



Impact of graphene oxide on removal of heavy metals using mixed matrix membrane



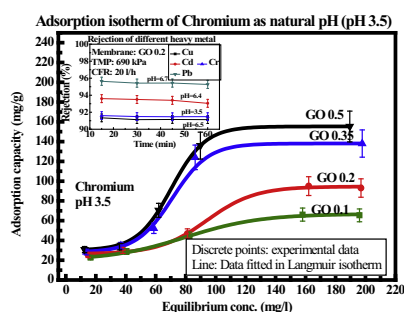
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HIGHLIGHTS

- Membrane became more hydrophilic and porous with addition of GO.
- Above 90% rejection of Cr, Cd, Pb and Cu was effected by MMM.
- Membrane performed better in basic pH, low pressure and flow rate.
- In-situ membrane regeneration was possible by acid treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

The graphene oxide (GO) impregnated mixed matrix membrane (MMM) was prepared by non-solvent induced phase inversion method. MMM was characterized in terms of molecular weight cut-off, porosity, permeability, average pore size, pore size distribution, contact angle, zeta potential and mechanical strength. The membrane became highly permeable, hydrophilic and charged on addition of GO. The surface morphology and roughness were studied through scanning electron and atomic force microscopy. Efficacy of MMM for removal of heavy metals was explored using the developed MMM. It showed high adsorption capacity for Pb^{2+} (79 mg/g), Cu^{2+} (75 mg/g), Cd^{2+} (68 mg/g) and Cr^{6+} (154 mg/g) at natural pH, 6.7, 6.5, 6.4 and 3.5, respectively. The steady state permeate flux was around $30\text{ l/m}^2\text{ h}$ for 50 mg/l feed concentration at 414 kPa transmembrane pressure drop and 40 l/h cross flow rate. Rejection of Pb, Cu, Cd and Cr were in between 90% and 96% for various operating conditions and the breakthrough time was around 10 h for different metal ions. The membrane was regenerated in-situ, by acidic solution at pH 5.5.

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1. Introduction

Graphene has been proposed as the “next generation material” owing to its remarkable electronic, optical, and thermal properties, chemical and mechanical stability, and large surface area [1]. Chemical structure of graphene oxide (GO) is reported as oxidized graphene, decorated with various oxygenated functionalities such as hydroxy, epoxy on the basal plane and carbonyl, carboxylic acid

at the edges [2]. These functionalities make the GO more versatile in application oriented research. Through the sharing of the lone electron pair on oxygen, it can efficiently bind the metal ion to form metal complex [2], thereby, rendering it an effective adsorbent for removal of heavy metal ions [3]. There are some recent reports demonstrating high adsorption ability of GO for Cd(II) [2], Pb(II) [3] and Cu(II) [4]. In spite of having high adsorption efficiency, GO has some disadvantages: (i) possibility of leaching of GO nanoparticles, due to high affinity towards water and (ii) high cost of synthesis [3]. Considering the above challenges, a mixed matrix membrane (MMM) of ultrafiltration (UF) grade can offer

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Nomenclature

C_e	equilibrium concentration	DMF	<i>N, N</i> -di methyl formamide
C_p	permeate concentration	EDX	energy-dispersive X-ray spectroscopy
C_f	feed concentration	FTIR	Fourier transform infrared spectroscopy
k	adsorption constant	GO	graphene oxide
q_e	adsorption capacity	MMM	mixed matrix membrane
ν_m	energy of adsorption	MWCO	molecular weight cut-off
<i>Abbreviation</i>		Pb	lead
AAS	atomic adsorption spectroscopy	PEG	polyethylene glycol
AFM	atomic force microscopy	PSF	polysulfone
BET	Brunauer–Emmett–Teller	Sb	antimony
Cd	cadmium	SEM	scanning electron microscopy
CFR	cross-flow rate	TMP	transmembrane pressure
Cr	chromium	U	uranium
Cu	copper	UF	ultrafiltration
Da	dalton	Zn	zinc

an attracting alternative. The GO impregnated MMM has the following advantages: (i) no leaching of GO nanoparticles; (ii) the membrane can be easily regenerated and (iii) as the required amount of GO is less, the process will be cost effective. GO impregnated MMM is reported using *N*-methyl-2-pyrrolidone as solvent for desalination [5]. However, the membrane technology utilizing the high adsorption capacity of GO for heavy metals remains unexplored.

Presence of heavy metals in aquatic environment poses a serious threat to the ecosystem. Heavy metals, like chromium (Cr), cadmium (Cd), copper (Cu) and lead (Pb) are among the most dangerous pollutants according to Environmental Protection Agency [6]. These metals are extremely toxic, even in trace amount [7]. Industrial and domestic workflow, leaching in effluents and degradation of earth cast are some of the major sources of severe contamination to water stream [8]. The common techniques for remediation of heavy metals include chemical precipitation, electro dialysis, activated carbon adsorption, phyto-extraction, reverse osmosis and ion exchange [9]. These methods have various disadvantages, like generation of high amount of toxic sludge and liquid waste, low throughput, high energy requirement, necessity of pre and post processing and high operating cost, limiting their applicability [10–12]. Hence, there is a need for efficient, high throughput and low energy intensive technique for removal of heavy metals. Ligand based anchoring nanomaterials have several advantages, like high adsorption capacity, selectivity and fast kinetics. However, performance of these mesoporous adsorbents during continuous operation using real life effluent, leaching of nanoparticles and their post treatment are not yet studied well [13–15]. Being physical separation process and less energy intensive, membrane technology can address this problem adequately. Reverse osmosis (RO) is most suitable technology in this context but at the cost of low throughput with high energy requirement [14]. Mixed matrix membrane (MMM) can be an alternative in this regard.

MMM is defined as a class of composite membrane where one or more inorganic additives are dispersed in polymeric matrix. Addition of inorganic additives offers various advantages, like higher adsorption capacity and surface charge leading to enhanced rejection maintaining high permeate flux [15]. Thus, the specific adsorption capacity of targeted smaller solutes by inorganic additive is achieved in MMM along with filtration of larger solutes by polymeric matrix, thereby widening its applicability. More studies on MMM have been undertaken in gas separation processes [16] and their application for treatment of liquid stream is scarce. Selection of GO nanoparticles as an inorganic additive in a polymeric

matrix can have huge potential for specific removal of heavy metal ions from aqueous stream.

In the present study, a novel GO impregnated MMM was designed and prepared using polysulfone (PSF) in *N, N*-dimethylformamide (DMF) for the adsorption of heavy metal ions such as Cr(VI), Cu(II), Pb(II) and Cd(II). As PSF has high thermal and chemical stability in aqueous environment, it is selected as the polymeric matrix [17]. The MMM was systematically characterized in terms of molecular weight cut-off (MWCO), porosity, permeability, average pore size, pore size distribution, contact angle, zeta potential and mechanical strength. The surface morphology and roughness were studied through scanning electron microscopy (SEM) and atomic force microscopy (AFM). The interaction of the oxygenated functional groups with metal ions was captured through Fourier transform infrared (FTIR) spectroscopy. Heavy metal ion removal efficiency under various operating conditions, e.g., transmembrane pressure drop (TMP), cross flow rate (CFR) and solution pH was quantified. In addition, the membrane was tested for long duration filtration of solutions containing individual heavy metal and also for solution of mixed metals under optimized conditions. The membrane was regenerated using in-situ chemical regeneration method. It has strong application potential for filtration of aqueous stream containing heavy metals, like, Cr, Cu, Pb and Cd.

2. Materials and method

2.1. Materials

Polysulfone (PSF, average molecular weight 22,400 Da) was purchased from Solvay Chemicals, Mumbai, India. *N, N*-dimethyl formamide (DMF) and polyethylene glycol (PEG) (molecular weight 6, 10, 20, 35 100 and 200 kDa), were procured from Merck (India) Ltd., Mumbai, India. Dextran (70 kDa) was obtained from Sigma-Aldrich, USA. For preparation of phosphate buffer, dipotassium hydrogen phosphate and potassium di-hydrogen phosphate, hydrochloric acid and sodium hydroxide were purchased from Merck (India) Ltd., Mumbai, India. Analytical grade chromium tri-oxide, lead nitrate, cadmium nitrate and copper sulfate were supplied by Merck (India) Ltd., Mumbai, India.

2.2. Preparation of graphene oxide (GO)

GO was prepared from natural graphite powder by the modified Hummers method using sulfuric acid, potassium permanganate

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