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Fabrication of antibacterial mixed matrix nanocomposite membranes using hybrid nanostructure of silver coated multi-walled carbon nanotubes

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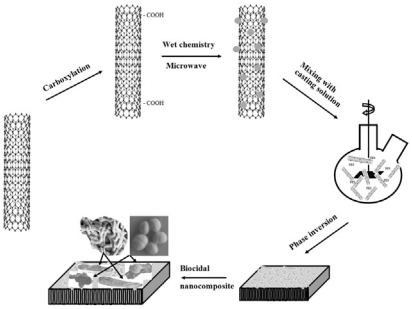
HIGHLIGHTS

- Novel, rapid and facile method for decorating MWCNTs surface with silver nanoparticles.
- Physical deposition of Ag NPs on MWCNTs via microwave treatment.
- Self-cleaning/antimicrobial nanocomposite membranes for wastewater and desalination applications.
- Long term antibacterial activity for polymeric membranes blended with Ag-MWCNTs.
- Role of Ag-NPs size on the bacteriostatic activity of nanocomposite membranes against *E. coli* and *St. aureus*.

GRAPHICAL ABSTRACT



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ABSTRACT

The function of separation membranes can be significantly improved by the integration of nanoparticles that can improve not only the mechanical properties of the membrane but also reduce the propensity of the surface to foul. The research of the paper presents the development of a novel nanocomposite membrane incorporating antimicrobial nanoparticles which have the potential to lower membrane biofouling; a major problem in many industries that exploit membrane technology. Antibacterial hybrid nanostructures (HNS) comprising of multi-walled carbon nanotubes (MWCNTs) coated with silver nanoparticles (AgNPs) were successfully synthesized via a facile and rapid method using a microwave treatment. The HNS were incorporated into polyethersulfone (PES) ultrafiltration (UF) membranes via the classical phase inversion technique in order to assess their antimicrobial properties against two bacterial species; *E. coli* and *S. aureus*. Different techniques were used to characterize HNS powders and a number of loading weights of the HNS were blended with PES flakes to assess the resultant nanocomposite membranes. The nanocomposite membranes displayed an increase in their antibacterial activity against the two species with increasing the loading weight of HNS.

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Nomenclature

HNS	hybrid nanostructure
10HNS	hybrid nanostructure prepared with 10 wt% silver
20HNS	hybrid nanostructure prepared with 20 wt% silver
MWCNTs	multiwalled carbon nanotubes
f-MWCNTs	functionalized multiwalled carbon nanotubes
p-MWNT	pristine multiwalled carbon nanotubes
AgNPs and nAg	silver nanoparticles and nanosilver
PES	polyethersulfone
UF	ultrafiltration
FTIR	Fourier transmission infrared
XRD	X-ray diffraction
EDX	X-ray energy dispersive spectroscopy
TGA	thermal gravimetric analysis
SEM	scanning electron microscope
ZOI	zone of inhibition
CNTs	carbone nanotubes
NPs	Nanoparticles
1D-NMs	one-dimensional nanoscale materials
2D-NMs	two-dimensional nanoscale materials
0D-NPs	zero-dimensional
NWs	nanowires
Cu_2O	copper oxide
TFC	thin film composite
GO	graphene oxide
CVD	chemical vapor diposition
PBS	Phosphate Buffer Solution
MHB	Mueller Hinton Broth
PEG	polyethylene glycol
PVP	polyvinylpyrrolidone
HNO_3	nitric acid
H_2SO_4	sulphoric acid
HCl	hydrochloric acid
NMP	N-methyl-2-pyrrolidone
mM	milimolar
ζ	zeta potential
CFU	colony forming unit
PWF	pure water flux
μp	mean pore size
σp	geometric standard deviation
EC	*E. coli*
SA	*S. aureus*

1. Introduction

As a new class of membranes, nanocomposite membranes are currently attracting a tremendous amount of research activity. These membranes that merge the beneficial properties of organic polymers and nanoscale materials are expected to revolutionize current membranes, and create innovative solutions in membrane technology through providing alternative strategies to control and mitigate membrane surface fouling; one of the key problems for the application of membranes.

Biological fouling is considered as the most inherently complex form of fouling. It is defined by microbial cell adhesion and subsequent colonization at the membrane surface forming microbial biofilm. Commonly, microbial colonization of the membrane surface is initialized by irreversible adhesion of one type of bacteria or more, followed by the growth and multiplication of the surface-bound cells in the presence of feed nutrients [1]. Once the microbes attach at the surface and form a matrix of extracellular polymers, their removal will extremely arduous even with the application of biocides [2]. Thus, biofouling represents the major concern for industries that exploit membrane technology including water, food and pharmaceuticals. As an alternative to disinfectant application, nanotechnology has impacted on the design and fabrication of nanocomposite membranes with the potential for creating self-cleaning and antimicrobial surfaces.

Within the wide range of commercially available nanoscale materials, silver nanoparticles (Ag-NPs) have gained special interest as unique antimicrobial additives in a broad range of applications. This is due to their unique antimicrobial properties alongside their electrical and optical characteristics [3,4]. Even though their antimicrobial mechanisms are still under debate and not comprehensively understood [5,6], their biocidal activity is well recognized as a highly desirable capability that they bestow on a membrane surface to resist biofouling and impart self-cleaning characteristics. On this basis, a large amount of work has been devoted for developing such nanocomposites membranes [7,8]. As an important consideration when developing nanocomposite membranes for the control of biofouling, it is crucial to sustain antimicrobial activity and avoid the rapid depletion of nanosilver (nAg) that could diminish the antibacterial activity of nAg, and increase environmental issues and cost constraints. Attempts have been stepped forward to ameliorate the interactions between these metallic nanoparticles and host polymeric chain to overcome these limitations. One of the most pragmatic approaches used for this target was through revising the surface characteristics of Ag-NPs via chemical treatment [9]. Various functional groups for modification/functionalization are available to increase their stability in the host polymer e.g., phosphoric acid, carboxylic acid, dopamine and silane coupling agents [10].

Apart from that, intensive researches are focusing on integration of one-dimensional nanoscale materials (1D-NMs) or two-dimensional (2D-NMs) with zero-dimensional (0D-NPs) into one hybrid nanostructure (HNS), which found their way in countless applications such as; fuel cells, photocatalysis, electrocatalysis, solar cells, sensors, supercapacitors, batteries and hydrogen storage applications. Applying these HNS could potentially exhibit not only the unique characteristics of 1D/2D and 0D at the nanoscale level, but also may possess novel chemical and physical properties as a result of the synergic effects of both nanomaterials that might not be obtainable to their individual component alone [11]. Several HNS targeting water and wastewater treatment have been reported in the recent few years [12,13]. Parallel to this steadily evolution in HNS structures and applications. Functionalization of polymeric membrane aiming for a specific application has been evinced with these tailored nanostructures. For antibacterial nanocomposite membranes, Xu et al. blended AgNPs decorated Cu_2O nanowires (NWs) with PSF via phase inversion. To facilitate the deposition of AgNPs on the surface of Cu_2O, *L*-dopa has been grafted onto NWs surface by *in situ* polymerization to create zwitterionic surface. The nanocomposite membranes held remarkable antibacterial against *E. coli* and *S. aureus* with controlled release of Ag^+ [14]. Recently, Faria et al. conducted a research to functionalize TFC membranes with GO-Ag. Results referred to a promising inactivation rate against attached *P. aeruginosa* (up to 80%) without sacrificing the intrinsic transport properties for the modified membranes [15]. A number of HNS employed in fabrication malfunctional nanocomposite membranes have been reported in the literature for waste water treatment and desalination applications [16–19]. Another interesting 1D nanoscale carrier is carbon nanotubes (CNTs). Since its discovery in 1991, CNTs have turned into