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# Performance of mixed matrix nanofiltration membranes in wastewater treatment: A review

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## Abstract

Water covered close to 70% of the Earth's surface; however, it is a struggle to access 3% of it as freshwater, suitable for human intake. The shortage of water has become a global issue due to climate change, pollution and the demand of growing freshwater. The fabrication of high performance and antifouling nanofiltration membranes are of utmost importance as a result of their capability of treating contaminated water and to also regulate the quality of freshwater. However, there is need for improvement in the fabrication of membrane using different nanofillers to produce mixed matrix membranes (MMMs). Mixed matrix nanofiltration membranes with nanofillers were reviewed for different applications.

**Keyword:** Nanofiltration, mixed matrix membranes, nanoparticles, carbon nanotubes

## 1. Introduction

Water is not equally dispersed after a while and more water is wasted, contaminated and not managed in a sustainable way; which has led to losing thousands of people yearly due to the shortage of water. Hence, shortage of water have pronounced effect on human health, growth of socio-economic and the surroundings [1-4]. Nanofiltration membranes (NF) could be utilized for diverse water and wastewater treatment industries due to their low cost for the removal of ions and organic substances. Present-day investigations in the field of nanofiltration technology is mainly apprehensive about the improvement of the performance of NF membranes in order to minimize fouling and reduce energy requirements of water treatment processes that is already in working operation. Researchers have presented ultra-modern concepts and design of developing new membranes with an aim of obtaining efficient performance they are applied in separation processes [5]. Several procedures utilized in fabricating nanofiltration membranes exist; nonetheless, the option of a pertinent method is contingent on the type of material and the application they will be used for. The properties of membrane may relatively be controlled, via proper selection of the preparation technique and working with the main process conditions.

Researchers have attempted to enhance the performance of NF, especially to upsurge the permeate flux and lessen the resistance of membrane through experiments with diverse membrane materials and configuration. Thin film composite membranes are inter-facially polymerized on top of a microporous substrate and they have been commercially successful with regards to their applications in industries as a result of their capability in the optimization of the selectivity and permeability of individual stratum [6-9]. It has been

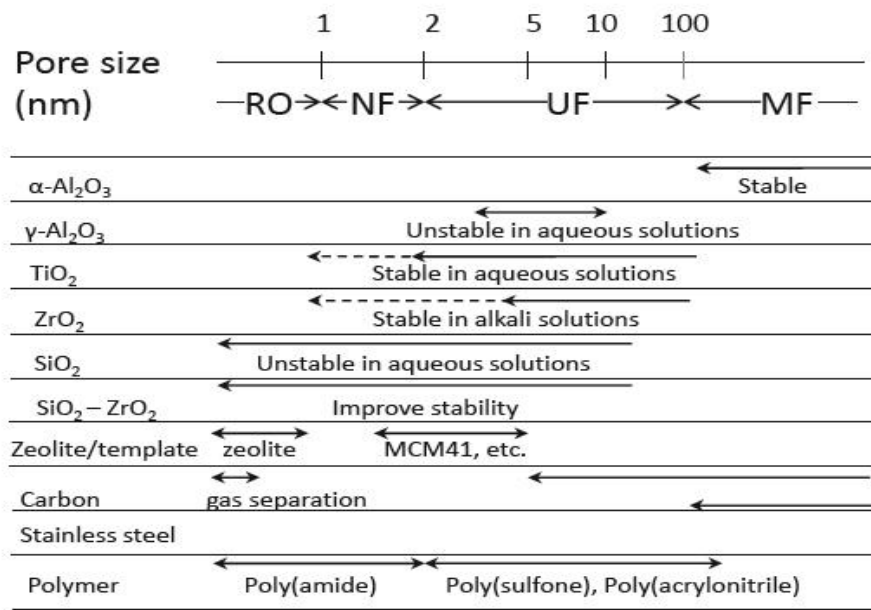


recently revealed that the integration of nanomaterials suchlike electrospun nanofibrous membrane layers to typical thin film composite membranes resulted in an increased permeate flux. The result obtained is due to the essential properties of electrospun nanofibrous membrane that favor the flux; consequently, there was reduction in membrane fouling and the cost of the pre-treatment energy [10]. Electrospun nanofibrous membranes possess exceptional properties like well coordinated pore configuration, high surface area to volume ratio, low trans-membrane pressure, high porosity, excellent mechanical properties and enhance permeability of water. These properties offers a main impact in the process of water treatment. These nanofibers have been used for several water treatment utilizations, and are dependent on thickness, porosity, and surface roughness [11]. Apart from the addition of electrospun nanofibrous membranes to conventional thin film composite membranes, nanofiltration membranes are fabricated with the integration of nanomaterials such nanoparticles, carbon nanotubes, nanoporous ceramics and nano-sponges. This review aim at giving an overview of mixed matrix membranes using nanofillers for wastewater treatment.

## 2. Mixed matrix nanofiltration membranes with nanofillers

Up to this present time, the advancement researchers have attempted on material science, for the improvement of membranes is contingent on nanocomposites. The overabundance of diverse kinds of nanomaterials has practically presented infinite high-tech benefits with regards to the variation and optimizing the current novel membrane for present-day desalination plants [12]. The assimilation of nanofillers such as nanoparticles within polymer matrix has offered an innovative method for fabrication of membranes with boosted permeability, selectivity in an elevated manner and enhanced properties for anti-fouling [13]. Mixed matrix membranes (MMMs) are aimed in taking the benefit of low cost simple method of preparing organic polymeric membranes that possess enhanced mechanical strength with effective working properties of inorganic materials [14]. Application of nanoparticles in the fabrication of membranes provides the capability to make membrane with preferred structure and their functionalities; this gives a practical level of control over membrane fouling [15, 16]. Figure 1 shows the dynamic of preparing functionalized nanofiltration membranes that will result to high performances. Furthermore, the integration of nanofillers in nanofiltration membranes should enhanced the properties of MMMs. Hence, MMMs should have high specific surface area, exceptional mechanical stability and physicochemical properties for the purpose of improving water-permeation of the membrane. Table 1 shows the stability of nano-materials used for synthesizing NF membranes for the treatment of contaminated water. The unique properties of these nanomaterials have resulted to their use in diverse applications, especially in the fabrication of commercial filtration membranes that is cost effective. This may perhaps, greatly cut down the cost of clean water production.

Table 1: Stability of polymers/nanomaterials in aqueous/alkali solutions [17]



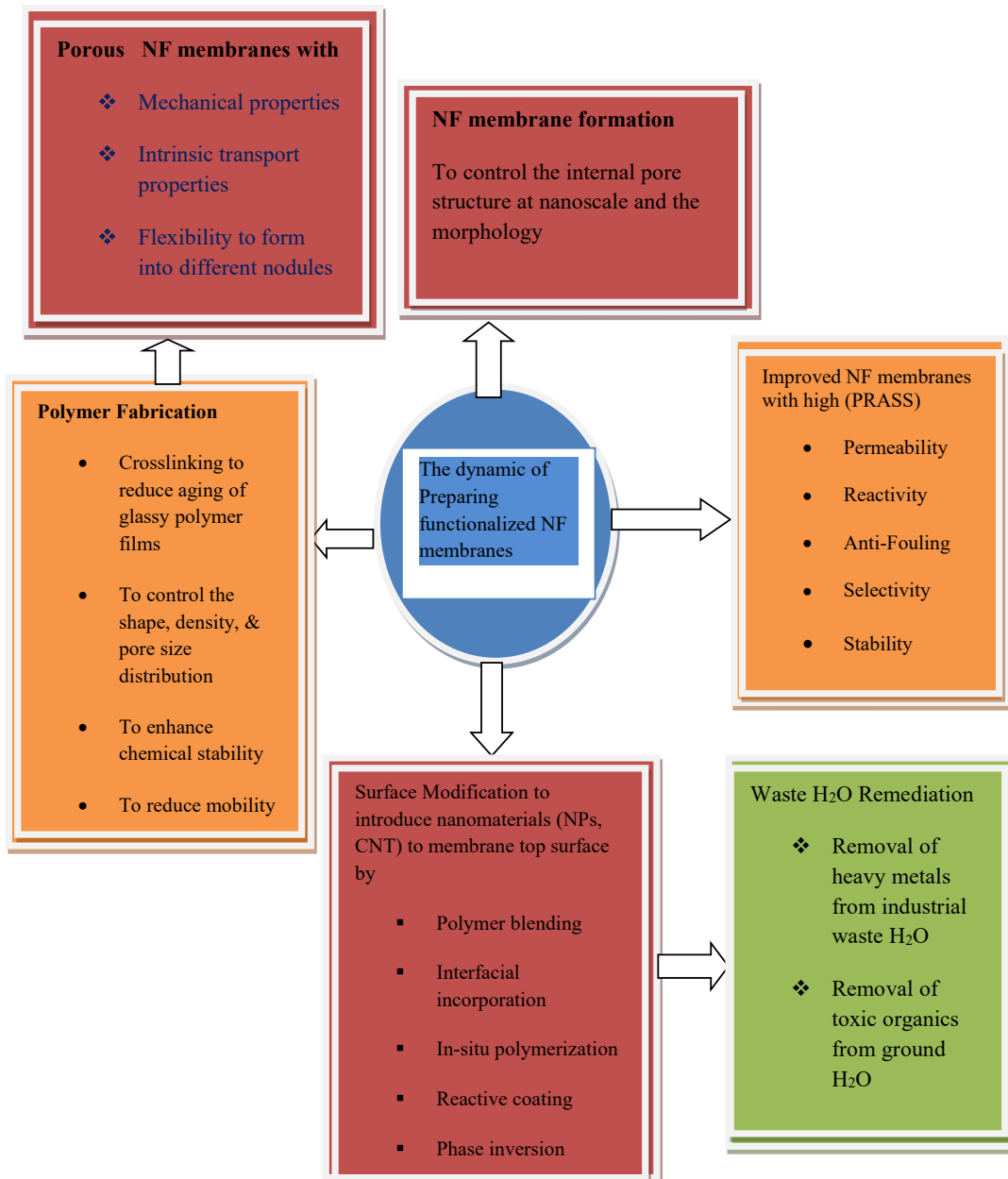


Figure 1: Schematic design of the dynamic of preparing nanofiltration membranes for excellent performance

## 2.1 Titanium dioxide (TiO<sub>2</sub>) Nanofiltration MMM for wastewater treatment

TiO<sub>2</sub> is among one of the nanoparticles that is mostly utilized in synthesizing MMMs. TiO<sub>2</sub> has severally been employed in the fabrication membranes for the purpose of enhancing the surface properties and structure modifications. This is because it has the capability of degrading natural organic matters, creating additional structures that are hydrophilic, and decomposing bacterial cells. [18]. TiO<sub>2</sub> nanoparticle has positively stimulated substantial reaction for the synthesis of nanofiltration membranes with regards to increase in flux and improved fouling resistance. Extensive research on the prospective application of TiO<sub>2</sub> nanoparticles for the reduction of membrane fouling has been conducted [19-21]. This involves the application of a nonporous coating of Titania over membrane surface. This has helped in resisting the internal fouling of the membrane as a result of the non-porosity of the coating. In addition, the super-hydrophilic nature of Titania offers resistance to fouling via the decreasing of adhesion of waste on the surface of the membrane [22]. However, different methods might be employed in the synthesis of mixed matrix nanofiltration membranes integrated with TiO<sub>2</sub>.

Peyravi et al. [23] developed an innovative thin film nanocomposite (TFN) solvent resistant NF membranes via interface layer integration of functionalized TiO<sub>2</sub> over a porous polyimide support; formerly coated by polyethyleneimine. From their study, Fourier transform infrared spectroscopy analysis depicted that the dispersion or stabilization of agents were covalently or physically bonded to the TiO<sub>2</sub>. Furthermore, exceptional stability in organic and aggressive solvents was observed as a result of the limitation of chain mobility found in the presence of rigid TiO<sub>2</sub> domains. Flux was enhanced together with high selectivity of the removal of dyes for TFN-SRNF membrane. Venkatesh et al. [24] investigated the removal of oil from water in emulsion by employing polyvinylidene difluoride mixed matrix nano-filtration membranes incorporated with 1D-PANI/TiO<sub>2</sub> NFs. In their study, 1-D PANI/TiO<sub>2</sub> nanofibers were integrated into polyvinylidene difluoride mixed matrix membranes for the separation of oil and water droplets from oily wastewater. From their study, there was an increase in pure water flux; from 80 to 132 L m<sup>-2</sup> h<sup>-1</sup>. This is an indicator that there was an impact in addition of the 1D PANI/TiO<sub>2</sub> NFs to polyvinylidene difluoride. From the mixed matrix membranes fabricated, PT-4 displayed the highest retention efficiency (99%) of oil at 5 bar. Khorshidi et al. [25] developed a novel methodology for fabricating robust thin film nanocomposite reverse osmosis membrane. This novel method involves the concurrent fabrication and surface functionalization of TiO<sub>2</sub> NPs in an organic solvent (heptane) through biphasic solvothermal reaction. The membranes fabricated were used for the treatment of high-temperature streams made of biological materials and these membranes demonstrated better thermal stability and anti-biofouling properties. Koutahzadeh et al. [26] studied the performance of a hybrid ultraviolet/hydrogen peroxide–mixed matrix membranes system for the removal of an azo dye, known as acid black, from a wastewater purification process. 270 (L/[m<sup>2</sup>·h]) permeation, 29% flux decline, 90% total organic carbon retention, 99% decolorization, and 99% flux recovery ratio were attained using hybridized process of ultraviolet/hydrogen peroxide-polysulfones /TiO<sub>2</sub>/multi walled carbon nanotube-mixed matrix membrane.

## 2.2 Aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) Nanofiltration MMM for wastewater treatment

Aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) is among the stable inorganic nanoparticles utilized in preparing membranes on the account of increasing the performance membranes. This nanoparticle is not expensive, non-toxic, highly abrasive and resistant. Al<sub>2</sub>O<sub>3</sub> nanoparticles was integrated into polyvinylidene difluoride (PVDF) membrane by utilizing dimethyl acetamide as solvent [27]. The influence of the concentration of Al<sub>2</sub>O<sub>3</sub> in the polymer dope on the permeation properties, structures of membrane and performances of membrane for antifouling were studied. The results obtained pointed to the fact that Al<sub>2</sub>O<sub>3</sub>-polyvinylidene difluoride

composite membranes showed weighty changes in surface and inherent properties owing to the addition of nanoparticles [28]. This suggested that a positive influence occur on reducing bio-fouling. Saleh and Gupta [29] synthesized polyamide (PA) nanocomposite membrane with the integration of alumina nanoparticles (which was homogeneously distributed) through in situ interfacial polymerization. It was observed that the membrane performed better than pristine membrane, with improved permeate flux and salt rejection. Bagheripour et al. [30] prepared polyvinylchloride-based nanocomposite NF membranes via the utilization of sodium dodecyl sulfate as additive. In addition,  $\text{Al}_2\text{O}_3$  nanoparticles was used as inorganic filler and dimethylacetamide/tetrahydrofuran as solvents through phase inversion technique. Khalili et al. [31] prepared bi-layered  $\gamma\text{-Al}_2\text{O}_3\text{-TiO}_2$  NF membranes based on  $\alpha$ -alumina supports for the removal of chloride ion. Compression was used in preparation of the supports and sol-gel dip-coating for the top-layer formation. Their results showed that the calcination temperature ( $600^\circ\text{C}$ ) results in different crystal structures including the brookite phase of  $\text{TiO}_2$ , the  $\gamma$  phase of  $\text{Al}_2\text{O}_3$ , and a combined phase of aluminium-titanium oxides. Filtration experiments confirmed the smooth rejection behaviour in an extended pH range because of the dual-layer structure with different isoelectric points (IEPs). Their results showed a higher rejection in the alkaline pH range over the acidic region.

### 2.3 Zinc oxide (ZnO) Nanofiltration MMM for wastewater treatment

ZnO is a semiconductor having an extensive bandgap comparable to  $\text{TiO}_2$  [32]. It is highly reactive with higher surface area, chemical stability, and non-toxic nature. It is an imperative multifunctional semiconductor material, remarkably significant for photo-catalysis application and it is also used as anti-bacterial materials. This is on the account of its outstanding electrical, optical, chemical and mechanical properties [33]. Embedding ZnO nanoparticles in membrane matrix could result into a steady system, making their physical properties to be connected to their size and having the chemical activity to be linked with their ease of use in the membrane. This will make the application of nano-size scale ZnO integrated in membranes full of promise for the advancement of cost effective and anti-fouling membrane technology [34]. Balta et al. [34] investigated the performance of fabricated MMMs integrated with ZnO nanoparticles. The novel membrane integrated with ZnO nanoparticles exhibited enhanced membrane topographies. Hence, the membrane performed much better than the neat membranes with regards to permeability, rejection of dye and resistance to fouling.

### 2.3 Carbon nanotube Nanofiltration MMM for wastewater treatment

A practical strategy of reducing fouling in wastewater treatment is the fabrication of advanced membrane materials that will possess antifouling properties [35]. Nowadays, carbon nanotubes (CNTs) have found utilizations in the field of nanofiltration membrane technology based on their inherent electrical, thermal, mechanical and separation properties. Among the utmost extensively studied artificial nanomaterials, single-walled carbon nanotubes (SWNTs) have found wider applications as a result of their uncommon basic properties. They are sized in nanometre range, with needle like cylinder prepared via a rolled-up graphene sheet to transform to a graphical shell [36]. A vital means used for tailoring the interactivities of SWNTs with their milieu is functionalization [37]. This kind of functionalization could be noncovalent such as coatings of surfactants, synthetic polymers, or biopolymers [38] or covalent, attained via chemical reactions responsible for linking addends to the nanotube sidewall carbon atoms [39, 40]. Carbon nanotube with a number of concentric graphite shells is termed multi-walled carbon nanotubes (MWCNTs) [41]. The distinctive properties of these carbon nanotubes are their exceptionally high separations;

inner diameter at nano and atomic scaled, smooth, hydrophobic graphitic walls. MWCNTs have appeared to be a novel nano membrane material (NMM) for the separation of liquid and gas because of their large surface area [42]. CNT with well-established framework have the capacity to function as robust pores in membranes used for the applications of water desalination and decontamination [43]. The CNT frictionless channels with specifically controlled pore entrances might at the same time aid the fast movement of water molecules and provide excellent capability for the rejection of salt [12]. For example, the hollow CNT structure affords frictionless movement of water molecules; hence, allow its suitability for the advancement of high flux separation methods [44]. Apposite pore diameters could create energy barricades at the channel entrances, leading to salt rejection and allowing permeation of water via nanotube hollows [45]. CNT pores can also be modified for the purpose of selectively sensing and rejecting ions [46].

Nano-sized particles and CNTs are considered excellent materials used as additives for polymeric membrane due to their high hydrophilicity, good chemical stability and high surface area. Nanocomposite nanofiltration membranes include a composite of carbon nanotubes coated or chemically bonded with metal oxide nanoparticles. They could be taken into consideration as a novel group of filtration materials made of mixed matrix membranes with high surface-functionality [47]. Vatanpour et al. [48] synthesized multi walled carbon nanotubes coated by anatase  $\text{TiO}_2$  nanoparticles through the precipitation of  $\text{TiCl}_4$  precursor on the acid oxidized multi walled carbon nanotubes in order to be employed in the synthesis of nanocomposite polyethersulfone (PES) membranes. Ultraviolet (UV) irradiation on the  $\text{TiO}_2$  contributed to the formation of holes and electrons. The electrons generated would reduce Ti (IV) cations to the Ti (III) state and the holes will oxidized  $\text{O}_2^-$  anions. Oxygen atoms are therefore removed and a group of oxygen vacancies are made on the surface. As a result, water molecules from the surroundings would take up the vacancy and the OH groups adsorbed would contribute to the formation of membrane surface with higher hydrophilicity. Additionally, the radicals produced can avoid the proteins deposition thus reduced membrane surface fouling. These researchers discovered that  $\text{TiO}_2$  nanoparticles used possessed low agglomeration during the application as the nanoparticles exhibited good compatibility with the polymeric components. Incorporation of these additives into the PES membrane matrix led to an increased pure water flux, higher hydrophilicity, better anti-biofouling and decreased membrane surface roughness. Vatanpour and some group [49] synthesized flat sheet asymmetric amine-functionalized MWCNTs/polysulfone nanocomposite NF membranes with diverse nanofiller contents via the phase inversion technique. The membranes were assessed with respect to pure water flux, static protein adsorption and bovine serum albumin filtration. The authors found that the hydrophilicity and pure water flux of the membranes were improved with the upsurge of  $\text{NH}_2$ -MWCNTs dosages. Sumisha et al. [50] fabricated nanofiltration membranes by using functionalized titanate nanotubes via phase inversion process. The neat membrane and MMMs were fabricated by employing polyethylenimine and  $\text{TiO}_2$  particles. The retention performances of  $\text{NaCl}$ ,  $\text{K}_2\text{SO}_4$  and  $\text{CaCl}_2$  ions increased. CNTs were also integrated into alumina/polyethersulfone hollow fibre membranes for the purpose of improving the mechanical property and the effectiveness of water treatment [51]. It was observed that the integration of CNTs could significantly hinder large surface pores formation, leads to reduction in void size of the support layers and enhance the porosity and pore connection of alumina/polyethersulfone membranes. Hence, enhanced flux and rejection.

### 3. Conclusion



The stage of nanotechnology development has totally offered diverse innovative functional materials that have the capacity to be used for the creation of advanced properties for nanofiltration membranes in water treatment. The progress of developing innovative materials with improved properties is a very important issue for the development of the nanofiltration membranes. With several use of integration of nanomaterials in nanofiltration membranes, mixed matrix membranes are being developed for creating advanced properties required for wastewater treatment.

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