



Synthesis and characterization of alumina nano-particles polyamide membrane with enhanced flux rejection performance

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ABSTRACT

This paper presents polyamide (PA) nanocomposite membrane containing alumina nanoparticles synthesized via in situ interfacial polymerization. Polymerization reaction occurred from the aqueous phase of *m*-phenylenediamine and the organic phase of trimesoyl chloride in which alumina nanoparticles were homogeneously dispersed. In the first part of the work, aluminum oxide (Al₂O₃) nanoparticles with average size of 14 nm, were synthesized by an aqueous sol–gel method using precursors of aluminum nitrate and citric acid mixed solution. The as-synthesized Al₂O₃ nanoparticles were characterized by X-ray diffractometer (XRD), scanning electron microscope (SEM), energy-dispersive X-ray spectroscopy (EDX) and Fourier transform infrared spectrometer (FTIR). In the second part, the Al₂O₃ were used to prepare nanoparticles entrapped PA membrane via interfacial polymerization. SEM analysis demonstrated that nanoparticles were dispersed in the membrane and embedded in polyamide chains. Elemental analysis by EDX demonstrated the presence of nanoparticles on the membrane surface. The FTIR data confirm the formation of polyamide layer with Al₂O₃. The performance of the nanocomposite membrane, which was cured at 80 °C for 5 min, was observed to be better than pristine membrane. The existence of nanoparticles in the membrane improves the permeate flux and maintains the salt rejection. It also resulted in enhanced hydrophilicity of the membranes proved by decreased water contact angle.

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1. Introduction

Water sustainable supplies are vital for agriculture, industry, recreation, energy production and domestic consumption. Thus, there is a need to improve the efficiency of water purification technology. Membrane technology is of a great potential for application in water treatment systems. It is used to remove fine particles from water with high separation efficiency [1]. Inorganic materials incorporation into the polymer matrix is being of great interest due to their completely hydrophilic characteristic and their certain functionality to benefit membrane properties of fouling reduction [2]. Therefore, extensive efforts are being devoted to incorporate inorganic nanoparticles into polymeric membrane. It was reported that inorganic materials could be incorporated into membranes by doping and coating technologies [3,4]. However, the doping technology has the disadvantage that inorganic materials are buried in the polymer matrix and in turn rendered non-functional. On the other hand, the coating technology has the disadvantage of the instability of inorganic materials onto polymer surface,

especially for those not subject to chemical bonds or physical restraints between the inorganic materials and membrane matrix. The concept for formation of mixed matrix reverse osmosis membranes, by interfacial polymerization of nanocomposite thin films on porous supports, has been reported [5]. The emergence of nano-technology in membrane materials science could offer an attractive alternative to polymeric materials [6].

Zeolite nano-particles have been used to prepare nanocomposite membrane where first zeolite nano-particles are synthesized via a template hydrothermal reaction followed by a series of complex processes involving template removal, carbonization, sodium exchange and calcination [7]. Titanium oxide nanoparticles have been incorporated into polyamide (PA) thin film membranes by applying an in situ interfacial polymerization procedure [8]. Polyethersulfone–TiO₂ nanoparticle composite membranes made from casting solution consisting of various compositions of polymer solvents (DMF and EtOH) and TiO₂ additive showed significant improvement in fouling resistance [9]. Silica oxide nanoparticles have been incorporated into PA thin film membrane via in situ interfacial polymerization process [10]. Zirconium oxide has been used as a bulk material in polyvinylidene fluoride membranes [11]. ZnO enhanced polyethersulfone membrane has been synthesized by diffusion induced phase inversion in *N*-methyl-pyrrolidone. It has reported that membrane materials embedded with ZnO nanoparticles have

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significantly improved membrane features. It showed lower flux decline and better permeability compared to neat polymeric membrane due to a higher hydrophilicity of the ZnO membranes. ZnO nanoparticles provide a remarkable improvement in the methylene blue rejection potential [12]. Aluminum oxide (Al_2O_3) nanoparticles were also incorporated into a membrane of polyvinylidene fluoride using dimethyl acetamide as solvent [13]. Same type of polymer was used to prepare a membrane by doping with anhydrous and hydrated aluminum oxide particles through in situ particle embedment and subsequent crystal growth under a hydrothermal environment [14].

Recently, an informative review on the use of nanoparticles in polymeric and ceramic membrane structures has been published [15]. Different strategies using a wide range of nanoparticles have been presented for the manufacturing of membrane materials with their potential applications in the field of water treatment. Despite the efforts, there is still a need for investigating different nanocomposite membranes with increased water permeability for energy savings and at the same time improved salt rejection for good water quality; along with long service life of the membrane that is resistant to fouling. Aluminum oxide (Al_2O_3) is one of the most stable inorganic materials, and thus it is used as ultra-filtration membranes [16]. In addition, it is inexpensive, non-toxic, highly abrasive and resistant. Remarkably, the use of alumina nanoparticles in membranes, especially in PA membrane synthesis by interfacial polymerization, has not yet been investigated to a large extent, in spite of their wide availability and applications in water treatment technology.

In this study, the Al_2O_3 nanoparticles were synthesized and characterized. Then, they were incorporated into a thin film polyamide membrane via interfacial polymerization process. The performance of the membrane was evaluated in terms of permeate flux and salt rejection.

2. Experimental

2.1. Reagents and chemicals

Aluminum nitrate, citric acid, polysulfone (PS) *m*-phenylenediamine (MPD), trimesoyl chloride (TMC), *n*-hexane and *N,N*-dimethylformamide (DMF) were obtained from Sigma Aldrich and used as received. The water employed in the syntheses processes was double distilled. The other chemicals and reagents were of high analytical purity, obtained from Sigma Aldrich and used as received.

2.2. Synthesis of alumina nanoparticles

The nanoparticles of alumina were prepared by the following process. The aluminum nitrate and citric acid were dissolved into deionized water. The concentrations of aluminum nitrate and citric acid were 0.5 M. The solution was continuously stirred for 8 h at a temperature of 80 °C, until it turned to a yellowish sol. It was then, rapidly heated to 100 °C and stirred constantly. The temperature of the system was then increased up to 200 °C and kept for 3 h until a fluffy, polymeric citrate precursor was obtained. It was then allowed to dry and then it was ground to a fine powder. The final step is the calcinations process, which was performed at 800 °C for 3 h. The resulted nanoparticles of alumina were characterized.

2.3. Preparation the nanocomposite membranes

The membranes were synthesized by depositing an active top layer on the polysulfone (PS) support. A typical membrane was prepared by initially immersing PS support in 2% (w/v) solution of *m*-phenylenediamine in water for 120 s, followed by immersing

into 0.1% (w/v) trimesoyl chloride in *n*-hexane solution for 40 s. This resulted in lamination of an ultra-thin film of PA over the surface of PS support. Then, the membrane was cured at around 80 °C for 5 min and then washed thoroughly. The alumina nanoparticles synthesized in this work were used to be embedded in polyamide membrane. The membrane was prepared by mixing the as-synthesized alumina nanoparticles (1 wt.%) in TMC solution. The dispersion was enhanced by sonication of the matrix for 1 h.

2.4. Characterization

The as-synthesized alumina nanoparticles were characterized by X-ray diffractometer (XRD) (Shimadzu XRD Model 6000) which was employed to determine crystalline phases and average crystalline size. The morphology and microstructure of the composite was examined by means of scanning electron microscope (SEM, FEI Nova-Nano SEM-600, Netherlands). The quantitative analysis of the nanoparticles was performed by the mean of energy-dispersive X-ray spectroscopy (EDS or EDX). Fourier transform infrared spectroscopy (FTIR), (FTIR-100 Spectrometer), spectra were measured at room temperature using the KBr Pellet technique. Pellets were prepared by mixing 1 mg of the alumina nanoparticles with 300 mg of KBr in an agitate mortar, and then pressing the resulting mixture successively under a pressure of 12 Pa for 3 min. The spectra of the composite's samples were measured between 4000 and 400 cm^{-1} .

The morphology and microstructure of the as-synthesized nanocomposite membrane was examined by means of scanning electron microscope (SEM, FEI Nova-Nano SEM-600, Netherlands). The quantitative analysis of the membranes was performed by the mean of energy-dispersive X-ray spectroscopy (EDX). Fourier transform infrared spectrometer (FTIR-100 Spectrometer) was used to confirm the formation of polyamide. The measurements were performed at room temperature. The spectra of the composite's samples were measured between 4000 and 400 cm^{-1} .

2.5. Membrane evaluation

The nanocomposite membrane was evaluated in a flow cell of the reverse osmosis test kit. The membrane sample was supported in the dead-end testing cell by a porous stainless steel disk. The performance was evaluated in terms of flux and solute rejection of feed solutions. The feed solutions, used in evaluating the performance of the membrane, were aqueous solutions of 1000 ppm NaCl. The tests were performed at a pressure of 1 MPa and a temperature of 25 °C. The effective membrane area (active area) of the test membrane was measured as 4 cm^2 . The flux was calculated by measuring permeates penetrated through the membranes per area of the membrane per unit time. The solute rejection was achieved by determining the concentration of the feed solutions and the permeate concentration. The rejection was calculated by the following equation:

$$\text{Rejection}(\%) = [(C_f - C_p)/C_f] \times 100$$

where C_f is the concentration of the feed solution and C_p the concentration of the permeate.

3. Results and discussion

3.1. Characterization of the synthesized Al_2O_3 nanoparticles

The prepared alumina nanoparticles were characterized by EDX, SEM, XRD and FTIR. The EDX spectrum is presented in Fig. 1a. It shows two peaks reflecting the presence of aluminum and oxygen. The quantitative analysis is presented in the inset of Fig. 1a. The

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