



High flux and antifouling filtration membrane based on non-woven fabric with chitosan coating for membrane bioreactors

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ABSTRACT

To prepare a high flux and antifouling filtration membrane used for submerged membrane bioreactors, non-woven fabric (NWF) was modified by coating chitosan (CS) on both internal and outer surface. Chemical structural and morphological changes were characterized. The changes of surface free energy were monitored by dynamic contact angle, which showed an increase after modification. The CS/NWF composite membranes were found to be with high flux, high effluent quality and excellent antifouling property. The results of fouling resistance distribution indicated that irreversible fouling resistance was decreased by coating CS. Especially, there were fewer gel layers existing on the outer surface. The adsorption of EPS on the NWF membrane internal surface decreased after being coated with CS. Modification improved filtration performance, and made fouling less troublesome and membrane regeneration efficient.

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1. Introduction

As a promising technology used for municipal and industrial wastewater treatment, submerged membrane bioreactors (SMBRs) have been actively used in the past decades (González et al., 2008). Owing to the effective biomass-effluent separation by membrane filtration, SMBRs are expected to lead the next generation of biological wastewater technologies. However, there are still many problems which need understanding such as high operation costs resulting from low flux of the membranes and excessive aeration strength required for minimizing membrane fouling (Meng et al., 2008). Thus, it is highly desirable to develop membranes with high flux and better performance for fouling control in SMBRs applications.

In the non-woven fabric (NWF) industry, one of the fastest growing segments is in filtration applications. Traditionally, wet-laid, melt-blown and spun-bonded non-woven articles, containing micron-size fibers, are most popular for these applications because of the low cost, easy processibility and good filtration efficiency (Mayer and Warren, 1998). In liquid filtration, NWFs have been used mostly as substrates to support porous membranes for ultra-filtration or nanofiltration (Zeman and Zydney, 1996). Recently, attempts have been made to use the NWF as a filtration membrane in

SMBRs (Seo et al., 2002; Chang et al., 2006). However, NWF membrane is relatively hydrophobic, and the potential application in aqueous fluid is limited due to the severe fouling (Meng et al., 2005).

Membrane fouling occurs more seriously on hydrophobic membranes than hydrophilic ones because of hydrophobic interaction between microbial cells, proteins and membrane materials (Huisman et al., 2000). As a result, much attention has been made to reduce membrane fouling by modifying hydrophobic membranes to relative hydrophilic ones (Korikov et al., 2006). Coating porous membrane with hydrophilic and functional polymer can enhance the performance of the membrane filtration (Shubhangi and Manohar, 2005) and thus it could be a suitable candidate to overcome its shortcomings. Yoon et al. (2006) has fabricated a high flux ultrafiltration membranes based on electrospun nanofibrous PAN scaffolds and chitosan coating. Zhang et al. (2008) has studied the permeability and antifouling property of the composite NWF membrane which was dip-coating by polyvinyl alcohol. Recently, we have proposed different methods for functionalization of NWF membrane by polyvinyl alcohol/4-vinylpyridine copolymer to prepare antifouling membranes (Wang et al., 2009). Chitosan (CS) is an N-deacetylated product of chitin and is the second most abundant natural polysaccharide in the ecosphere after cellulose. The seafood industry waste product CS has good physical and biological properties. CS molecules contain a large number of reactive hydroxyl (–OH) and amine (–NH₂) groups. It is suitable

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for being fabricated into membranes from aqueous acid solutions. Zeng and Fang (2004) have prepared microporous chitosan membranes by selective dissolution of blend. The results showed that highly porous structure was induced in the case of chitosan/polyethylene glycol because of their poor compatibility and multiphase structure. Clasen et al. (2006) has synthesized and characterized the hydrophilic polymer membranes based on macromolecular chitosan networks. The structure of the membrane has been altered in several ways during the formation to adjust the properties, particularly with regard to the elasticity, tensile strength, permeability and surface structure. But there was few researchers have investigated the antifouling property of chitosan membrane. It will be interesting that modified NWF membranes by coating CS to enhance their antifouling properties.

Fane et al. (1983) has concluded that although the adsorption was largely on the top surface and the amount of protein adsorbed in the pores could be a fraction of the total quantity adsorbed, it could have a significant effect on membrane performance. So, if the hydrophilicity of the internal pores of membrane is increased as well as that of the external surface through suitable treatment, it is expected that the repression of macromolecular adsorption in the pores could improve antifouling performance and increase in separation efficiency and membrane permeation stability (Li et al., 2000). However, most methods for increasing the hydrophilicity of membranes focused on the formation of the hydrophilic external surface and ultra-thin skin layer of the membranes instead of improvement of antifouling properties of the internal pores of membrane (Zhang et al., 2008; Yoon et al., 2006). Furthermore, it is also apparent that the relatively weak mechanical stability of coating layer in aqueous conditions could limit the long-term operation. An internal and outer surface coating method with the glutaraldehyde crosslink is expected to overcome the problem.

In this work, CS coating combined with glutaraldehyde crosslink in both outer and internal surface was developed and this membrane was used in a SMBR for wastewater treatment. The performances of the composite membrane were measured to determine the antifouling properties.

2. Methods

2.1. Materials

NWF membranes (0.5 μm of the pore size) were supplied by Meida Co. Ltd., Tianjin, China. CS (average molecular weight was about 150 kDa and the degree of deacetylation was 95%) was purchased from Dalian Xindie Co. Ltd., China. Glutaraldehyde (GA) was purchased from Shanghai Chemical Reagent Co., Shanghai, China. The water used in the following experiments was deionized water. Ethanol, acetic acid and sodium hydroxide are all analytical reagents and used without further purification.

2.2. Membrane preparation

NWF membrane discs were shaken in ethanol for 24 h at room temperature, to wash the impurities on the membrane surface. After that, the membranes were treated with 25 wt.% GA for 6 h at 60 °C to activate it. (The oligomers of GA were adsorbed on the NWF surface after treatment.) Afterwards, the membranes were washed three times with water. CS was dissolved into a 2% acetic acid liquid at room temperature to form a 0.2–1 wt.% solution. GA (crosslinking agent) solution was added at a constant concentration to a given CS solution under stirring. The CS/NWF composite membranes were prepared by filtering 35 mL CS solution through the NWF membranes at 40 kPa using a stirred cell, and the membrane outer surface area was 19.6 cm^2 . Then the

membranes were dried in an oven at 60 °C for 2 h. This CS/NWF composite membrane was then immersed into NaOH (2 wt.%) for 30 min for neutralization, and washed with deionized water.

2.3. Filtration and antifouling properties measurements

2.3.1. Permeation properties

The permeability of the membranes was examined using a permeation cell under constant pressure. The supernatant of SMBR was used as a model solution. A round-shaped membrane was installed into the cell and the pressure in the cell was maintained at 40 kPa with nitrogen gas. The determination of flux was depend on the permeate volume within identical filtration time. The pure water was forced to permeate through the membrane, and the flux was recorded (J_w). After that, supernatant of MBR was forced to permeate through the membrane at the same pressure and the flux was recorded as J_p . To confirm the water flux recovery property of these membranes, pure water flux was measured (J_R) after cleaning by vibrating in a 0.1 M NaOH solution for 2 h and washing with pure water three times. The relative flux reduction (RFR) and the flux recovery ratio (FRR) were calculated as follows:

$$\text{RFR (\%)} = \left(1 - \frac{J_p}{J_w}\right) \times 100, \quad (1)$$

$$\text{FRR (\%)} = \left(\frac{J_R}{J_w}\right) \times 100. \quad (2)$$

2.3.2. Antifouling evaluation in a SMBR

The laboratory-scale SMBR system was designed to characterize the filtration and antifouling properties of original NWF and CS/NWF composite membranes. It consisted of three flat membrane modules which were placed in the bioreactor with a working volume of 12 L. The modules were made from original NWF, 0.5 and 1.0 wt.% CS/NWF composite membranes, respectively. The surface area of each membrane module was 0.05 m^2 . Initially, the SMBR was filled with activated sludge sampled from the local wastewater treatment plant. Mixed liquor suspended solids (MLSS) concentration was maintained at 5 ± 0.2 g/L. The aeration through a tubular sparger was located at the bottom of the bioreactor in order to create sufficient cross-flow velocities on the membrane surface and to ensure aerobic conditions throughout the experiment, with the air flow rate at 80 L/h. A synthetic wastewater was used in the experiments, and the chemical oxygen demand (COD) concentration was 300 mg/L. The synthetic wastewater was fed to the reactor through elevated water tank. A balance tank was used to control feed flow. Treated water was pumping through the membrane by a peristaltic pump under a constant flux 20 L/m^2 h, and the effluent was intermittent with 3 min operation and 1 min stop. Concurrently, suspended solid (SS), turbidity and COD levels were measured.

The fouled membrane modules were taken out from the bioreactor at the end of each operation cycle (7–15 days). When the transfer membrane pressure (TMP) reached 40 kPa, the deionized water flux was measured at 40 kPa. After that the membranes were washed with tap-water, the deionized water flux was measured at 40 kPa also. For microfiltration at a constant TMP, the resistances for a new membrane make R_m (m^{-1}) measured by filtration of pure water, R_t is total membrane resistance (m^{-1}) after operation in the SMBR, R_c is cake layer fouling resistance (m^{-1}) due to reversible cake formation (which can be washed off by water from the membrane surface) and R_g is gel layer fouling resistance (m^{-1}) due to irreversible adsorption and pore blocking (which could be washed off by chemicals mostly). The calculate method of resistances were described by Chae et al. (2006). With the proceeding of microfiltration operation, pore blocking and cake formation will cause R_g and

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