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Nano-modification of polyamide thin film composite reverse osmosis membranes by radiation grafting

M.B. El-Arnaouty [a], A.M. Abdel Ghaffar [a], M. Eid [a], Maysara E. Aboulfotouh [a, *], N.H. Taher [a], El-Sayed Soliman [b]

[a] Polymer Chemistry Department, Radiation Industrial Division, National Center for Radiation Research and Technology, Atomic Energy Authority, P.O. Box 29, Nasr City, Cairo, Egypt
[b] Faculty of Science, Chemistry Department, Ain Shams University, Cairo, Egypt

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ABSTRACT

Radiation synthesis of reverse osmosis membranes were carried out by grafting of *N*-Isopropyl acrylamide (NIPAM) and ZnO nanoparticles incorporation onto polyamide thin film composite reverse osmosis membranes PA(TFC). The effect of monomer concentration, radiation time and concentration of ZnO nanoparticles on the grafting percent were investigated. The properties of the prepared grafted reverse osmosis membranes were characterized by using different analytical tools such as contact angles goniometer, Fourier transform IR (FTIR), X-ray diffraction (XRD), Field Emission-Scanning Electron Microscope (FESEM). The performance of the reverse osmosis process of the neat and the modified PA(TFC) membranes in terms of water flux and salt rejection (%) was investigated. The chlorine and biofouling resistance properties of the neat and the modified PA(TFC) membrane were evaluated. It is found that, the performance of the modified ZnO NPs/P(NIPAM)-g-PA(TFC) membrane is much better than the neat PA(TFC) membrane.

1. Introduction

The rapid increase demands on water resources, fresh water shortage has become an important issue affecting the economical and social development in many countries. As one of the main technologies for producing fresh water from saline water and other wastewater sources, reverse osmosis (RO) has been widely used so far. Nowadays, the most part of conventional seawater desalination plant use either RO or MSF technology. While thermal desalination is the most frequently applied technology in the Middle East, RO now surpasses thermal processes in new plant installations since RO uses less energy than MSF.

The global market for RO components is currently greater than $2.6 billion and is expected to reach $3.7 billion by 2018. RO have developed into the predominant membrane-based desalination technology mainly because it is the most energy-efficient desalination technology to date. The energy cost for the RO has reduced from 5 kWh/m^3 in the 1990s to 1.8 kWh/m^3 nowadays, which is several times lower than that for other technologies, such as thermal-based desalination methods. For membrane desalination, decreasing costs and producing superior water quality are among a number of significant reasons why this technology continues to be the water treatment technology of choice in the world.

Historically cellulose acetate (CA) polymer was developed by Loeb and Sourirajan in 1962 as the first commercially viable RO membrane which had been used widely until the commercial introduction of TFC membranes in 1981. TFC membranes are stable over a wider pH range and operable at lower pressure than CA membranes, as shown in Table 1.

Polyamide thin film composite (PA-TFC) membranes are becoming more widely used for water desalination both in industrial and experimental plants and also in reverse osmosis (RO) process due to their superior properties. However, trade-off between the permeability and the salt rejections, fouling and chlorination are seriously restricting their better operational functions. Therefore, various strategies have been explored to tackle these problems, among surface modifications by grafting and

* Corresponding author.
E-mail address: drmaysara@consultant.com (M.E. Aboulfotouh).
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Table 1
Comparison of polyamide TFC membrane with cellulose acetate (CA) membrane.

Parameters		PA Membrane	CA Membrane
Operating pH range		2~12	4~6
Operating Pressure (Kg/cm^2)		15	30
Salt Rejection(%)	TDS	99+	98
	Silica (SiO_2)	99+	<95
Salt Rejection Change after 3 years		99%→98.7%	98%→96%
Chlorine Tolerance		<0.1 ppm	1 ppm
Membrane Fouling		High	Low

nanoparticles incorporations have been identified to be the most effective ones, (Balta et al., 2012; Dihua, Xuesong, Sanchuan, Meihong, & Congjie, 2010; Freger, Gilron, & Belfer, 2002; Isawia, El-Sayeda, Xianshe, Hosam, & Mohamed, 2016; Moshe et al., 2014; Wei, Wang, Chen, Wang, & Wang, 2010a, Wei, Wang, Zhang, Wang, & Wang, 2010b; Zhe, Zhi, Jixiao, & Shichang, 2013).

Dihua et al. (2010) focused on the surface modification of the commercial aromatic polyamide (PA) thin-film composite (TFC) reverse osmosis (RO) membranes by grafting with thermo-responsive copolymers as poly(*N*-isopropyl acrylamide-*co*-acrylamide) (P(NIPAM-*co*-Am)) for improving membrane properties. Guo-Rong, Jiao-Na, and Cong-Ju (2013) provided comprehensive information and give an outlook on the surface modifications and nanoparticles incorporations, which might supply some clues to explore more advanced and innovative strategies for improving the performance of the PA-TFC RO membranes.

In this study, the grafting of *N*-isopropyl acrylamide and ZnO NPs onto polyamide thin film composite reverse osmosis membrane PA(TFC) RO has been carried out using gamma irradiation in one single step. The performance of the modified membranes has been studied in terms of water flux, salt rejection (%), the chlorine resistance and antifouling.

2. Experimental

2.1. Materials

N-isopropyl acrylamide of purity 98.9% are purchased from Sigma Aldrich (Germany). Polyamide thin film composite PA (TFC), for water desalination from DOW FILMTEC™ reverse osmosis (RO) membranes, (USA). Membrane operating limits: Membrane Type (Polyamide Thin-Film Composite). Maximum Operating Temperature: 113 °F (45 °C). Maximum Operating Pressure: 150 psig (10 bar). Maximum Feed Flow Rate: [2.0 gpm (7.6 lpm)]. pH Range Continuous Operation: (2–11). Maximum Feed Silt Density Index (SDI): (5). Free Chlorine Tolerance: (<0.1 ppm). Other chemicals used such as solvents and other reagents were reagent grade and used as received. The structure of NIPAM and PA(TFC) membrane are represented in Scheme 1.

2.2. Procedures and measurements

2.2.1. Gamma radiation sources

The samples are irradiated at the required dose using Cobalt-60 source at dose rates ranging from 1.19 to 1.23 Gy/sec. The irradiation facility was constructed by National Center for Radiation Research and Technology, Cairo, Egypt (Atomic Energy Authority). γ-Chamber 4000A°, isotope group of BHAHA, Atomic research center of INDIA which contains 10 400 Ci of Cobalt-60.

2.2.2. Graft copolymerization

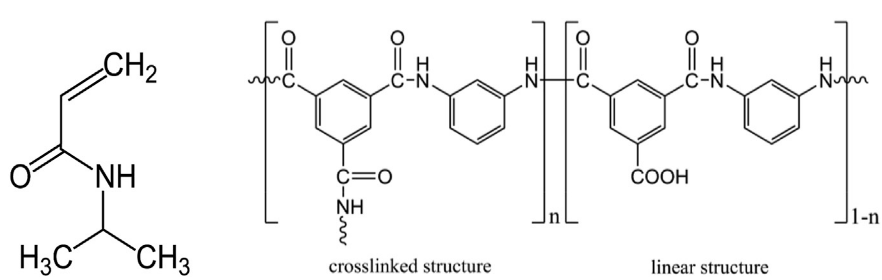
Strips of Polyamide thin film composite PA(TFC) membrane of 2 × 2 cm dimensions were washed with acetone, dried in vacuum oven at 35 °C for 1 h, weighed and then immersed in glass ampoules with the monomer solution. The direct radiation-induced grafting technique was used in which the glass ampoule containing the membrane and the monomer solution was deaerated by bubbling nitrogen gas for 5 min, and subjected to gamma irradiation at the desired dose rate. After irradiation, the modified membranes were washed thoroughly with hot distilled water to extract the residual monomer and the homopolymer occluded in the membrane. The membranes were dried in a vacuum oven at 30–40 °C for 24 h and weighed. The grafting percent (G%) is determined by the percentage increase in weight according to equation (1).

$$G\,(\%) = (Wg - W_0)\;/\;W_0 x\;100 \qquad (1)$$

Where, W_0 and Wg represent the weight of the virgin and grafted membranes, respectively.

2.2.3. FTIR spectroscopic measurements

The spectra were recorded on Nicolet spectrometer 380 (Thermo scientific, USA) using ZnSe crystal (25 × 5 × 2 nm) at a nominal incident angle of 24° yielding about 12 internal reflections at the sample. All spectra 100 scans at resolution of at least 4 cm^{-1} were recorded at 25 °C. The instrument was purged with dry nitrogen to prevent interference of atmospheric moisture with the spectra; the analysis was performed in National Institute of Standards (NIS), Cairo, Egypt.



Scheme 1. The structure of *N*-isopropyl acrylamide and polyamide thin film.

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