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Synthesis, characterization and performance studies of polysulfone/bentonite nanoparticles mixed-matrix ultra-filtration membranes using oil field produced water

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ABSTRACT

The present paper deals with the performance studies of newly formulated polysulfone–bentonite ultra-filtration membrane for the purification of oily produced water and the results were compared with standard polysulfone–silica membrane. Membranes were prepared via phase inversion technique by blending silica and bentonite nanoparticles separately with polysulfone and N-methyl-2-pyrrolidone solution. Particle size distribution, specific surface area, morphology, FTIR, elemental analysis and cation exchange capacity of silica and bentonite nanoparticles were determined. Viscoelastic properties of membrane casting solution were measured to determine the maximum applied strain (i.e., $\leqslant 70\ s^{-1}$) to avoid structural degradation while casting. The surface morphology of membranes was analyzed by field emission scanning electron microscopy and atomic force microscope. Porosity, contact angle and equilibrium water content of individual membrane was also determined. The membranes were subjected to ultra-filtration characterization such as compaction factor, hydraulic permeability, pure water flux, percent oil rejection and molecular weight cut-off for performance studies. Finally, membrane performance of silica and bentonite blended polysulfone membranes was compared by measuring permeate flux, flux declination rate and oil rejection. An improved performance of polysulfone–bentonite membranes was obtained for desired percent rejection over polysulfone–silica membrane.

1. Introduction

A large volume of water is usually produced as a byproduct (called produced water) during oil and gas production, which is mixed with dispersed oil, grease, dissolved and suspended solids [1]. Discharging of produced water pollutes surface and underground water as well as soil. As per environmental regulation the permitted oil and grease limits for discharged produced water is $10\ mg\ L^{-1}$ [2]. The typical oil concentration in produced water generated during oil and gas operation is between 100 and $1000\ mg\ L^{-1}$ and even may go higher depending on the oil well, which cannot be discharged directly to the environment without treatment [3]. Produced water may also be injected to some extent to the reservoir for enhanced oil recovery after removing oil and suspended solids to avoid the damage in oil formation [4]. Moreover, the direct disposal of produced water to the environment causes disturbance to ecology. Thus, efficient and economic treatment of produced water before disposal is very important.

Several techniques are available in the literature for the treatment of produced water. In this regard, the gravity settling separation and mechanical coalescence methods are the well-known traditional treatment processes [5]. Chemical emulsion breaking is also an effective way to separate oil from produced water [6]. The coagulation and air flotation [7], electrostatic and electro coagulation separation methods [8] were also applied to separate oil and water from produced water. However, these methods would lead to a huge production of sludge with complicated operational problems. Microwave treatment [9] and heat treatment [10] have been occasionally applied to treat oily water in recent years. But above methods are highly capital intensive with high maintenance and operating cost. Moreover, these are environmentally unfit and difficult to separate lower size oil droplets [11–14].

Under such circumstances, the use of membranes offers a potential solution to the problem of oily produced water purification. Common membrane separation techniques employed for the

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Nomenclature

A	area of membrane (m^2)
c_f	oil concentration in feed ($mg\ L^{-1}$)
c_p	oil concentration in permeate ($mg\ L^{-1}$)
G^*	complex viscosity of casting solution (Pa s)
G'	elastic (storage) modulus of casting solution (Pa)
G''	viscous modulus of casting solution (Pa)
J_p	permeate flux ($L\ m^{-2}\ h^{-1}$)
L	membrane thickness (m)
P	trans-membrane pressure, TMP (Pa)
P_m	hydraulic permeability of membrane ($m Pa^{-1}\ s^{-1}$)
R	oil rejection (%)
t	time of filtration (h)
T	transmittance (%)
V_p	volume of permeate collected (L)
w_0	weight of wet membrane (kg)
w_1	weight of dry membrane (kg)

Greek symbols

Δ	difference
γ	strain (or amplitude) applied to rotational viscometer (s^{-1})
φ	membrane porosity (%)
ρ_w	density of water ($kg\ m^{-3}$)
ω	frequency applied to rotational viscometer (Hz)

separation of oil from oily waste water are: (i) microfiltration [15], (ii) nanofiltration [16], (iii) prevaporation [17], (iv) electrocoagulation [18], (v) reverse osmosis [19,20] and (vi) ultra-filtration (UF) [3].

Polysulfone (PSf) is one of the most popular thermoplastic polymer which is used frequently in the casting of UF membranes for water treatment [3,21,22]. It offers excellent mechanical strength and thermal stability during operation. The phase inversion technique is commonly employed to prepare PSf membranes. However, due to hydrophobic nature, PSf membrane exhibits strong adsorption and deposition of foulants (e.g. colloids, proteins and particles) on the membrane surface and as well as inside the membrane pores. This leads to the lowering of permeation flux and deterioration in performance [23]. Fouling of PSf membranes during filtration could be reduced by improving the hydrophilic characteristic in the membrane. In this regard, several studies have been made to improve the hydrophilicity of membrane via redox initiated grafting, plasma treatment, UV induced graft polymerization, etc. [24,25]. However, these methods are suffered by the drawback of complicated steps and the use of harsh chemicals. Surface coating with a hydrophilic polymer on membrane surface is another way to increase the hydrophilicity of PSf membrane [26]. The drawback of this method is frequent de-lamination of coating layer from the surface. The common properties that control the performance of PSf membranes are: porosity, molecular weight cut-off (MWCO), average pore size, hydrophilicity, permeability, surface roughness, etc., and these properties are dependent on casting solution composition (i.e., base polymer, additive and solvent), casting temperature, solvent evaporation rate and thickness of cast solution. Different additives and their dosages in the casting solution have significant influence to control the membrane properties. Several inorganic additives (e.g., Al_2O_3, SiO_2, TiO_2, ZrO_2, LiCl, Fe_2O_3, CdS, etc.) and polymeric additives [e.g., polyethylene glycol (PEG), polyvinylpyrrolidone (PVP), etc.] are blended with casting solution to obtain desired membrane properties [27–31]. Song et al. [29] studied the effect of silica nanoparticles in PSf membrane and found that the pure water flux (PWF) of membrane containing 1.0 wt% silica was 2.3 times higher than that of PSf membrane. Wu et al. [31] prepared the PSf–silica membranes and found that the water flux was nearly twice that of plain PSf membranes when silica in the membranes was 0.3 wt%. Chakrabarty et al. [3] studied the effect of PVP with different molecular weight on the structure and permeation properties of PSf membrane and reported that number of pores and pore area were increased with a molecular weight of PVP. But due to the soluble nature of PVP in water, membrane performance reduced rapidly due to washing-out of PVP and membranes get reduced hydrophilicity. Many researchers have reported the effect of PEG with different molecular weight on pure water flux, average pore size, porosity and hydraulic permeability of membranes [3,32]. Panda and De [32] also studied the effect of different molecular weight of PEG on membrane porosity and found that the porosity of the membrane increased with PEG dosing up to 8.0 wt% and thereafter decreased rapidly. Arthanareeswaran et al. [33] studied the effect of LiCl and SiO_2 separately in cellulose acetate membranes and found that marginal improvement in pure water flux from 29.60 to 34.28 $L\ m^{-2}\ h^{-1}$ was obtained when additive dosing kept at 5 wt%. Eren et al. [34] prepared PSf membrane by blending poly[2,2′-(m-phenylene)-5,5′-dibenzimidazole] in casting solution and found that marginal pure water flux improvement in composite membrane was observed over PSf membrane.

Bentonite (montmorillonite) is frequently used as an additive to improve the viscosity of drilling fluid. It consists of several closed packed platelets and each platelet has tetrahedral–octahedral–tetrahedral layered structure. Bentonite has a strong affinity toward water and swelled easily through the crystalline and osmotic swelling when it was suspended in water [35]. Therefore, bentonite nanoparticles may be considered as an additive in PSf membrane that can be used for the purification of produced water.

Though the use of different additives in PSf membrane was reported in literature, to the best of our knowledge, there is no published literature on the additive effect of bentonite nanoparticles on morphology and permeation properties of PSf membrane. Therefore, the objective of the present study is to examine the effect of bentonite nanoparticles in PSf membrane for the purification of produced water. In this paper, bentonite and silica nanoparticle based PSf membranes were prepared using N-methyl-2-pyrrolidone solvent. The effects of nanoparticles to the blended membranes were determined by water contact angle, equilibrium water content (EWC), porosity, compaction factor, pure water flux, membrane hydraulic resistance, separation of oil from synthetic produced water. The MWCO of the prepared membranes was determined by using neutral solution of polyethylene glycol (PEG) with different molecular weight. The bentonite and silica particles present in the blended membranes was also analyzed by electron diffraction spectra (EDS). The morphology of the membranes was studied by field emission scanning electron microscopy (FE-SEM). The surface roughness of the blended membranes was also studied by atomic force microscope (AFM).