



Polymeric and mixed matrix polyimide membranes



Valentina Grosso^{a,b}, Danilo Vuono^b, Mohammed A. Bahattab^c, Gianluca Di Profio^a, Efrem Curcio^a, Saad Abdullah Al-Jilil^c, Fahed Alsubaie^c, Mohammed Alfife^c, Janos B.Nagy^b, Enrico Drioli^{a,b}, Enrica Fontananova^{a,*}

^a Institute on Membrane Technology of the National Research Council of Italy (ITM-CNR), at University of Calabria, Via P. Bucci, Cubo 17/C, 87036 Rende (CS), Italy

^b Department of Environmental Engineering and Chemical Engineering, University of Calabria, 87036 Rende (CS), Italy

^c King Abdulaziz City for Science and Technology (KACST), Riyadh, Saudi Arabia

ARTICLE INFO

Article history:

Received 6 May 2014

Received in revised form 13 June 2014

Accepted 14 June 2014

Available online 22 June 2014

Keywords:

Charge effect

Dyes

Multiwalled carbon nanotubes

Polyimide membranes

ABSTRACT

Three different polyimide type polymers were used to prepare porous asymmetric membranes by non-solvent induced phase separation (NIPS): a homopolymer (Matrimid) and two co-polymers (P84 and Torlon). The effect of the casting solvent on membrane morphology and transport properties was studied. Moreover, the co-polyimide P84 was selected for further investigations in the form of mixed matrix membranes (MMMs) containing functionalized multiwalled carbon nanotubes (oxidized or aminated MWCNTs). The introduction of polar functional groups was found to be fundamental in order to have a good dispersion of the MWCNTs in the casting solution before, and in the formed membrane, after NIPS, thanks to the formation of hydrogen bonds between the solvents, the MWCNTs and the polymer. The presence of the MWCNTs in the membrane matrix improved the performance in the filtration of dyes solutions in water or ethanol (enhanced permeance and reduced fouling, with similar or higher rejection), with respect to reference polymeric membranes (without MWCNTs). The separation properties of the membranes produced were also compared with literature data obtained with commercial and other laboratory made mixed matrix membranes.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

The presence of dyes in textile industrial wastewater is presently one of the most worrisome ecological problems in recent years [1]. Discharge the industrial effluents without an appropriate treatment is dangerous because it determines undesirable chemical and physical changes in the recipient stream. High concentrations of colorful pollutants reduce the light transmittance and, as a consequence, the photosynthesis and the oxygenation of the water is reduced with consequent reduction of aquatic plant growth and survival of animal species [2]. Numerous textile industrial processes require a large volume of water for treatments, like: dyeing, washing and cleaning, producing a variety of wastewaters in terms of composition and toxicity. In addition to dyes, these industrial effluents have a high content of salts and dissolved solids (organic and inorganic) [3,4]. Many of the dyes present in industrial wastewater are difficult to be eliminated in nature because of their complex structure and synthetic origin. There are several conventional methods for treating wastewater including:

biological treatment, ozonation, coagulation–flocculation, adsorption on powdered activated carbon, electrochemical processes and photocatalysis [5–12].

However, the need for more efficient and environmental friendly wastewater treatments together with the increasingly stringent discharge regulations has attracted the attention of many scientists and environmental engineers toward membrane technology for wastewater treatment [2–4]. Membrane operations have the advantage to use intrinsically more clean and energy-efficient separation routes, compared to conventional separation technologies [13].

Numerous examples of research work can be found in literature in which membrane process are used for wastewater treatment containing dyes, including: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) [14–24]. However, one of major problem in these pressure driven liquid phase membrane process is the flux decline caused by concentration polarization and fouling which induced additional resistances to the transport through the membrane.

Several strategies have been proposed to limit concentration polarization and fouling. Some examples are: increasing feed velocity, use of turbulence promoters and solution pre-treatment.

* Corresponding author. Tel.: +39 0984 492010; fax: +39 0984 402103.

E-mail address: e.fontananova@itm.cnr.it (E. Fontananova).

Nomenclature

AFM	atomic force microscopy	NMP	1-Methyl-2-Pyrrolidone
ATR	attenuated total reflectance	PA	polyamide
CNT	carbon nanotube	Pe	permeance
DMSO	dimethylsulfoxide	PES	polyethersulfone
ΔU	internal energy	PI	polyimide
$E_{cohesive}$	cohesive energy	PVA	Poly(vinyl alcohol)
FT-IR	Fourier transform infrared analysis	PVDF	polyvinylidene difluoride
J	flux	R	rejection
J_R	relative flux	RO	reverse osmosis
MD	Molecular dynamics	SEM	scanning electron microscopy
MF	microfiltration	SPESEKK	sulfonated poly(ethersulfone ether ketone ketone)
MMM	mixed matrix membrane	TEM	transmission electron microscopy
MWCNT	multiwalled carbon nanotube	THF	tetrahydrofuran
MWCNT-NH ₂	aminated multiwalled carbon nanotube	TMP	transmembrane pressure
MWCNT-OX	oxidized multiwalled carbon nanotube	UF	ultrafiltration
NF	nanofiltration	V_{molar}	molar volume
NIPS	non-solvent induced phase separation		

Moreover, the use of advanced membranes with low fouling properties, like mixed matrix membranes (MMMs) is a promising tool for fouling limitation [25]. Hybrid organic–inorganic membranes are reported in the literature to have attractive characteristic such as increased hydrophilicity, high permselectivity, fouling resistance and macrovoids-free structure [26–29]. Titanium dioxide (TiO₂) has been widely utilized to improve the permeability and antifouling properties of membranes due to the increasing membrane hydrophilicity and, in some cases, the exploitation of its photocatalytic activity as UV-activated self-cleaning membrane. In the last years is growing the interest toward the inclusion in artificial membranes of new forms of pure carbon such as carbon nanotubes (CNTs) [30].

CNTs thank to their diameter in the nanometer range and their atomically smooth surfaces, offer unique systems for fast molecular transport [31,32]. Molecular dynamics (MD) simulations showed spontaneous and continuous filling of a CNT with a one-dimensionally ordered chain of water molecules. CNTs have been entrapped in mixed matrix membranes made of various polymeric material by several techniques including: dispersion in the casting solution and successive phase separation, entrapping in the membrane pores using a polymer binder, in situ crosslinking of a polymer matrix around oriented CNTs [33]. The resulting hybrid membranes offers, in many cases, relevant advantages in comparison with the polymeric samples. Poly(vinyl alcohol) (PVA)/multiwalled carbon nanotubes (MWCNTs) membranes have been realized for pervaporation separation, obtaining significant improvement in Young's modulus and thermal stability, as compared to pure PVA membranes [34]. The entrapment of MWCNTs in polyethersulfone (PES) membranes, reduced the fouling problems in water treatment [35]. MMM consisting of sulfonated carbon nanotubes (sCNTs) and sulfonated poly(ethersulfone ether ketone ketone) (SPESEKK) were also fabricated via the solution casting method [36]. The proton conductivity of the SPESEKK membrane increased while the methanol permeability decreased as the sCNTs content increased. MWCNTs have been covalently linked to aromatic polyamide (PA) membranes by a polymer grafting process [37]. Measurements of mechanical properties of this composite showed an increased membrane mechanical strength of the MMMs. The addition of MWCNTs also improved the rejection of both salt and organic matter relative to the PA polymeric membranes [37].

MWCNTs have been also immobilized in the pores of a hydrophobic membrane improving the water-membrane interactions

to promote vapor permeability in membrane distillation process [38]. In this case The CNTs dispersion was forced under vacuum into the pores of a polypropylene (PP) membrane, using polyvinylidene difluoride (PVDF) as binder.

In this work three different polyimide (PI) polymers were used to prepared porous asymmetric membranes by NIPS: a homopolymer (Matrimid) and two co-polymers (Lenzing P84 and Torlon) (Fig. 1). PIs combine an easy processability in the form of membranes, with a high chemical and thermal stability, over a wide range of operative conditions. Membrane made of PIs, have shown excellent transport properties in both gaseous and liquid phase separations [39].

The effects of the membrane preparation conditions on the membrane morphology and transport properties, were investigated. Moreover, mixed matrix membranes based on co-polyimide P84 and functionalized multiwalled carbon nanotubes (oxidized or aminated MWCNTs), were prepared and compared with literature data obtained with commercial membranes and other laboratory made membranes containing MWCNTs.

2. Materials and methods

2.1. Materials

Co-polyimide Lenzing P84[®] was purchased from HP polymer GmbH (Austria); the co-poly(amide–imide) Torlon[®] 4000 TF was kindly supplied from Solvay Solexis (Italy); the polyimide Matrimid[®] 5218 was purchased from Ciba Specialty Chemicals (North America). 1-Methyl-2-Pyrrolidone (NMP), dimethylsulfoxide (DMSO), tetrahydrofuran (THF), Ethanol, Orange II Sodium salt (MW 350.32 g/mol), Safranin O (MW 350.45), Sudan Blue II (MW 350.82 g/mol), NaY zeolite, H₂SO₄, HNO₃, cobalt acetate tetrahydrate, iron nitrate nonahydrate, were purchased from Sigma–Aldrich. HF was purchased from Carlo Erba.

2.2. Supported catalyst preparation for MWCNTs synthesis

2.11 g of Cobalt acetate tetrahydrate and 3.62 g of iron nitrate nonahydrate were dissolved in 6.08 mL of distilled water. After manual homogenization for 5 min, the solution was treated by an ultrasound bath for 1 h. The solution was mixed with 9 g of NaY zeolite and homogenized (with a spatula for 5 min) to obtain a wet product of uniform color (red). The product was dried in an

Download English Version:

<https://daneshyari.com/en/article/641065>

Download Persian Version:

<https://daneshyari.com/article/641065>

[Daneshyari.com](https://daneshyari.com)