



Biofunctionalized poly (amic) acid membranes for absolute disinfection of drinking water



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ABSTRACT

The integration of biological building-blocks with synthetic nanomaterials may permit unprecedented ability to detect, disinfect and completely remove pathogens in water. We hereby described the synthesis of biodegradable, interpenetrating polymeric networks of poly (amic) acid (PAA), glutaraldehyde-derivatized PAA (PAA-GA) and chitosan-modified poly (amic) acid (PAA-CS) using phase-inversion procedures. The characterization data from NMR, FT-IR, SEM and cyclic voltammetry confirmed the successful formation of electroactive, bifunctional, glutaraldehyde-linked PAA membranes. Toxicological, electrochemical and mechanical characterization data showed the successful formation of non-toxic, biodegradable, porous, free-standing and mechanically strong membranes. PAA-GA showed the highest modulus of 568.1 Mpa followed by PAA-CS-GA (495.0 Mpa). The optimized membranes were tested against three of the most common drinking water contaminants, namely *Escherichia coli*, *Citrobacter freundii* and *Staphylococcus epidermidis* with 100% removal achieved using dead end filtration and tangential flow filtration.

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1. Introduction

Water treatment has been accepted as one of the most crucial topics for a sustainable environment [1–3]. Most sources of public water supplies are not of suitable quality for consumption without some form of treatment [4,5]. In accordance with the Safe Drinking Water Act (SDWA), the U.S. EPA has ruled that all surface water must be filtered and disinfected before consumption unless the purveyor can significantly justify avoidance of filtration [6,7]. Therefore, the limitation of the availability of resources in small communities can create tremendous difficulties in complying with the SDWA. Consequently, there is a need to develop low-cost, innovative technologies that can efficiently remove microbial contaminants from drinking water.

Disinfection is the most obvious approach for remediation of microbial contaminants. There are two main approaches to disinfection: The first approach relies on physically blocking microorganisms from entering into the drinking water. The second focuses on deactivating the microorganisms already present in the media [4]. Filtration is commonly used for blocking microbial

entry into drinking water, and can be driven by temperature, osmotic difference and pressure. Membrane-based water filtration (MWF) processes are currently among the most important and versatile technologies for conventional drinking water production, wastewater treatment, desalination, and reusable water [8,9]. MWF technologies are very appealing due to their in situ usability and their application in emergency situations, as well as in small communities trying to comply with the SDWA. The pore size of the membrane is one of the most important parameters for capturing and inhibiting the microorganisms [10], and 0.2 µm is the effective pore size to prevent bacterial leakage through membranes [11,12]. However, current MWF technologies are energy intensive and very costly [3].

Newer technologies have been developed to address these challenges. For instance Liu et al. reported Ag/TiO₂-based nanofiber membrane in which nearly 100% of *Escherichia coli* was inactivated in a dead-end membrane separation system [13]. Similarly, Lin et al. reported silver, nanoparticle-enhanced, alginate beads in which disinfection and filtration were simultaneously performed in a packed column system [14]. Quang et al. reported silver nanoparticle-enhanced silica beads in which disinfection and filtration were also simultaneously performed in a packed column system [15]. Lewis et al. have reported the development of reactive, nanostructured membranes for water purification using enzyme catalysis and iron-catalyzed, free radical reactions within pore-functionalized synthetic

Abbreviations: PAA, Poly (amic) acid; GA, Glutaraldehyde; CS, Chitosan

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membrane platforms [16,17]. Interfacing some nanomaterials with drinking water may present new hazards to human health and the environment. Safer design of materials may prevent making these a new source of contamination after usage [18]. Previously, we have reported the synthesis of a new class of flexible, nanoporous phase-inverted PAA membranes [21–24]. Safer nanomaterials can be designed by integrating biodegradable polymers and controlling the chemistry, porosity, and the method of desolvation. These include the incorporation of natural products and biopolymer—such as green tea extract, cinnamon bark oil, terpenes, and chitosan [18–21]. The aim of the present work is to develop free-standing, flexible, nano-sized porous, biodegradable, nontoxic and biofunctional PAA membranes for disinfection of tap water. We hereby report a very promising approach to MWF membranes with subsequent application for absolute disinfection of water.

2. Materials and methods

2.1. Materials

The following reagents were purchased from Sigma-Aldrich (St. Louis, MO, USA); Chitosan (degree of deacetylation 85% and MW 1.9×10^5 Da). Glutaraldehyde (Grade I, 50% in H_2O). Ham's nutrient mixture F12, fetal bovine serum, 96-well Corning® Costar®, Thiazolyl Blue Tetrazolium Bromide, dimethyl sulfoxide, sodium bicarbonate, pyromellitic dianhydride (PMDA), 4,4'-oxydianiline (ODA), chitosan, glutaraldehyde solution (50 w/v), agar plates, anhydrous *N,N*-dimethylacetamide (DMAC), Corning centrifuge tubes, Mueller Hinton broth, sodium phosphate monobasic, hexan, dichlorometane, acetone, ethanol, dimethylformamide, tetrahydrofurane, Mueller Hinton agar, sodium hydroxide, hydrochloric acid and sodium acetate. *E. coli* ATCC® 25922™ *Citrobacter freundii* ATCC® 8090 and *Staphylococcus epidermidis* ATCC® 12228™ were purchased from American Type Culture Collection (ATCC) (Manassas, VA, USA). Dimethyl sulfoxide ($DMSO-d_6$) was purchased from Cambridge Isotope Laboratories (Andover, MA USA). PBS buffer was used as 50 mM pH 7.2 if not otherwise specified. Unless otherwise stated, all solutions were prepared with triply distilled Nanopure water with resistivity of 18 M Ω .

2.2. Microbial culture

S. epidermidis ATCC® 12228™, *E. coli* ATCC® 25922™ and *C. freundii* ATCC® 8090 were cultured in Mueller–Hinton broth at room temperature for 72 h. The viable cell number was determined by conventional agar plate counting [25].

2.3. Synthesis and modification of PAA membrane

PAA membranes were synthesized via phase-inverted method [21,22] using six different concentrations of viscous PAA polymeric solutions viz: 0.20, 0.25, 0.30, 0.32, 0.35 and 0.38 M. The flow chart for membrane preparation is shown in Fig. 1.

Briefly, viscous solution of PAA was prepared by dissolving ODA in DMAC followed by the addition of PMDA and the solution was incubated overnight under darkness. Functionalization of PAA was achieved with CS by direct addition of chitosan into the ODA that was dissolved simultaneously in the DMAC. Functionalization was used to create a platform needed for absolute disinfection of water-borne pathogenic organisms. Modification with glutaraldehyde was performed by using 0.25 mg/mL glutaraldehyde solution as the non-solvent instead of water during the phase inversion process. 0.25 mg/mL glutaraldehyde-water solution served as a linker between individual PAA, CS polymers or PAA-CS heteropolymers. Based on

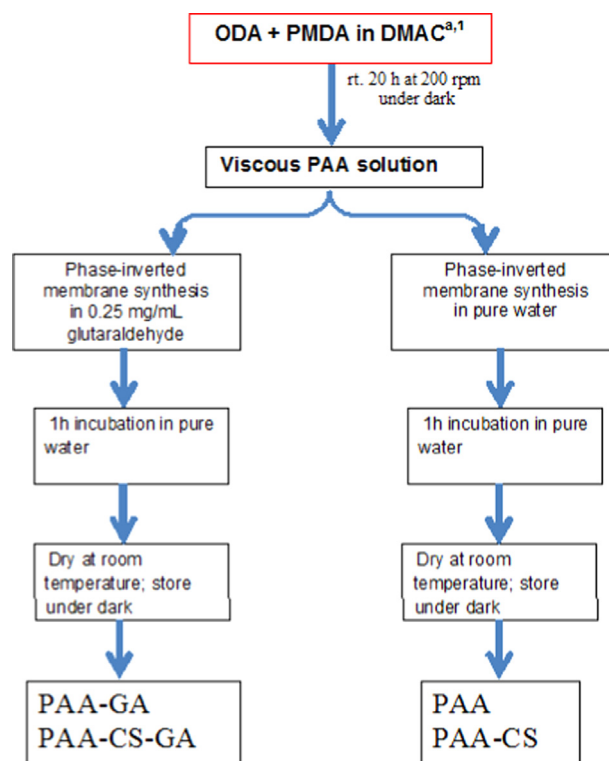


Fig. 1. Flow chart for the preparation of (A) PAA-GA and PAA-CS-GA membrane; (B): PAA and PAA-CS. ¹:implies that in the case of PAA-CS or PAA-CS-GA membrane formation CS is used with ODA + PMDA. ODA; 4,4'-oxydianiline; PMDA: pyromellitic dianhydride; DMAC: *N,N*-dimethylacetamide (a porogenic solvent suitable for forming pores and/or displacing the polymer chains during polymerization.); PAA; poly(amic)acid; GA: glutaraldehyde; CS: Chitosan.

the procedures described above, the following sets of PAA membranes were fabricated: PAA refers to poly (amic) acid membranes phase-inverted in water; PAA-CS represents co-polymers of poly (amic) acid-chitosan membrane prepared in water; PAA-GA represents poly (amic) acid cross-linked in 0.25 mg/mL glutaraldehyde solution; and PAA-CS-GA represents copolymers of poly(amic)acid-chitosan membranes prepared in 0.25 mg/mL glutaraldehyde solution.

2.4. Characterization of PAA and PAA-CS membranes

2.4.1. SEM characterization

Characterization of the PAA membrane morphology was carried out on a Zeiss Supra 55 VP field emission scanning electron microscope (SEM). The membranes were imaged both before and after filtration. All samples were coated with 5 nm gold layer for SEM imaging.

2.4.2. NMR characterization

The PAA membrane and the functionalized derivatives were dissolved in $DMSO-d_6$ and then subjected to 1H NMR, ^{13}C NMR, and 1H-correlation spectroscopy (COZY), 1H- ^{13}C -heteronuclear single quantum coherence (HSQC), 1H ^{15}N HSQC characterizations. A Bruker AM 600 spectrometer operated by Topspin™ 3.0 NMR software was used for spectra

2.4.3. FT-IR characterization

All parent and functionalized PAA membranes were characterized using FTIR via dissolution in DMSO at 5 mg/mL concentration. Same samples were also characterized using ATR/FT-IR [Perkin-Elmer

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