Journal of Membrane Science 492 (2015) 547–558

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

A simple but efficient zwitterionization method towards cellulose membrane with superior antifouling property and biocompatibility

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ARTICLE INFO

Article history:
Received 8 April 2015
Received in revised form
7 June 2015
Accepted 8 June 2015
Available online 23 June 2015

Keywords:
Cellulose membrane
Zwitterionic polymers
Surface modification
Protein adsorption
Biocompatibility

ABSTRACT

Cellulose membrane has been widely used for bioseparation and blood purification but suffers from fouling issue and insufficient biocompatibility. Surface grafting of zwitterions onto membrane surface can effectively alleviate this issue. Current grafting methods are mostly based on free radical polymerizations and have limitations of multi-step process, the requirement of inert atmosphere and inevitable flux decline. Herein we put forward a simple method for cellulose membrane (CM) zwitterionization. The modification was carried out via a one-step method based on the alkoxysilane condensation reaction using three silane coupling agents including (3-carboxypropylbetaine-propyl)-trimethoxysilane (CPPT), (3-sulfopropylbetaine-propyl)-trimethoxysilane (SPPT), and (3-sulfobutylbetaine-propyl)-trimethoxysilane (SBPT). Membrane surfaces were characterized by ATR-FTIR, XPS, SEM, water contact angle (WCA) and streaming potential. Membrane mechanical performance, pore size and its distributions were measured. The antifouling property, hemocompatibility and cytocompatibility of the membranes were evaluated by protein adsorption, platelet adhesion, cell attachment and MTT assay experiments. The results show that sulfobetaines and carboxybetaines were successfully grafted on the CM surface. The obtained zwitterions-grafted membranes show excellent resistance to protein adhesion, platelet adhesion and cell attachment. More importantly, the biocompatibility of the CM membrane was retained.

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1. Introduction

As a major renewable natural material, cellulose has been widely used in artificial organs, medical devices, implant materials, tissue engineering scaffold, blood-contacting devices and disposable clinical apparatus [1–3]. Cellulose membrane has also shown promise as an excellent membrane material used in bio-separation and blood purification in the last half century. However, its biocompatibility (especially hemocompatibility) is not adequate for its bio-related applications due to its nonspecific protein adsorption.

Nonspecific adsorption of proteins is a major issue for biomaterials application, because it can cause many problems, such as bacterial infection, thrombus formation as well as other undesired bio-reactions and bio-responses [4,5]. Therefore, it is important to develop materials and surfaces with superior protein resistance. Polyethylene glycol (poly (ethylene glycol)s (PEGs) or oligo(ethylene glycol)s (OEGs) are widely studied as a class of polymers to reject non-specific protein adsorption on membrane surface [6,7]. PEG can effectively reduce protein adsorption and platelet adhesion due to (1) its strong binding of water through hydrogen bond forming a stable hydration layer, which can block the interaction between the surface and proteins [8]; (2) high mobility of the polymer chains enabling steric repulsion of the biomolecules [9]; (3) its uncharged nature excluding electrostatic attraction while maintaining normal conformation of biomacromolecules [10–13]. However, it has been found that when the temperature reaches above 308 K, the membrane modified with PEG is easy to lose its resistance to protein adsorption [14]. More importantly, PEG is susceptible to chain cleavage and oxidative degradation in aqueous systems, especially in the presence of the transition metal ions [15,16].

In the last decade, zwitterionic polymers have shown great promise as a novel and alternative anti-fouling material. According to IUPAC, a zwitterionic polymer is “an ampholyfic polymer in which ionic groups of opposite sign are incorporated into the same pendant group” [17,18]. Several studies indicated that zwitterionic surfaces had comparable protein repellency to PEG/OEG coated

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http://dx.doi.org/10.1016/j.memsci.2015.06.024
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ones [19,20]. In addition, zwitterionic polymers show better long-term stability and higher efficacy on thrombosis resistance [21]. Typical zwitterionic polymers have phosphobetaines, sulfobetaines and carboxybetaines. In this regard, early research had been focused on phosphobetaine. However, their applications are limited because the phosphoester group is easy to hydrolyze and the monomer is difficult to synthesize [22,23]. In recent years, polysulfobetaines (PSB) and polycarboxybetaines (PCB) have attracted increasing attention due to their similar biocompatibility to phosphobetaine and much more cost-effective synthetic methods.

Various methods have been exploited to introduce zwitterions onto membrane surface. The main strategies include blending, surface coating and surface grafting [22,24–27]. Surface grafting is mostly based on controlled free radical polymerizations and is believed to render the most stable and efficient modification in a well-defined manner. The Chen group grafted poly(p-vinylbenzyl sulfobetaine) from cellulose membrane (CM) surface using surface-initiated atom transfer radical polymerization (SI-ATRP) for blood compatibility improvement [28]. Then they compared the protein adsorption and platelet adhesion of three cellulose membranes grafted with different zwitterionic polymers using the same method [29]. The Yuan group grafted carboxybetaine brush from CM surface via Activator Regenerated by Electron Transfer ATRP (ARGET-ATRP) for blood compatibility improvement. Then they grafted zwitterionic polysulfobetaine brushes onto cellulose membrane (CM) via surface-initiated reversible addition–fragmentation chain-transfer (SI-RAFT) polymerization for improving hemocompatibility and antibiofouling property [30,31]. Despite the high efficiency modifying membrane surface, these methods suffer from limits of (1) multistep process with prior anchoring of initiator groups as the first step; (2) required elimination of oxygen by the polymerization; and (3) undesired flux decline.

In this study, we aim to develop a very simple method for cellulose membrane surface zwitterionization via a one-step operation based on alkoxysilane polycondensation. The reaction mechanism is show in Scheme 1. Three alkoxysilane coupling agents with pendant zwitterions, (3-carboxypropylbetaine-propyl)-trimethoxysilane (CPPT), (3-sulfopropylbetaine-propyl)-trimethoxysilane (SPPT), and (3-sulfobutylbetaine-propyl)-trimethoxysilane (SBPT), were synthesized and grafted onto the CM surface. Surface compositions were characterized by attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR), X-ray photoelectron (XPS). Membrane surface morphology was observed by scanning electron microscopy (SEM). Water contact angle (WCA) and permeation measurements were conducted to investigate the effects of the surface zwitterionization on surface wettability and membrane permeability. The resistance to nonspecific protein adsorption, the blood compatibility and the cytocompatibility of the CM and modified membranes were compared.

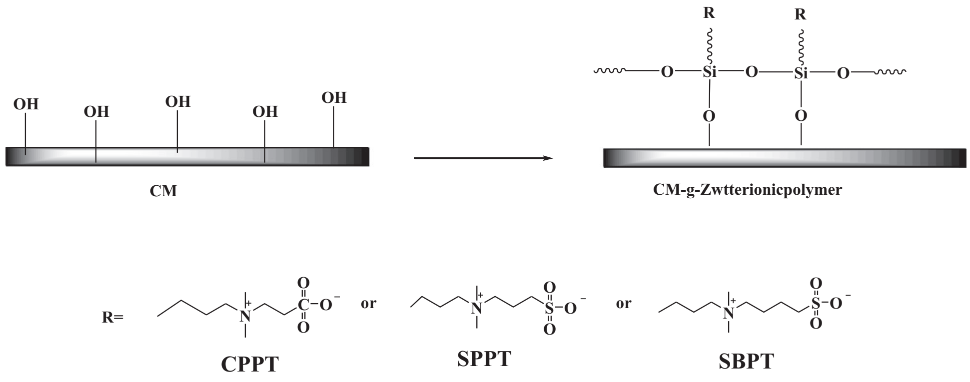
2. Experimental section

2.1. Materials

Cellulose membrane with an average pore diameter of 0.45 μm was purchased from Sartorius Stadium Biotech, Germany. (N, N-dimethyl-3-aminopropyl) trimethoxysilane ($\geq 97\%$) was obtained from Aladdin, China. 1, 3-Propanesultone (99%), 1, 4-Butanesultone (99%) and β-Propiolactone (98%) were purchased from J&K Chemical Co. (China). Acetone was distilled with CaH_2 before use. All the other chemicals were used as received without further purification. Albumin Fraction V (BSA) was obtained from Sigma-Aldrich. Bovine fibrinogen (BFG) and lysozyme were purchased from J&K Chemical Co. (China). Phosphate-buffered saline solution (PBS, pH 7.4) was prepared with 8.0 g NaCl, 0.2 g KCl, 1.44 g Na_2HPO_4, and 0.24 g KH_2PO_4 in 1 L of water and was used for the BSA, BFG and lysozyme adsorption measurements. Ultrapure water (18.2 MΩ cm at 298 K, 1.2 μg/L TOC) was obtained from a Millipore Milli-Q Advantage A10 water purification system (Billerica, MA, USA).

2.2. Synthesis of alkoxysilane coupling agents

(3-Carboxypropylbetaine-propyl)-trimethoxysilane (CPPT) was synthesized using a method similar to the literature [32]. β-Propiolactone (1.8 g, 25 mmol) was dissolved in 20 mL of anhydrous acetone and was added dropwise to a solution of (N,N-dimethyl-3-aminopropyl)-trimethoxysilane (5.175 g, 25 mmol) dissolved in 50 mL anhydrous acetone. The reaction mixture was stirred under nitrogen protection at 288 K for 3 h. The white precipitate was rinsed with 50 mL of anhydrous acetone and 100 mL of anhydrous ether and was dried under reduced pressure. The final product was stored at 275-281 K before polymerization. ^1H NMR (DMSO-6D, 400 MHz): δ 0.5–0.6 (t, 2H, Si–CH_2), 1.5–2.0 (t, 2H, C–CH_2–C), 2.5–3.0 (m, 2H, C–CH_2–N), 3.2–3.5 (m, 2H, N–CH_2–C), 3.0 (s, 6H, CH_3–N–CH_3), 2.5–2.6 (t, 2H, CH_2–COO–), 3.0–3.5 (s, 9H, CH_3–O–Si). (3-Sulfopropylbetaine-propyl)-trimethoxysilane (SPPT) was synthesized using a method similar to the literature [33]. Anhydrous acetone was added into a 50 mL of flask, after which 7.5 mL of (N, N-dimethyl-3-aminopropyl) trimethoxysilane and 4.55 g of 1, 3-propanesultone were dissolved in 37 mL of anhydrous acetone. Then the mixture was stirred for 12 h at 298 K. The crude product was rinsed with acetone for three times followed by drying under vacuum at 303 K. The monomer was stored at 298 K before polymerization. ^1HNMR (DMSO-6D, 400 MHz): δ 0.4–0.6 (t, 2H, O–Si–CH_2), 1.6–1.8 (m, 2H, C–CH_2–C–Si), 1.9–2.0 (m, 2H, S–C–CH_2–C), 2.4–2.5 (t, 2H, C–CH_2–N), 3.0 (S, 6H, CH_3–N–CH_3), 3.1–3.3 (m, 2H, N–CH_2–C), 3.3–3.4 (m, 2H, C–CH_2–S), 3.5 (s, 9H, CH_3–O–Si). (3-Sulfobutylbetaine-propyl)-trimethoxysilane (SBPT) was



Scheme 1. Schematic illustration of grafting zwitterionic polysiloxane on CM surface.

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