



Adsorptive removal of phenolic compounds using cellulose acetate phthalate–alumina nanoparticle mixed matrix membrane



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HIGHLIGHTS

- Composite membrane of cellulose–acetate–phthalate and alumina nanoparticle is cast.
- Surface charge of the membrane changes with nanoparticle concentration and pH.
- Separation of phenolic compounds occurs due to adsorption.
- The removal efficiency is maximum for 20% nanoparticle with 91% removal of catechol.
- Transmembrane pressure drop has negligible effect on solute separation.

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ABSTRACT

Mixed matrix membranes (MMMs) were prepared using alumina nanoparticles and cellulose acetate phthalate (CAP) by varying concentration of nanoparticles in the range of 10 to 25 wt%. The membranes were characterized by scanning electron micrograph, porosity, permeability, molecular weight cut off, contact angle, surface zeta potential, mechanical strength. Addition of nanoparticles increased the porosity, permeability of the membrane up to 20 wt% of alumina. pH at point of zero charge of the membrane was 5.4. Zeta potential of the membrane became more negative up to 20 wt% of nanoparticles. Adsorption of phenolic derivatives, catechol, paranitrophenol, phenol, orthochloro phenol, metanitrophenol, by MMMs were investigated. Variation of rejection and permeate flux profiles were studied for different solutes as a function of various operating conditions, namely, solution pH, solute concentration in feed and transmembrane pressure drop. Difference in rejection of phenolic derivatives is consequence of interplay of surface charge and adsorption by alumina. Adsorption isotherm was fitted for different solutes and effects of pH were investigated. Catechol showed the maximum rejection 91% at solution pH 9. Addition of electrolyte reduced the rejection of solutes. Transmembrane pressure drop has insignificant effects on solute rejection. Competitive adsorption reduced the rejection of individual solute.

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1. Introduction

Petrochemical and other chemical industries produce aromatic and semi-volatile phenol and phenolic derivatives which are extremely toxic [1] and carcinogenic [2]. This class of compounds is in the 11th position out of 126 chemicals, which are labeled as priority pollutants according to United States Environmental

Protection Agency. Exposure to phenol leads to high irritation to eyes, skin and mucous, causes headache and dizziness. Long term exposure results in high blood pressure, liver and kidney damage. Existing technologies for removal of phenolic compounds are solvent extraction [3–6], adsorption [7], chemical oxidation [8,9], biological treatment [10] and distillation [11]. However, all these methods require high input of energy and/or additional chemicals and hence are cost intensive. Moreover, low separation efficiency and post processing of streams are important issues. Thus, there is a need to develop energy efficient and economical method for separation of phenolic compounds. Membrane based processes can offer attractive alternative in this regard. Reverse osmosis (RO) can be used to remove phenol [12]. But, low permeability and higher pressure limit the commercial applicability of RO membranes. Micellar enhanced ultrafiltration (MEUF) is another alternative for removal of phenol [13]. However, it involves surfactant that increases the operating cost and hence, recovery of surfactants from the

Abbreviation: np, nanoparticle; CAP, cellulose acetate phthalate; CAPAI-0, membrane with CAP and 0% alumina nanoparticle; CAPAI-10, membrane with CAP and 10% alumina nanoparticle; CAPAI-15, membrane with CAP and 15% alumina nanoparticle; CAPAI-20, membrane with CAP and 20% alumina nanoparticle; CAPAI-25, membrane with CAP and 25% alumina nanoparticle; CC, catechol; PNP, *para*-nitrophenol; OCP, orthochlorophenol; Ph, phenol; MEUF, micellar enhanced ultrafiltration; MMM, mixed-matrix membrane; MNP, meta-nitrophenol.

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Nomenclature

A	membrane area (m^2)
C_f	concentration of solute in the feed (kg/m^3)
C_p	concentration of solute in the permeate (kg/m^3)
D_i	Dielectric constant
J_w	pure water flux ($\text{l}/\text{m}^2 \text{ h}$)
l	membrane thickness (m)
M_w	molecular weight cut off (kDa)
Q	amount of permeate collected (m^3)
R	rejection (%)
r_m	pore radius, (cm)
V_p	Streaming potential coefficient (V/Pa)
w_l	dry weight (kg)
w_o	wet weight (kg).

Greek symbols

ΔE	potential difference across the membrane (V)
ΔP	transmembrane pressure (Pa)
Δt	time difference (s)
ε	porosity
λ	conductivity of bulk solution (S/m)
μ	viscosity of water (Pa s)
ρ_w	density of water (kg/m^3)
ζ	zeta potential (V).

retentate is a critical issue in MEUF. Composite ultrafiltration membrane with high selectivity toward phenolic compounds can be proven as viable alternative owing to higher permeability and lower pressure requirement.

Choice of base polymer plays a crucial role in selection of membrane. Derivatives of cellulose acetate (CA) are popular polymers to cast organic membranes. Advantages of CA over other polymers are, good mechanical strength, biocompatibility, good desalting and high permeate flux. Hydrophilic nature of CA also enhances the anti-fouling property [14–17].

Nanoparticle doped mixed matrix membrane (MMM) has generated considerable interest among researchers recently [18–20]. This is primarily due to the fact that tailor made membranes can be fabricated targeting specific application [21,22]. Presence of charged nanoparticles in the membrane matrix has potential application in wastewater treatment by electrostatic interaction [23–25]. MMMs improve various properties, like mechanical [26], thermal [27], magnetic [28] and electrostatic. It can enhance solute diffusivity [24], antibacterial property [29] and reduce flux decline [30,31]. MMMs have widespread application in case of gas separation and pervaporation [32]. In case of treatment of liquid stream, some of its applications include separation of sulfur from gasoline [33], silver using functionalized silica [34], humic acid using TiO_2 [35], metal ions using polyelectrolyte [36], lead by hydrous manganese dioxide [37], arsenic [38] from aqueous stream. MEUF for removal of methylene blue using titanium dioxide–polyvinylidene fluoride MMM is also reported [39]. Zeolite, modified silica and graphene impregnated composite membranes are also used for separation of salts in desalination applications [40–42].

Addition of alumina as an inorganic additive has been studied considerably in past. MMM of granular alumina and CA shows interesting properties like flux enhancement, good mechanical strength [43]. Alumina nanoparticles in PVDF membranes at low percentage improved membrane hydrophilicity, porosity and protein retention [44]. Most studies reported doping of nanoparticle in small quantities, typically less than 5 wt%. While added in large proportion, significant amount of nanoparticles are retained in the matrix even

after leaching during phase inversion to impart desirable properties like porosity, permeability, etc. [45].

In the present work, MMM was prepared using cellulose acetate phthalate (CAP) as base polymer, di-methyl formamide (DMF) as solvent and alumina nanoparticle as dopant. The superiority of CAP compared with cellulose acetate is presence of numerous acidic and carbonyl functional groups on its structure. These groups, in addition to impregnation of nanoparticle in the membrane induce more surface charge. The morphology, wettability and porosity of the membrane are also modified. This is manifested through the removal efficiency and performance during filtration of polar phenolic compounds.

2. Materials and methods

2.1. Materials

Cellulose acetate phthalate (CAP) was purchased from GM Chemicals Company Limited, Mumbai, India. *N,N*-dimethyl formamide (DMF), polyethylene glycol (PEG) (Molecular weight 35, 20, 10, 6, 4 and 0.4 kDa), all phenolic derivatives, *meta*-nitrophenol (MNP), catechol (CC), *para*-nitrophenol (PNP), orthochlorophenol (OCP) and phenol were procured from Merck (India) Ltd., Mumbai, India. Dextran (molecular weight—70 kDa) was purchased from Sigma Aldrich Chemicals (USA). The alumina nanoparticle was provided by US Research Nanomaterials Inc. (Houston, USA). Bovine Serum Albumin was purchased from Sisco Research Laboratories Pvt. Ltd., Mumbai, India.

2.2. Membrane preparation

MMM using CAP and alumina nanoparticle was prepared by phase inversion method. Initially, DMF was heated to 60 °C and alumina nanoparticles were mixed in various weight percentage (10%, 15%, 20% and 25%). The mixture was sonicated for 2 h to prevent agglomeration [45,46]. CAP being a nanoparticle-stabilizer itself was added (15 wt%) slowly to the suspension under continuous stirring. The mixture was again sonicated for 6 h with occasional stirring to prevent agglomeration [43]. Casting solution for membrane without alumina was prepared by mixing CAP (15 wt%) in DMF (75 wt%).

Non-woven polyester fabric (product number: TNW006013, supplied by Hollytex Inc., New York, USA) was attached to the glass slides and was fixed by tape. The cast solution was drawn manually with a speed of 20 mm/s using a doctor's blade with gap 150 μm . The membranes were put in water bath at 27 °C for 16 h to complete the phase inversion and labeled as CAPAI-0, CAPAI-10, CAPAI-15, CAPAI-20, CAPAI-25 indicating weight percent of nanoparticles in casting solution as 0%, 10%, 15%, 20% and 25%, respectively.

2.3. Experimental set-up

Membrane compaction, rejection of protein and phenolic compounds were carried out in a 600 ml batch cell with 34 mm diameter and effective filtration area 36.3 cm^2 . A nitrogen cylinder was used to pressurize the cell. Cumulative weight of permeate was collected on a digital balance, connected to a computer via data-logging device to monitor cumulative weight as a function of time. The schematic of the experimental set-up is shown in Fig. 1.

2.4. Experimental procedure

The feed solution of each batch was prepared by mixing measured amounts of solutes in distilled water. The concentration of catechol, PNP, MNP, OCP and phenol, were 100, 100, 50, 100 and 100 ppm, respectively. In case of solution of mixed solutes (PNP,

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