative process that has a great potential in the specific area of desalination of highly saline water although it is in the developing stage.

Membranes, therefore, can be applied in treating water as well as determining the technological and economic efficiency. Improving the membranes opens the possibility of greatly improving the efficiency of today's technology. Selecting the material and pore size of the membranes, however, must obviously rely on the kind of applications. Pressure-driven membrane process, for example, is the driving force for membrane operations in liquid filtration. The membranes used in liquid filtrations, therefore, are most commonly classified based in accordance with the range of their operating pressures, and those depend on the range of pore sizes of the membranes [10, 46, 47], as illustrated in Figure 5. Recently, NF has been actively used for efficient separation of divalent ions, as well as proteins, macromolecules, and even sub-molecular organic groups from water and wastewater streams [48, 49]. However, in order to remove the monovalent ions and produce a safe and drinkable water, RO is the most promising option [50, 51]. Here in this section, the applications of electrospun membranes with nanofibrous structure applied to the pressure-driven (MF, UF, NF, and RO) and thermally-driven (MD) membrane processes for water treatment are investigated.

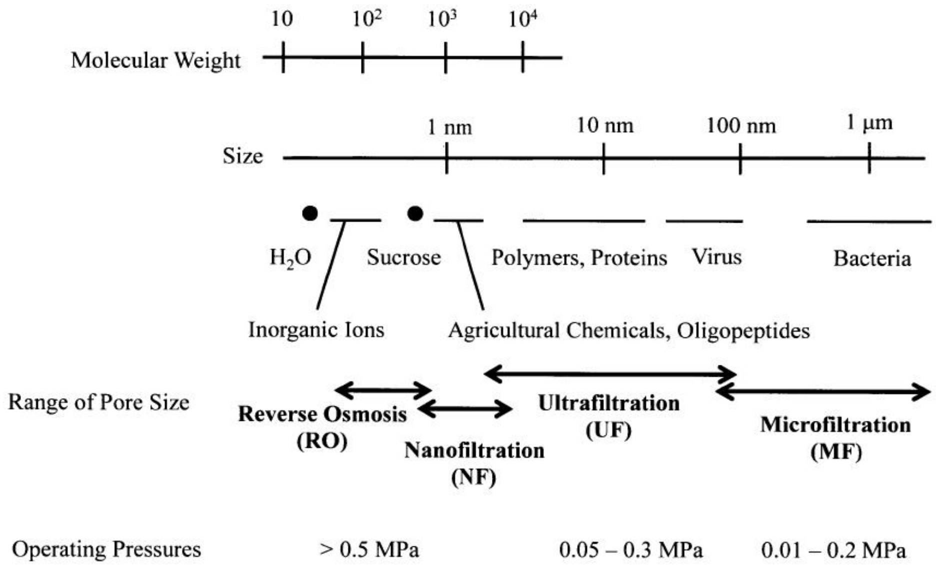


Figure 5 The pore size range for different membrane separation processes [10, 46]

3.1 ENMs for MF Process

MF, which is the most mature membrane system, operates based on sieving filtering theory [107]. Typical pore sizes of MF insect membranes are based on 0.1-1.0 µm. Solid barriers with a larger pore size than 1.0 µm are actually a filter, not a membrane, and are often used as previous filters to remove large particles. High porosity and pore size distribution makes MF-ENMs another promising alternative to standard MF. The first attempt to use MF-ENMs to filter water was reported by Gopal and his colleagues [52]. In this work, the authors used polyvinylidene fluoride (PVDF) to form a nanofibrous membrane to remove polystyrene particles with a diameter of 1, 5, and 10 µm. The results showed that the repaired membrane could effectively remove more than 90% of the tiny-micro particles. Based on the results of the classification, the synthetic membrane showed the same characteristics as those of the MF trading layer. This work has opened a new window examining the use of ENMs in water treatment applications.

In another work, Barhate and his colleagues studied the effect of electrospinning conditions, such as high voltage applied, tip-to-collector distance, and collector rotation speed in morphology and penetration of the nanofibrous PAN membrane [53]. The results of this work showed that electrospinning parameters significantly affect the structure of the fiber while collecting nanofibers. In addition, coordinating drawing and collecting levels can control the distribution of pore size. To better understand the effect of nanofibrous structure on ENM separation operations, various polymers used to design membrane samples, such as polyether sulfones (PES), polycarbonate (PC), polyacrylonitrile (PAN), nylon 6, polyethylene terephthalate (PET), and polysulfone (PSU) [54, 55, 56, 57, 58]. In these activities, the performance of ENMs with various characteristics was studied, including the nanofiber width and membrane size. In almost all of the aforementioned activities, it was concluded that the natural structure and structure of the investigated ENMs significantly affects the performance of the filter. Commercial polymers such as PSU and PVDF become more hydrophobic when electrospun becomes a nanofibrous membrane compared to the material of a virgin polymer. However, hydrophilic membranes are very helpful in direct filtering of water. Therefore, low tendency for overcrowding and deceptive tendencies are expected. Table 3 lists the recently published activities for MF-ENMs.

Table 3 Recently published works on MF-ENMs and their performances [a*]

Year and reference	Materials and ENMs methods	Performances
2020 [59]	Materials PES (Mw = 53 kDa), SPES, PS-NPs (2.5 w/v%), MB (≥ 70.0%), Pb (II) (99%), PAR, and DMF. Electrospinning methods • Dope: 6.0 wt% to 12 wt%	•Feed: Wastewater treatment • Flux: 140- 320 LMH • Rejection: 96.2- 100%

Year and reference	Materials and ENMs methods	Performances
	• Flow: 16 μL/min • Voltage: 20.0 kV • Tip-to-collector: 10 cm • Temperature 14°C • Humidity: 30%	
2019 [60]	Materials PSU (Mw=79,000 g/mol), DMF, THF, HA, NaOH, HCl, IPA, and FHC. Electrospinning methods • Dope: 20 wt.% • Flow: 2.5 mL/h • Voltage: 16.0 kV • Tip-to-collector: 10 cm •Temperature 20- 25°C • Humidity: 38- 41%	• Feed: Wastewater treatment • Flux: 147-133 Kg/m^{2}h • Rejection: 81.9-99.9%
2018 [61]	Materials PAN (Mw = 150,000 g mol), and MDF. Electrospinning methods • Dope: 7, 10, and 12 wt.% • Needle ID: 0.4 mm • Voltage: 18 kV • Flow: 0.5 mL/h	• Feed: Suspended particles • Flux: 712-6810 LMH • Rejection: 11.5–99.3%
2017 [62]	Materials PAN (Mw: 150,000 g/mol), and DMF. Electrospinning methods • Dope: 11 wt.% • Voltage: 15 kV • Tip-to-collector: 15 cm • Flow: 1 mL/h • Humidity: 35% • Temperature: 40°C	• Feed: Oily wastewater • Flux: 6898–18,614 LMH • Rejection: 42.8–98.1%

[a*]Abbreviations: PES: Polyethersulfone, SPES: Sulfonated-polyethersulfone, PS-NPs: Polystyrene nanoparticles, MB: Methylene-blue, Pb (II) :Lead(II) nitrate, PAR: 4-(2-pyridylazo) resorcinol, DMF: N, N-dimethylformamide, PSU : Polysulfone, THF: Tetrahydrofuran, HA: Humic acid ,NaOH : Sodium hydroxide, HCl: Hydrochloric acid , IPA: Isopropyl alcohol, FHC: Fluorinated hydrocarbon, PAN: Polyacrylonitrile

3.2 ENMs for UF Process

In the process of UF, water molecules or small solute particles can pass through the membrane under the hydrostatic pressure while large molecular particles are rejected. The pore size of UF membrane is in the range of 0.01–0.1 μm [63]. UF can trap suspended particles, colloids and microbial viruses, and it is often used to purify and concentrate water in many water treatment fields such as drinking water treatment, desalination, and water reuse [64]. In practical applications, UF membranes prepared by phase inversion method are controversial due to the shortcomings of low throughput and susceptible to pollution. During the infiltration process, the closed large pores in UF membrane prepared by phase inversion method are not conducive to the diffusion of water molecules, which highlights the necessity of a porous support layer and a thin selective layer for the preparation of UF-ENMs [65]. Because the pore size of ENMs is difficult to meet the requirements of UF, it is common to perform various modifications on the surface of ENMs when preparing nanofiber UF membranes. The feasibility of several UF- ENMs modification technologies has been confirmed. One way is to incorporate nanoparticles into the polymer solution before preparing the membrane [66]. For example, a new type of composite hollow fiber membrane produced by mixing alumina nanoparticles into PAN has been developed [67]. The SEM image shows that after adding alumina, the shape of hollow fiber changes from finger-like pores to a teardrop-like structure. This is because alumina is inherently hydrophilic and it will attract more water to the polymer matrix to make the hole larger. Moreover, the presence of charged nanoparticles improves properties such as hydrophilicity, pore size, mechanical strength, and selective nitrate adsorption capacity. This also illustrates the potential application of ENMs with charged nanoparticles in wastewater treatment through electrostatic interactions.

In other studies, TiO_2, Ag NPs, CNTs and other substances have been shown to improve the performance of electrospun UF membranes [68,69]. However, the cost of nanoparticles, uneven distribution on the membrane, and secondary contamination limit the applicability of these methods [70]. Researchers have applied surface modification such as coatings to build new denser modified layers. Bahmani *et al*. [71] successfully prepared a TFC-UF membrane by applying a PAN coating on the PAN nanofiber membrane. Compared with the UF membrane prepared by phase inversion method, the flux of TFC membrane is N1.7 times, and the rejection for arsenic ions is 1.1–1.3 times higher than that of the UF membrane, which is attributed to the porous support layer structure. It has also been found that the hydrophilic modification improves the wettability of the UF membrane, thereby enhancing water permeability and minimizing fouling. Due to its good mutual solubility and hydrophilicity, the nylon-6,6/CS blend was anchored on the hydrophobic PVDF nanofiber support layer to prepare a hydrophilic composite UF membrane [72]. Subsequently, the results show that the modified membrane exhibited high hydrophilicity, and the rejection of bovine serum albumin (BSA) was improved obviously. Meanwhile, the attachment of proteinaceous pollutants could be effectively reversed and its antifouling performance was 2.1 times higher than that of pristine PVDF membrane. In summary, a desirable nanofiber UF membranes for wastewater treatment should has a porous substrate with excellent mechanical properties and chemical stability, and combine with the modification of hydrophilic materials to further optimize the structure or top coating to enhance hydrophilicity, permeability and antifouling performance. Table 4 lists the recently published works on the UF-ENMs.

Table 4 Recently published works on UF-ENMs and their performances [a*]

Year and reference	Materials and ENMs methods	Performances
2019 [73]	Materials PES (Mw: 58,000 g/mol), DMF (> 99%) and NMP (> 99%), PVP (MW: ~ 29,000 g/mol), and HMO NPs (average size: 50–75 nm). Electrospinning methods • Dope: 26 wt.%. • Voltage: 26.2 kV. • Tip-to-collector: 12.8–14.0 cm. • Flow: 0.8 mL/h for 3 h followed by 0.6 mL/h for 3 h and 0.5 mL/h for 1 h. • Humidity: 55%. • Temperature: 25°C.	• Feed: Synthetic oily solution (5000 and 10000 ppm oil). • Flux: 4384 - 7023 L/m^2 h at 1 bar. • Rejection: 90.03–97.98% (5000 ppm oil). 75- 94.04% (10000 ppm oil).
2019 [16]	Materials PES (Mw: 58,000 g/mol), DMF, NMP, PVP (MW: ~ 29,000 g/mol), and Fe_3O_4 NPs (average size: 4.5 nm). Electrospinning methods • Dope: 26 wt.%. • Voltage: 26.2 kV. • Tip-to-collector: 12.8–14.6 cm. • Flow: 0.8, 0.5 and 0.3 mL/h. • Humidity: 55%. • Temperature: 25°C.	• Feed: Synthetic oily solution (12000 ppm oil). • Flux: 2846- 3227 L/m^2 h at 1 bar. • Rejection: 87.16- 94.01%.
2019 [74]	Materials Formic acid (98–100%), Glacial	• Feed: Produced water filtration via solvent vapour

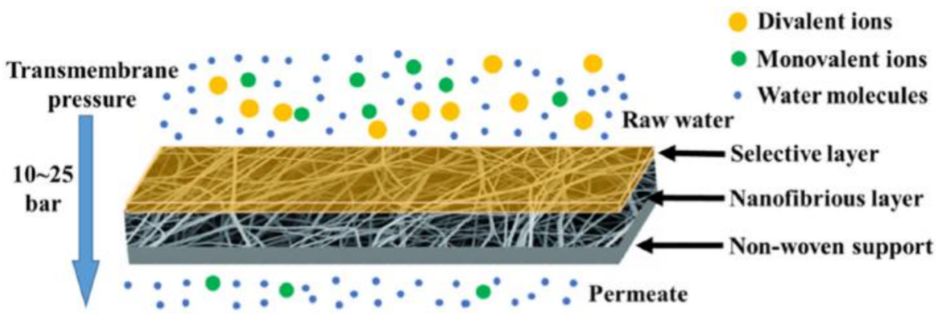
Year and reference	Materials and ENMs methods	Performances
	acetic acid (99.85%) and_Nylon 6,6 pellets. Electrospinning methods • Dope: 14 wt.%. • Voltage: 26.0 kV. • Tip-to-collector: 15 cm. • Flow: 0.4 mL/h.	treatment. • Flux: 533- 733 L/m^2 h at 1 bar. • Rejection: 94.40- 100%.

[a*]Abbreviations: PES: Polyethersulfone, DMF: N, N-dimethylformamide, NMP: N-methyl-2 pyrrolidone, PVP: Polyvinylpyrrolidone, HMO-NPs: Hydrous manganese dioxide nanoparticles, Fe_3O_4 NPs: Iron oxide nanoparticles

3.3 ENMs for NF Process

As a pressure-sensitive membrane separation technology, NF which is widely used in water treatment, can capture particles from 100 to 1000 Da [75]. It has been reported to have high cooler emissions, trace organic pollutants and ions [76-78]. In the present study, most NF membranes are TFC membranes formed by the IP process in a painful porous support layer. As mentioned above, as with all TFC membranes, the porosity and thickness of the support layer affect the availability of NF layers. ENMs have been identified as the preferred candidate as the TFC-NF membrane support layer. The TFC membrane scheme is shown in Figure 6 (a). Compared with the traditional UF membrane, the TFC NF membrane with ENM as a support layer has a higher porosity, connected voids and a nanocomposite structure of the selected layer, leading to higher elevation (flux) and disposal (rejection) [79]. To compare the effects of the various layers of support on the function of the NF membrane, an IP process was used on the surface of the transformation film and ENMs to prepare for the NF membrane [80]. It can be found from SEM that the same selective and thin layer of polyamide on nanofiber is formed by IP reaction (Figure 6 (b)). The results showed that the nanofiber NF membrane had a much higher dissolution of water and salt than the NF membrane of the penetration support phase i.e., much higher water flux and salt rejection than the NF membrane.

(a)



(b)

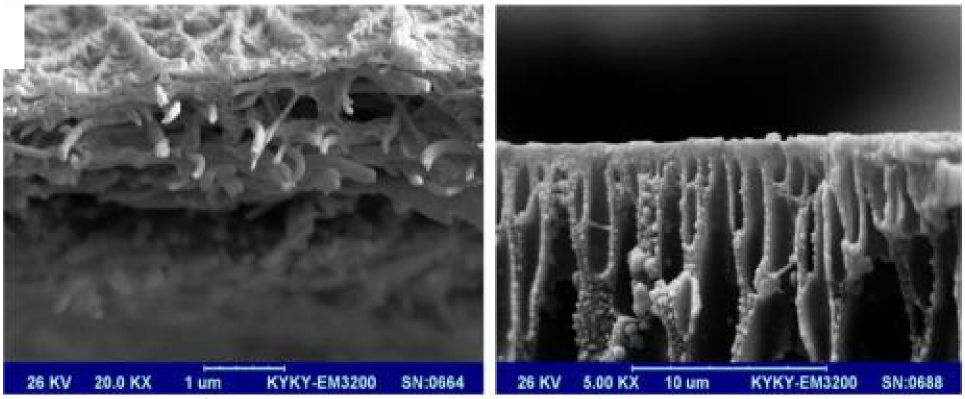


Figure 6 (a) TNF-ENMs system and (b) SEM cross sectional views of TFC-NF (left) and TFC-NF ENMs (right) [80]

In addition, the NF membrane also had a high rejection concentration of divalent ions (such as SO_4^{2-} and Mg^{2+}). The excellent NF efficacy of wastewater management has been demonstrated in a large open pore structure, low hydraulic resistance of nanofiber, and a small nanocomposite layer of the NF-ENMs process. It is expected that more efforts will be made in transforming NF-ENMs into water treatment applications. Covering suitable polymer material in ENMs is a common method of modification. Considering the characteristics of a very high density, the coating or coating/ self-assembly of vinyl monomers [81, 82], TiO_2 [83], polyelectrolytes [84] and other surface-free surfaces to improve the blank/hollow NF membrane to remove dye and the desalination is currently a hot topic for research. The properties of the NF membrane of nanofiber including hydrophilicity, roughness, micro-porosity, durability and mechanical strength are considered to be the most important factors affecting NF performance. Although some problems have been alleviated by the use of ENMs and the selected IP layer, the pressure-driven NF process causes damage to the NF membrane with a nanofiber support layer. Therefore, how to improve the stability of the NF layers is an urgent problem that needs to be addressed.

3.4 ENMs for RO Process

Similar to NF, RO is also a pressure-driven membrane technology. The pore-size distribution of the RO membrane ranges from 0.1–1 nm [85]. Unlike other membrane technologies, the RO membrane is resistant to the smallest contaminants/ pollutants (Cl^- and Na^+), making it the most widely used water treatment and desalination technology in the world [86]. In addition, for effective salt processing applications, the potential of RO ENMs at high operational pressures must be addressed. Wang *et al*. [87] developed the TFC RO membrane, which exhibits robust RO activity, by preparing PAN ENMs, and then a layer of cellulose nanofibers (CNs) from biomass is incorporated into the lower PAN to form the PAN CNs substrate. SEM shortcuts for PA / PAN and PA / CNs RO membranes are shown in Figure 7 (a) and (b). The PA layer on the PA / PAN membrane is very hard, and the size distribution is uneven, while the surface of the PA / CNs layer is smooth, and maintains a high level of water and salt disposal under high operating pressures. This also suggests that a thin layer of PA can be formed in a smooth surface, leading to stable RO performance [88]. In the production of RO-ENM films, the effect of nanofiber substrates on the construction of barrier layers may be more important than expected. The development of slow-moving ENMs with large grip sizes and a hydrophilic surface can be a great research study to prepare for high RO-ENM performance.

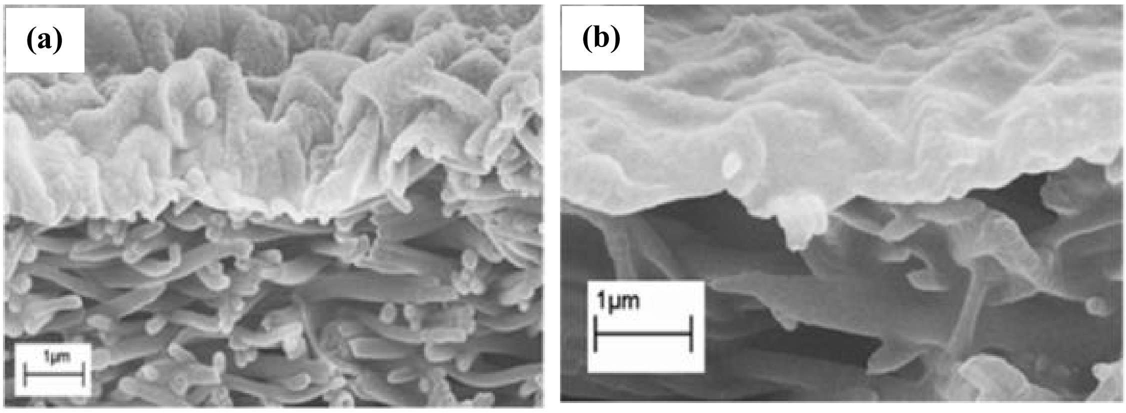


Figure 7 SEM cross-sectional images of RO membranes based on (a) PA/PAN and (b) PA/CNs [87]

3.5 ENMS for MD Process

The previously discussed pressure-driven membrane processes are all isothermal separation techniques. Recently, a new non-isothermal membrane separation technique has been introduced, which is a combination of the conventional distillation and the membrane separation. This separation technique, which uses the vapour-pressure difference as the driving force, has been called "membrane-distillation" process [89]. MD is an impressive separation technique wherein a porous hydrophobic membrane is used to separate the feed channel (hot side) and the permeate channel (cold side) [90] as presented in Figure 8 (a). It is based on the phenomenon that pure water can be extracted from aqueous solutions by evaporation, with the vapour passing through a hydrophobic microporous membrane when a temperature difference is established across the membrane.MD process has four major configurations. All these configurations are the same for the feed channel, where the hot stream as the process liquid is in direct contact with the hydrophobic surface of the applied microporous membrane. The surface hydrophobicity of the used membrane prevents the process liquid from penetrating into its pores. This causes to form the liquid-vapor interface at the entrance of the pores on the membrane surface [91]. The four main MD configurations include DCMD (direct contact membrane distillation); SGMD (sweeping gas membrane distillation); AGMD (air-gap membrane distillation); and VMD (vacuum membrane distillation). Figure 8 (b) shows the general scheme and defines the differences among the MD configurations.

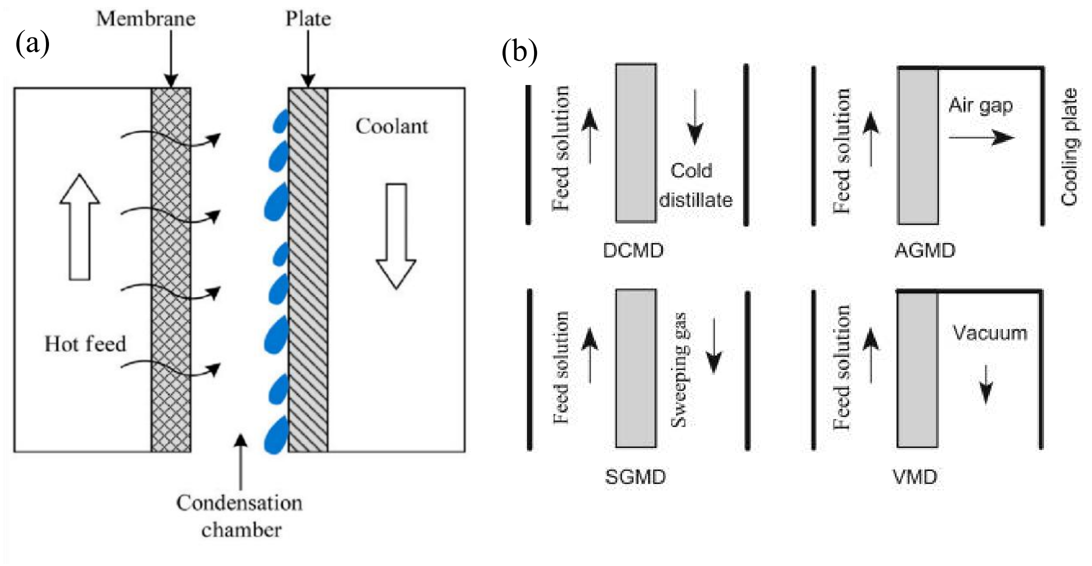


Figure 8 Schematic diagram of (a) principle of the MD process and (b) MD configurations [92, 93]

There are a few crucial characteristics required for an MD membrane that significantly affect the performance, as well as the overall efficiency of the MD process. The applied membrane should have high LEP (liquid entry pressure) value, must be hydrophobic (or even superhydrophobic), must be as porous as possible with narrow pore-size distribution, and must have low tortuosity factor [94]. The MD membrane should also have proper chemical and mechanical properties. Nonetheless, the first generation of the MD membranes has been commercially available MF membranes, which have been made of hydrophobic polymers [95]. However, commercial MF membranes are still being used in various MD processes [96]. The second generation of the MD membranes is fabricated using commercially available polymers [97]. Recently, the ENMs have attracted a lot of attention as the third generation of the MD membranes. This is attributed to their promising and exclusive characteristics including high porosity, three-dimensional interconnected pore structure, and reproducibility [96]. Mechanical durability is one of the most challenging bottlenecks of the ENMs. Li and co-workers [98] studied on improving the mechanical features of the ENMs with the use of nonwoven fabrics and spacer fabrics as the backing layer. The ENMs samples were fabricated using the PVDF polymer. The authors investigated the effect of the support layer on the membrane characteristics (e.g., permeability, porosity, morphology, pore size and pore size distribution, hydrophobicity, and mechanical durability).

Based on the obtained results, a 3D bead-fiber interconnected open structure and a rough membrane surface were observed for the newly developed membrane. The membrane samples were all hydrophobic with surface contact angles above 140°. Moreover, the stress at break and the elastic modulus of the new membrane samples increased by 4.5–16 times and 17.5–37 times, respectively, as compared with the nanofibrous membrane made of pure PVDF. Depending on the results obtained, space fabrics are used as the support layer provided higher water flow compared to the unsupported membrane. The authors conclude that this may be due to the resistance to the mass transfer of space fabrics. The highest water flow in this study was 49.3 kg/m^2/h when the temperature of the hot stream was set at 80 ° C. In addition, the new composite membranes showed moderate to long-term desalination/abortion. Only a handful of research groups have used ENMs with MD procedures until 2014. This study was thoroughly reviewed by Tijing and colleagues [99]. In another review paper, Shirazi and colleagues [100] also reviewed published literature covering the design and implementation of ENMs from 2014 to 2017. Since then, continuous practice has been observed for the preparation and application of nanofibrous layers by electrospinning for water treatment using MD procedures. Table 5 summarizes some examples of recently published activities for the use of ENMs in MD procedures [101].

Table 5 Recently published works on MD-ENMs application and their performances [a*]

Year	Configuration	Materials and ENMs methods	MD key features	Performance
2018 [102]	AGMD	Materials Polymer: PVDF (Mw: 275 kg/mol), Solvents: DMF and Acetone. Electrospinning methods • Dope: 15 wt.%. • Needle: 18 G. • Dope injection: 0.2 ml/h. • Tip-to-collector: 150 mm.	• Pore size: 0.4- 0.5 μm. • CA: 127–153°. • LEP: 15–25 psi. • Porosity: 83–90%.	• Flux: 19.3– 22.5 LMH. •Rejection: >90%.
2018 [103]	DCMD	Materials Polymer: PVDF (Mw: 275 kg/mol), Solvents: DMAc and acetone. Electrospinning methods • Dope: 25 wt.% • Voltage: 27 kV • Needle ID/OD: 0.6/0.9 mm • Dope injection:1.23 mL/h • Tip-to-collector: 27.5 cm • Temperature: 23°C • Humidity: 36%	• Pore size: 509– 945 nm • LEP: 7.9– 17.4 kPa • Porosity: 77– 92%	• Flux: 35- 50 kg/m^2 h • Rejection: >99%
2018 [104]	DCMD	Materials Polymer: SBS (C540 Galprene), and Solvent: DMF-THF (75/25) Electrospinning methods • Needle: 22 G • Voltage: 1.0 kV/cm • Temperature: 21°C • Humidity: 43%	• Pore size:0.58 μm • Contact angle:132° • Porosity: 81%	• Flux: 11.2 L/m^2 h • Rejection: < 99%

[a*]Abbreviations: PDF: Polyvinylidene difluoride, DMF: N,N-dimethylformamide, DMAc: Dimethylacetamide, SBS: Poly(styrene-butadiene-styrene), THF: Tetrahydrofuran

To apply to the MD procedures, ENMs must be able to maintain the distinction between process liquid and full product. Therefore, hydrophobic membrane or superhydrophobic membrane will work as considered the most effective option for water treatment applications [104]. Among the various polymers, PVDF is the most investigated option for making ENMs. This is due to the efficient process capacity of this polymer, especially for the membrane. Both beaded and non-beaded nanofibers have been investigated for MD-based water fixation using ENMs. Surprisingly, beaded nanofibers exhibit a lower amount of surface energy (i.e., higher hydrophobicity), while the full flow (permeate flux) from smooth nanofibers was much better than that of beaded nanofibers [105, 106]. More studies are needed on new polymers to make superhydrophobic ENMs, such as thermoplastics and elastomeric polymers with better performance.

4.0 CONCLUSION AND FUTURE OUTLOOKS

Over the years, ENM established itself as a globally approved filter media for a variety of applications. The unique specifications of ENMs, including high porosity (up to 90%), triple-pore structure, and performance, make them very promising and attractive to both academic researchers and industrial applications. However, before any material can be used for industrial level applications, the following challenges must be addressed.

i. Weak mechanical properties (machine structures) are always a drawback to industrial use. As a result, the area is well suited for further research, and this will increase the mechanical properties of sustainable development nanofibers.
ii. In addition, there are other applications that need to be investigated further for future progress/advancement. Many analyzes can be made on the use of nanofibrous membranes as a scaffold in a thin-film with a composite structure of UF, NF, and FO
iii. In the event of an MD procedure, a superhydrophobic membrane is required. In this regard, ENMs with a wide range of activities with very low potential can be used to advantage
iv. Noble materials/Good equipment is needed for oil refining and industrial wastewater treatment in the future. The use of certain wet materials to separate oil and contaminated water is a new phenomenon. It is a concept that contains many unknown challenges and these should be investigated in great depth. In addition, the use of building materials known to be harmful to the environment may be a key factor in sustainable use.

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