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# Electrically conducting nanofiltration membranes based on networked cellulose and carbon nanostructures

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#### HIGHLIGHTS

· Electrically conductive membranes have been fabricated using networked cellulose and carbon nanostructures

· Networked cellulose helps control the pore size and hydrophilicity, allowing the membrane to reject divalent salts

· Electrocatalytic activity of the membrane is promising for potential use as self-cleaning

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#### ABSTRACT

Electrically enhanced fouling control is increasingly applied to membrane-based separation and requires conducting membranes with controlled properties. In this work, electrically conductive membranes based on networked cellulose (NC) and carbon nanostructures (CNS) were fabricated via vacuum filtration, followed by drying at 40 °C. The morphology, structure, mechanical and electrochemical properties of these NC-CNS membranes were characterized and compared with CNS membranes. The effect of incorporating NC on the electrocatalytic activity has been analyzed. It is found that networked cellulose helps to decrease the contact angle of water from 105° to 73°. It is also found that the improved surface hydrophilicity of CNS-NC membrane assists the regeneration of electrode surface during electrolysis process. Networked cellulose yields a more dense structure with the tensile strength exceeding ten times that of CNS alone. The compaction of pore structure via incorporation of NC translates into promising results with respect to nanofiltration of divalent ions, with a rejection efficiency of 60% for MgSO<sub>4</sub> and 47% for CaCl<sub>2</sub>, while maintaining a high flux  $\geq 100 \text{ Lm}^{-2} \text{ h}^{-1}$ , making them suitable for pretreatment of RO feeds.

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#### 1. Introduction

Membrane based water treatment plays a crucial role in obtaining potable water through reclamation of wastewater and seawater desalination. Membranes offer several advantages over other processes, as they are simple, do not require chemical additives and can be easily scaled up. Despite the rapid rise in the use of membrane processes, the most widely used polymeric membranes are susceptible to fouling, which increases energy costs for cleaning the membrane and often requires replacement as frequent cleaning deteriorate membrane quality with time. Recently, impregnation of electrically conductive material such as carbon nanotubes (CNTs) in polymeric membranes has led to the development of electrically enhanced fouling control through oxidation of foulants [1,2] as well as gas bubble generation [3,4]. CNTbased conductive membranes prevent fouling through several possible

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http://dx.doi.org/10.1016/j.desal.2016.09.005 0011-9164/© 2016 Elsevier B.V. All rights reserved. mechanisms including electrophoretic transport of foulants, direct oxidation, bubble generation, inherent antimicrobial properties of CNTs as well as bacterial detachment due to cathodic current [5]. This relatively new approach allows the non-destructive, energy efficient control of membrane fouling. However, the challenge is to develop membrane materials that are electrically conductive and electrocatalytically active without compromising their flux and selectivity.

Although electrically conductive membranes have been fabricated for simple filtration, only a few studies have focused on developing conductive membranes for pressure-driven processes such as ultrafiltration, nanofiltration and reverse osmosis [6–9]. These materials rely on CNT-polymer composites to maintain the selectivity and flux of polymeric membranes while enhancing their conductivity with CNTs. Despite progress in NF membrane materials, fouling mitigation to recover flux still largely relies on chemical cleaning techniques. However, common chemical cleaning agents such as caustic acid have been shown to alter NF membrane surface properties, and in turn their separation efficiency [10]. Periodic electrolysis has previously been applied

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Fig. 1. Membrane filtration unit.

for successful flux recovery in carbon nanostructure membranes, through cathodic hydrogen bubble generation [4]. The aim of the current study is to attempt to control the pore size and surface Table 1

Porosity of membranes with and without NC.

Membrane	Thickness (µm)	Porosity
CNS NC/CNS	120 80	$\begin{array}{c} 45\pm1\\ 5\pm1\end{array}$

characteristics of such conductive membranes to be suited to the separation of multivalent salts, in order to allow electrolytic cleaning to be used with NF.

Cellulose and cellulose-based derivatives are widely used as membrane materials for various separation processes. Cellulose is the world's most abundant natural raw material with several attractive properties, namely biodegradability, biocompatibility and excellent chemical stability. Through controlled dissolution in sulfuric acid followed by regeneration, native cellulose can be processed into a gel-like suspension known as networked cellulose (NC) [11]. NC has a more amorphous and more accessible structure than native cellulose and has previously been used to control the swelling of polyvinyl alcohol (PVA) RO membranes [12].

Carbon Nanostructures (CNS) consist of entangled and covalently bonded CNTs that are made through a scalable and low cost process by Applied Nanostructured Solutions (ANS), a subsidiary of Lockheed Martin (USA). CNS are structures of highly entangled CNTs that may share common walls. They are characterized by improved processability, high electrical conductivity and large surface area [13]. In our previous work, CNS were used to fabricate microfiltration membranes that worked successfully as electrically conductive self-cleaning membranes [4].

In this work, self-supporting NC/CNS membranes in which NC serves to enhance the mechanical properties and compact the pore structure while retaining high electrical conductivity have been developed. The novel material is characterized for morphology, structure and mechanical properties. The performance of this novel membrane material is



Fig. 2. SEM images of a) CNS membrane, b) NC/CNS membrane and c) NC/CNS at high magnification.

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