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Cross-linked graphene oxide membrane having high ion selectivity and antibacterial activity prepared using tannic acid-functionalized graphene oxide and polyethyleneimine

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ABSTRACT

A covalently cross-linked graphene oxide (GO) membrane was prepared by a facile vacuum filtration method followed by a cross-linking process using tannic acid-functionalized GO (TA-GO) and hyperbranched polyethyleneimine (PEI). The TA-GO was prepared by a simple and environmentally friendly method using GO and a renewable plant-based polyphenol, tannic acid (TA), and it was covalently crosslinked by PEI via the reaction between catechol groups in TA and amine groups in PEI to form covalent bonds in a weak alkaline environment producing the cross-linked GO membrane. The cross-linked GO membrane showed excellent dimensional stability and ions separation performance. In particular, the cross-linked GO membrane containing TA showed excellent antibacterial activity against Escherichia coli (E. coli) due to the intrinsic bactericidal properties of the TA moieties. Furthermore, the membrane exhibited higher water flux and rejections toward NaCl and MgSO₄ than the commercial nanofiltration (NF) membranes.

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1. Introduction

Globally, the growing scarcity of fresh water is the risk and challenge faced by the human race [1]. Accordingly, new advanced water treatment technologies have been investigated over the past few decades [2]. In particular, membrane technologies are one of the most important and effective strategies for practical applications because membrane-based water separation processes are highly efficient, cost-effective, energy-saving, and environmentally sustainable [3]. Nevertheless, there are still inevitable technical limitations in using the traditional membranes in the real-world water treatment applications such as chlorine resistance, fouling resistance, productivity, and energy efficiency [4–7].

Recently, graphene derivatives have attracted intensive interests as a new class of membrane materials in the field of water separation applications due to their outstanding physical properties, chemical stability, and unique two-dimensional structures [8-16]. In particular, graphene oxide (GO) has been suggested as a very promising candidate for the membrane materials because it

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http://dx.doi.org/10.1016/j.memsci.2016.08.067 0376-7388/© 2016 Elsevier B.V. All rights reserved. can be stacked layer by layer, forming two-dimensional nanochannels between GO sheets that can be used as pores for highly selective molecular and ionic transport of GO-based membranes for gas and liquid separations. Furthermore, GO can be readily modified with various kinds of organic molecules using the oxygen functional groups on its edge and basal planes and such surface modification can enhance the important membrane characteristics such as selectivity, antibacterial activity, and antifouling performances [17-19]. Still the practical application of the GO-based membranes in the real-world water treatment applications have been limited due to the poor structural stability of the membrane in an aqueous environment [20,21]. GO membranes are readily disintegrated by the hydration and the electrostatic repulsion between the GO sheets. Therefore, it is very much desirable to impart the physical stability into the GO-based membranes to be used as the highperformance separation membranes under the aqueous conditions. Currently, it has been reported that stable GO membranes under the aqueous conditions could be prepared by the introduction of various interactive forces, including covalent bonds, between the GO sheets [19,20,22-24].

In this work, a cross-linked GO membrane was prepared using a simple vacuum-assisted filtration followed by cross-linking of





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Fig. 1. Schematic diagram of the preparation of TA-GO.

tannic acid-functionalized GO (TA-GO) prepared by the reaction of tannic acid (TA) and GO. TA was intentionally used to modify the surface of GO because it can form a TA layer on various substrates by oxidative coupling polymerization [25]. Moreover, the catechol groups in TA can react with amine groups by Michael addition and/or Schiff base reaction under basic conditions to form crosslinked layers having improved dimensional stability [25]. Therefore, the resulting cross-linked GO membrane prepared with TA-GO and polyethyleneimine (PEI) showed excellent dimensional stability in an aqueous environment. Furthermore, it also showed highly selective ion transport property and antibacterial activity due to the heavy metal binding ability and antibacterial activity of TA on the surface of the membrane [26,27]. We believe this to be the first report of the GO membrane having the unique properties including stability, hydrophilicity, selective ion penetration property, and antibacterial activity, which could be mediated by the TA-modified surface structures. The synthesis of GO functionalized with TA and the characterization of the cross-linked GO membrane are discussed in this manuscript.

2. Experimental

2.1. Materials

Graphite (Graphite UF 99.5) was obtained from BASF Korea. Tannic acid (TA), polyethyleneimine (PEI) with average M_n =60,000, phosphate buffer saline (PBS, pH 7.4), and bovine serum albumin (BSA, Mw=67 kDa) were purchased from Aldrich. *Escherichia coli* (*E. coli*; ATCC 8739) was received from the American Type Culture Collection (ATCC). DifcoTM Nutrient Broth and BactoTM Agar were supplied from Becton, Dickinson and Company (BD). All other chemicals and solvents were used without further purification.

2.2. Preparation of tannic acid-functionalized graphene oxide (TA-GO)

Graphene oxide (GO) was prepared from graphite using the modified Hummers method [28]. Tannic acid-functionalized graphene oxide (TA-GO) was prepared by the reaction of TA and GO in

a basic buffer solution following the process described in detail in our previous paper [25].

2.3. Preparation of cross-linked TA-GO membrane

5 mg of TA-GO was added to 50 mL of PBS (pH 7.0), followed by ultrasonication for 30 min to produce TA-GO dispersion (0.1 mg mL⁻¹), and 5 mg of PEI was also dissolved in 5 mL of PBS to produce a PEI solution. Then, the TA-GO dispersion and the PEI solution were mixed and sonicated. The mixture of TA-GO and PEI was vacuum-filtered through a cellulose acetate (CA) membrane with pore size of 0.2 μ m to form a PEI intercalated TA-GO layer. To prepare a cross-linked TA-GO membrane, 50 mL of 0.1 M Trizma-buffer solution (pH = 8.5) was vacuum-filtered through the PEI intercalated TA-GO layer. The resulting membrane was washed with DI water several times to remove any unreacted PEI and remaining buffer solution, and then dried at room temperature. For comparison, a GO membrane was prepared as a control using GO instead of TA-GO with the same vacuum filtration method.

2.4. Ion penetration tests

Ion penetration tests were performed using a homemade U-type tube divided into feed and permeate compartments by the membrane [24]. For penetration tests of a single salt solution, the feed compartment was filled with aqueous KCl or K_4 Fe(CN)₆ solution (0.05 or 0.20 mol L⁻¹). For penetration tests of mixed salts solutions, the feed compartment was filled with mixtures of aqueous MgCl₂, KCl, CaCl₂ and NiCl₂ solutions, so that each component had the same concentration (0.05, 0.10 or 0.20 mol L⁻¹). The permeate compartment was filled with only pure water. The concentration of the permeate compartment was measured by inductively coupled plasma atomic emission spectrometer (ICP-AES; Optima 8300, Perkin Elmer).

2.5. Membrane filtration experiments

Filtration experiments were performed using a dead-end filtration cell with an effective filtration area of 14.6 cm². All experiments were carried out at room temperature and the feed side was stirred at an agitation speed of 200 rpm and pressed by Download English Version:

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