



Investigation of antifouling and disinfection potential of chitosan coated iron oxide-PAN hollow fiber membrane using Gram-positive and Gram-negative bacteria



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ABSTRACT

Chitosan coated iron oxide nanoparticles were impregnated into polyacrylonitrile based hollow fiber membrane. The molecular weight cut off was varied in the range of 120 to 145 kDa with the concentration of nanoparticles. Incorporation of nanoparticles improved the permeability, mechanical property and hydrophilicity of the membrane. The contact angle of the membrane decreased from 80° to 51° and the permeability increased by 31% at 0.5 wt% nanoparticles concentration. The antibacterial and antifouling property of the membrane were investigated with two biofilm causing Gram positive and Gram negative bacteria. The damage of cell membrane was directly confirmed by release of cellular constituent absorbing in 260 nm. The cellular deformation on the membrane surface was evident by direct microscopic observation in FESEM. This damage was likely caused by electrostatic interaction between NH_3^+ group of nanoparticles and anionic components of phosphoryl group of bacteria. The hollow fiber membrane shows promising antibiofouling property even after long experimental run as evident by 95% flux recovery ratio. The effect of operating conditions on rejection and flux profile was investigated during long experimental run. The result indicated that there was no detectable iron in the permeate sample that could impose adverse health hazard.

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1. Introduction

The lack of clean and safe drinking water is a significant health concern throughout the world. Water resources are presumed to be vulnerable to disease causing microorganisms and hence, must be disinfected before consumption [1]. Long-term exposure of common disinfectants helps to build up microbial resistance among pathogens and has several drawbacks like high operational and maintenance cost [2]. Many of them even produce harmful disinfection by products, raising issues for environmental safety [3].

Membrane based separation process is the most versatile disinfection technology due to its high efficiency, ease of implementation, cost effectiveness and low environmental impact [4]. Polyacrylonitrile (PAN) has been widely used as base polymer for membrane fabrication due to its excellent chemical stability and mechanical strength [5].

However, PAN based membranes tend to be fouled by proteins and microorganisms during the separation process due to its hydrophobicity [6]. Recent study shows that some of the biofilm causing bacteria are resistant to chlorination and eventually contributes to severe membrane fouling [7]. This rapidly decreases the permeability and throughput of the membrane. Besides, frequent chemical cleaning not only shortens the life of the membrane but also increases the operating cost [8]. To reduce the fouling and contamination caused by water borne bacteria, it is necessary to endow the membrane with hydrophilic antimicrobial agent [9].

Chitosan (CS), being hydrophilic and natural nontoxic biopolymer, has been studied extensively for its role as an antimicrobial agent against a broad spectrum of bacteria [10]. In recent years, CS based hollow fiber (HF) mixed matrix membranes were used to remove heavy metals [11]. Vincent et al. [12], prepared CS based HF module for solvent extraction of chromate [12]. Han et al. [13], reported efficient adsorption of copper ions using real wastewater by CS based HF membrane containing CS as high as 18 wt% [13]. Mirmohseni et al. [14], prepared neat CS based HF membrane by dry wet spinning method for adsorptive removal of reactive blue 19 [14]. The antibacterial potential of PAN-CS films and nanofibers against *Escherichia coli*, *Staphylococcus aureus*, and *Micrococcus luteus* was reported by Kim et al. [15]. However, pure CS membrane has some major drawbacks, like, acid solubility of CS and poor mechanical property [14,15].

Abbreviations: PAN, polyacrylonitrile; MWCO, molecular weight cut off; EPA, Environmental Protection Agency; WHO, World Health Organization; *E. coli*, *Escherichia coli*; *S. aureus*, *Staphylococcus aureus*; SEM, scanning electron microscopy; DMF, dimethylformamide; PEG, polyethylene glycol; HF, hollow fiber; FTIR, fourier transform infrared; AFM, atomic force microscopy; CFU, colony forming unit; TMP, transmembrane pressure drop; CFR, cross flow rate.

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Modification of CS with nanoparticles has triggered strong interest due to rapid dissolution, enhanced antibacterial activity, high surface area and positive charge density compared to unmodified CS [16,17,18]. Iron oxide nanoparticles (IONP) can be a good candidate for this modification due to the large surface area and outstanding antimicrobial property [19]. In our previous work, we have physically coated CS layer on IONP incorporated PAN based ultrafiltration flat sheet membrane [20,21]. The membrane offered *anti*-adhesive surface with excellent antibacterial property during disinfection of surface water [20]. The prepared membrane also showed high adsorption capacity of humic acid in spiked solution [21]. However, these membranes became dense, less permeable and hydrophobic due to pore blockage during physical coating of CS [21]. Thus, it is necessary to blend some hydrophilic nanoparticles to obtain good antibacterial membrane without compromising hydrophilicity and permeability. It is widely accepted that CS coated IONP (CSNP) can be of great potential to improve hydrophilicity and antifouling property due to the active amino and hydroxyl groups [22]. Additionally, CSNP exhibits higher dispersion ability compared to pure CS [23]. Though, research has already been done to assess the antimicrobial and antifouling potential of pure IONP-PAN based flat sheet membrane [24], no report is still available on antibacterial and antifouling properties of CSNP incorporated PAN based HF membrane.

The present study was undertaken to prepare PAN based HF membrane containing CSNP with high antibacterial property to reduce the membrane fouling caused by bacteria during water filtration without compromising the permeate flux. To the best of authors' knowledge this is the first ever attempt to explore the antibacterial efficiency of CSNP doped HF membrane against biofilm causing Gram positive and Gram negative bacteria.

2. Materials and methods

2.1. Materials

PAN co-polymer of average molecular weight 150 kDa was purchased from M/s, Technorbital Advanced Materials Pvt. Ltd., Kanpur, India. *N,N*-dimethyl formamide (DMF), sodium hydroxide (NaOH) and potassium nitrate were procured from M/s, Merck (India) Ltd., Mumbai, India. Polyethylene glycol (PEG) (molecular weight 4, 6, 10, 20 and 35 kDa) was supplied by M/s, S. R. Ltd., Mumbai, India. Dextran (average molecular weight 70 kDa) and polyethylene glycol of average molecular weight 100 kDa and 200 kDa were procured from M/s, Sigma Chemicals and M/s, Aldrich Chemicals, USA, respectively. CS (molecular weight: 190 kDa; degree of deacetylation: ~85%) was supplied by M/s, Aldrich Chemicals, USA. Acetic acid (glacial) of analytical grade was procured from M/s, Central Drug House, New Delhi, India. *Pseudomonas aeruginosa* (NCIM 2036, length: 1.5 μ m, thickness: 0.8 μ m) and *Staphylococcus aureus* (NCIM 5345, diameter: 1 \pm 0.2 μ m) were purchased from National Collection of Industrial Microorganism (NCIM) Pune, India.

2.2. Synthesis of CSNP

CSNP was synthesized by previously described method with some modification [25]. Typically, 0.5 g of solid CS were dissolved in 50 ml 1% (v/v) aqueous acetic acid to prepare a clear solution. 2 g of ferric

chloride and 1 g of ferrous sulphate were added to this solution and stirred for 30 min until the color of the solution changes to pale yellow. Afterwards, 20 ml sodium hydroxide solution (0.08 M) was slowly added and sonicated for 40 min. This procedure facilitates the coating of chitosan simultaneously with the synthesis of Fe₃O₄ nanoparticles [25]. Finally, the black precipitate was collected through magnetic separation and thoroughly washed several times with distilled water to remove impurities. Finally, the nanoparticles were dried at room temperature and ground into fine powder.

2.3. Characterization of nanoparticles

Morphology of nanoparticles was observed using a field emission scanning electron microscopy (model: JSM-7610, JEOL, Japan). The size and zeta potential of the nanoparticles were measured using zetasizer (model: Zetasizer nano ZS 90) supplied by M/s, Malvern instruments, Worcestershire, England. Brunauer–Emmett–Teller (BET) surface area was measured by BET instrument, supplied by Quantachrome instruments, Florida, USA (model: AUTOSORB-1). The presence of surface functional groups on the nanoparticles was investigated by Fourier transform infrared (FTIR) spectroscopy (supplied by M/s, Perkin Elmer, Connecticut, USA; model: Spectrum 100) in the range of 400–4000 cm⁻¹.

2.4. HF module preparation

2.4.1. Preparation of spinning solution

Initially, DMF was heated to 60 °C and various dosages (0.2, 0.4 and 0.5 wt%) of CSNP were added and sonicated for 1 h. 15 wt% PAN was added slowly to the suspension under continuous stirring by mechanical stirrer. The mixture was again sonicated for 5 h to obtain homogeneous solution. Casting solution for membrane without CSNP was also prepared by mixing PAN in DMF and named as pure PAN. Three casting solutions with different concentration of CSNP (0.2, 0.4 and 0.5 wt%) were prepared and named as PANCNP0.2, PANCNP0.4 and PANCNP0.5. The detailed composition of different HF membranes is given in Table 1.

2.4.2. Fiber spinning and module preparation

The heart of the HF spinning set up was the needle assembly containing disposable syringe. Corresponding specifications are presented in Table 2. In brief, a smaller needle (inner diameter 0.0005 m) was inserted into an outer needle (inner diameter 0.0012 m) about 0.01 m above the tip. A leak proof junction was prepared using epoxy putty with brand name M-seal. The polymer tank was directly connected to the needle assembly. The spinning solution was immediately transferred to the polymer tank of HF spinning machine. HF was spun using distilled water as the bore fluid at 25 °C temperature. To flow the polymer solution, pressure was maintained at 690 kPa using a nitrogen cylinder in the polymer tank. The valves of both polymers and water lines were opened at this pressure. The water flow rate was fixed at 20 ml/min and the flow rate of polymer solution was 3 g/min. Extruded HF's were allowed to fall on a water bath, maintained at room temperature and was kept inside the gelation bath (water bath) for 5 min. After that, it was wound by a spool and kept in distilled water for 24 h to allow the phase separation to be complete. The fibers were cut in 20 cm length. A bundle of total 70 fibers was packed in a 1/

Table 1
Antibacterial efficiency of the membranes.

	Polymer (PAN) (wt%)	Solvent (DMF) wt (%)	CSNP wt (%)	MWCO (kDa)	Number of colonies/ml		Antibacterial efficiency (%)	
					<i>P. aeruginosa</i>	<i>S. aureus</i>	<i>P. aeruginosa</i>	<i>S. aureus</i>
Pure PAN (control)	15	85	–	–	102	120	–	–
PANCNP0.2	15	84.8	0.2	120 \pm 6	35	40	65 \pm 3	67 \pm 3
PANCNP0.4	15	84.6	0.4	133 \pm 8	27	15	78 \pm 4	85 \pm 4
PANCNP0.5	15	84.5	0.5	145 \pm 9	0	0	100	100

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