NOVEL ULTRAFILTRATION FLAT SHEET MIXED MATRIX MEMBRANES FOR ARSENIC AND LEAD REMOVAL AND FOULING MITIGATION

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To my parents, my wife and my son

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In the name of Allah, the beneficent and merciful.

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ABSTRACT

The aim of this work is to develop novel, efficient and environmental friendly water treatment technology with low cost and low energy consumption for adsorptive removal of selected heavy metals such as arsenic (As) and lead (Pb) from aqueous system as well as membrane fouling mitigation. In order to overcome the shortages of adsorption and membrane technology, porous asymmetric nanocomposite flat sheet ultrafiltration (UF) mixed matrix membranes (MMMs) incorporated with hydrophilic metal oxide nanoparticle adsorbents were prepared through the phase inversion process. Prior to the fabrication and characterization of MMMs, metal oxide nanoparticles, i.e. Fe-Mn binary oxide (FMBO) with high As adsorption capacity and hydrous manganese dioxide (HMO) with high Pb adsorption capacity were synthesized and used as inorganic fillers and adsorbents in flat sheet polyethersulfone (PES)-based MMMs. The effects of impregnating inorganic metal oxide nanoparticles on the PES-based MMMs morphology, pure water flux, adsorption capacity, surface pattern formation and membrane fouling mitigation were studied by varying the loading of the metal oxide nanoparticles. Both flat sheet PES/HMO and PES/FMBO MMMs were characterized using scanning electron microscope (SEM), contact angle goniometer, atomic force microscope (AFM) and Fourier transforms infrared (FTIR) spectrometer. The best performing membranes prepared from the FMBO/PES ratio of 1.5: 1 demonstrated the pure water flux as high as 94.6 L/m^2.h.bar and maximum As(III) uptake capacity of around 73.5 mg/g. On the other hand the experimental results showed that with increasing HMO:PES weight ratio from zero to 2.0 times, the membrane water flux was increased from 39.4 to 573.2 L/m^2.hr.bar (more than 14 times) and the optimized membranes fabricated from the HMO/PES ratio of 2.0 : 1 showed the highest Pb(II) adsorption capacity i.e. 204.1 mg/g. The continuous UF experiments showed that the optimized MMMs could achieve promising results by removing selected heavy metals from water samples by producing permeate of high quality to meet the maximum contaminant As level set by World Health Organization (WHO), i.e.<10 μg/L As and <15 μg/L Pb. Furthermore, the adsorptive performance of MMMs could be easily regenerated using alkaline and acidic solution. This work also contributed to the novel membrane design with present simple method to control nano-sized pattern formation (alignment of macromolecular nodules) on the polymeric membrane surface. Unlike the lithographical method, the proposed method allows the control of smaller nano-sized patterns of a large membrane area at a lower cost and further shows promising results in reducing membrane fouling due to the protein adsorption. Antifouling property of PES membrane was improved with increasing HMO:PES weight ratio from zero to 1.5 and most importantly, the initial pure water flux of the membranes could be nearly completely recovered by a simple deionized water washing.

ABSTRAK

Matlamat penyelidikan ini dijalankan adalah untuk membangunkan teknologi rawatan air yang terkini, cekap dan juga mesra alam disamping dapat mengurangkan kos dan penggunaan tenaga untuk menyingkirkan logam berat seperti arsenik (As) dan plumbum (Pb) daripada sistem akueus dan dalam masa yang sama dapat mengurangkan kotoran membran. Bagi mencapai matlamat ini, membran kepingan rata ultraturasan (UF) komposit nano berliang tak simetri yang digabungkan dengan partikel nano oksida logam telah dihasilkan melalui proses fasa-balikan. Sebelum proses pembuatan membran campuran (MMMs) dijalankan, oksida dedua Fe-Mn (FMBO) yang berkapasiti jerapan tinggi untuk As dan mangan hidrus dioksida (HMO) yang berkapasiti jerapan tinggi untuk Pb telah disintesis terlebih dahulu dan digunakan sebagai bahan pengisi bukan organik dan bahan penjerap dalam membran kepingan rata berasaskan polietersulfona (PES). Kesan penggabungan oksida logam bukan organik terhadap morfologi membran berasaskan PES, fluks air tulen, kapasiti jerapan, pembentukan corak permukaan dan pengurangan kotoran dikaji dengan mengubah suai kandungan oksida logam tersebut. Kedua-dua membran kepingan rata PES/HMO dan PES/FMBO telah dicirikan dengan menggunakan mikroskopi imbasan elektron (SEM), goniometer sudut sentuh, mikroskop daya atom (AFM) dan spektroskopi inframerah jelmaan Fourier (FTIR). Prestasi membran yang terbaik telah dicapai menggunakan membran yang mempunyai kadar nisbah FMBO/PES sebanyak 1.5:1 dengan kadar fluks air tulen setinggi 94.6 L/m^2.jam.bar dan kapasiti pengambilan As(III) yang maksimum sebanyak 73.5 mg/g. Selain itu, keputusan eksperimen menunjukkan bahawa dengan peningkatan nisbah berat HMO: PES daripada sifar kepada dua kali ganda, fluks air tulen meningkat daripada 39.4 kepada 573.2 L/m^2.jam.bar (lebih 14 kali ganda) manakala membran optimum dengan kadar nisbah HMO/PES sebanyak 2.0:1 menunjukkan kapasiti penjerapan Pb(II) tertinggi iaitu 204.1 mg/g. Ujikaji UF secara berterusan menunjukkan MMMs yang optimum boleh mencapai keputusan yang memberangsangkan melalui penghasilan resapan yang berkualiti tinggi sehingga mencapai tahap aras pencemaran maksimum yang ditetapkan oleh Pertubuhan Kesihatan Sedunia (WHO) iaitu <10 μg/L bagi As dan <15 μg/L bagi Pb dalam menyingkirkan logam berat tersebut. Selain itu, prestasi penjerapan MMMs boleh dijana semula dengan menggunakan larutan beralkali dan berasid. Hasil kerja ini juga menyumbang kepada reka bentuk baru dalam mengawal pembentukan corak bersaiz nano pada permukaan membran polimer melalui kaedah penjajaran nodul makromolekul. Tidak seperti kaedah litografik, kaedah yang dicadangkan ini dapat mengawal corak bersaiz nano yang lebih kecil pada kawasan membran yang besar dengan kos yang lebih rendah dan seterusnya menunjukkan keputusan yang lebih baik dalam mengurangkan kotoran pada membran yang disebabkan oleh penjerapan protein. Sifat anti kotoran yang ditunjukkan oleh membran berasaskan PES dapat ditingkatkan dengan meningkatkan kadar nisbah berat HMO: PES daripada sifar kepada 1.5 dan yang paling penting, fluks air tulen awal yang dicapai oleh membran boleh diperolehi semula sepenuhnya secara mudah dengan membasuh membran menggunakan air ternyahion.

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LIST OF ABBRIVATIONS

AAS	-	Atomic absorption spectroscopy
AC	-	Activated carbon
AFM	-	Atomic force microscope
As	-	Arsenic
As(III)	-	Arsenite
As(V)	-	Arsenate
BSA	-	Bovine serum albumin
CA	-	Cellulose acetate
Cd	-	Codmium
DI	-	Dionized water
ED	-	Electrodialysis
EU	-	European Union
FAAS	-	Flame atomic absorption spectroscopy
FMBO	-	Fe-Mn binary oxide
FTIR	-	Fourier transform Infrared spectroscopy
GFAAS	-	Graphite furnace atomic absorption spectrometry
GS	-	Gas separation
HMO	-	Hydrous manganese dioxide
IOCS	-	Iron oxide coated sand
MAC	-	Maximum acceptable concentration
MC	-	Membrane contactor
MCL	-	Maximum contaminant level
MD	-	Membrane distillation
MF	-	Microfiltration
MMM	-	Mixed matrix membrane
NIL	-	Nanoimprint lithography
NF	-	Nanofiltration

NMP	-	N-Methyl-2-pyrrolidone
PAN	-	Polyaniline
PES	-	Polyethersulfone
PI	-	Polyamide
PP	-	Polypropylene
PSF	-	Polysulfone
PVDF	-	Polyvinylidene fluoride
PVP	-	Polyvinylpyrrolidone
RO	-	Reverse osmosis
SEM	-	Scanning electron microscope
TEM	-	Transmission electron microscope
TMP	-	Transmembrane pressure
UF	-	Ultrafiltration
USPEA	-	United States Environmental Protection Agency
WHO	-	World Health Organization
XRD	-	X-ray diffractometers
Zn	-	Zinc

LIST OF SYMBOLS

A	-	Membrane surface area
b	-	Equilibrium adsorption constant
C_e	-	Eequilibrium concentration
C_f	-	Protein concentration in feed
C_p	-	Permet concentration
C_0	-	Initial concentration of adsorbate
D	-	Diameter
F	-	Permeat flux
g	-	gram
h	-	Hour
J_p	-	Permeat flux
J_w	-	Pure water flux
K_f	-	Freundlich constant
K_1	-	Pseudo-first order rate constant
K_2	-	Pseudo-second t order rate constant
L	-	Liter
m	-	Meter
n	-	Pseudo-first order rate constant
q_e	-	Amount of metal ion adsorbed at equilibrium
q_m	-	Maximum capacity of the adsorbent
q_t	-	Amount of adsorbate adsorbed at any given time
q_0	-	Maximum solid phase concentration of the solute
R	-	Portein rejection
R_a	-	Surface roughness
$R_{a,par}$	-	Surface roughnessin the paprallel direction of casting
$R_{a,per}$	-	Surface roughnessin the perpendicular direction of casting
R_F	-	Flux recovery

R_{ir}	-	Ireversible resistance
R_r	-	Reversible resistance
R_t	-	Total resistance
t	-	Time
V	-	Volume

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CHAPTER 1

INTRODUCTION

1.1 Heavy Metals and their Removal Technologies

Heavy metals are classified as metallic elements with atomic number between 63.5 and 200.6, and a density more than 5.0 g/cm^3 that are found in the earth's crust (Srivastava and Majumder, 2008). Heavy metals can enter the water sources through the natural erosion of soil and rocks. However, the majority of heavy metals contamination also comes from rapid development of anthropogenic activities, especially in developing countries (Fu and Wang, 2011). These hazardous materials, contrary to some organic pollutants, metabolically are not degradable and have tendencies to accumulate in bodies of living beings. Many of them are well-known to be toxic or carcinogenic and their accumulation at higher levels even causes death to human. The most common hazardous heavy metals found in the surface and ground water sources are arsenic and lead. The following is a brief description on these two heavy metals.

Arsenic, is a silver-grey semi-metallic chemical element with earthly abundance of around 2.5 mg/kg. It is ubiquitous and ranks twentyish element in the earth's crust and 14th in seawater. Arsenic has relative molar mass of 74.92 g/mol; density of 5.73 g/cm^3 at 25˚C and melting point of 817˚C (Mandal and Suzuki, 2002). Arsenic normally occurs in two oxidation states: arsenate and arsenite. In surface waters it exists in the form of arsenate, As(V). In ground waters it mostly exists in the form of arsenite, As(III). As(V) is easily removed when compared to As(III). On the other hand, As(III) is considerably more toxic, soluble and mobile

than As(V) where As(III) is sixty times more toxic than As(V) (Singh and Pant, 2004; Hossain, 2006).

Arsenic contaminated drinking water is a worldwide problem. The existence of arsenic in drinking water has been reported in more than 70 countries like USA, China, Bangladesh and Cambodia (Jain and Ali, 2000; Smedley and Kinniburgh, 2002). Figure 1.1 shows a map of the regions affected by high arsenic concentrations and arsenic poisoning.

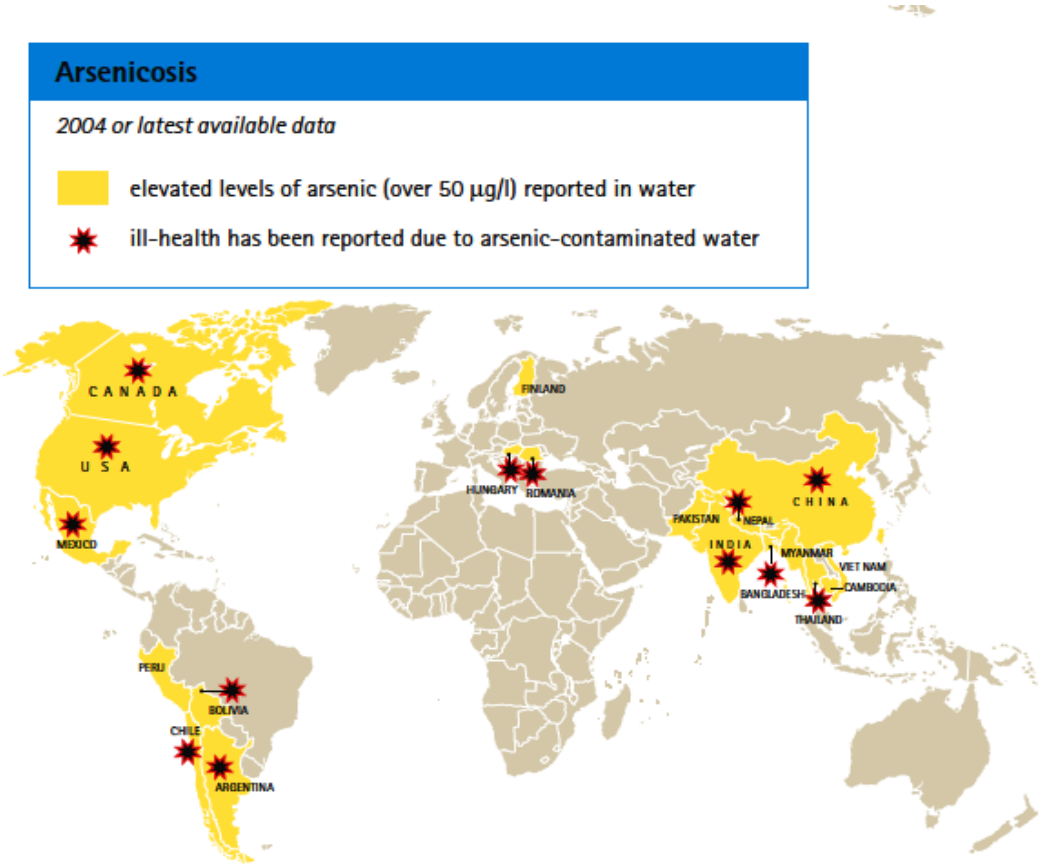

Figure 1.1 Map of the regions affected by high arsenic concentrations and arsenic poisoning (WHO, 2004).

Generally, human exposure to arsenic compounds comes from polluted water, food, and air contaminated by industrial and agricultural activities (DeSesso *et al.*, 1998; Santra *et al.*, 2013). This is of special concern for the reason that in liquid form arsenic is odorless and colorless, making it impossible to recognize by sight only. Arsenic is extremely toxic to human being; some studies show that long term

drinking of arsenic contaminated groundwater can lead to cancer of the bladder, lungs, skin, kidney and liver (Santra *et al.*, 2013). The World Health Organization (WHO) has been well known to establish standards for arsenic in drinking water since 1958. Nowadays, guideline for acceptable arsenic concentration in drinking water is 10 ppb (WHO, 1996).

On the other hand, lead a chemical element with symbol Pb, is a member of group 14 of the periodic table and can be found in the environment in oxidation states: 0, +2, and +4, whereas it infrequently occur in elemental state, Pb(0) (Weast, 1974), While in aqueous solution lead generally can form two classes of compounds: namely plumbous, Pb(II) and plumbic, Pb(IV). Sources of lead which can be released in the environment can be divided in two main categories: natural and anthropogenic sources (Wilkin, 2007; USEPA, 2010). The majority of lead polluted drinking water sources are related to industrial and wastewater effluents, pesticides and waste leachate from lead-acid batteries, paints and pigments to surface and ground water sources (USEPA, 2010).

Lead is very harmful material and has been recorded as the second most toxic and hazardous material after arsenic by the 2007 cerclapriority list of hazardous materials (ATSDR, 2007). Lead has the ability to accumulate in the body by lodging in the folds of the intestine or by being absorbed by the body in dissolved form and depositing in the bones. It is reported that the human exposure to lead has harmful effects on kidney, central nervous and reproductive systems and is more hazardous to children. At the present time, lead toxicity is well-known and many famous organizations around the world have set the maximum contaminant level (MCL) of lead in drinking water at the accessible lowest levels. The United States Environmental Protection Agency (USEPA) guidelines have regulated the maximum permissible concentration of lead in drinking water at 15 ppb (Momčilović *et al.*, 2011).

Traditionally, techniques such as chemical precipitation, coagulation and flocculation and ion exchange resins have been used for removing heavy metals (González-Muñoz *et al.*, 2006; Smara *et al.*, 2007; Pang *et al.*, 2011). The market for

nanotechnology used in water and wastewater worldwide reached USD 1.6 billion in 2007 and is expected to reach USD 6.6 billion in 2015 (Kaiser, 2006). Recently, applicability of nanosized metal oxide adsorbents,extensively have been studied for effective removal of some toxic heavy metals from aqueous effluents. Hua *et al.* (2012) have reviewed the use of nanosized metal oxides for decontaminating hazardous heavy metals from water/wastewater. Zhang *et al.* (2007c), investigated using of Fe-Mn binary oxide (FMBO) particles for arsenic decontamination, while Su *et al.* (2010) used hydrous manganese dioxide (HMO) adsorbent for removing some toxic heavy metals like Pb (II), Zn (II) and Cd (II). However, the above mentioned technologies are incapable of decreasing concentration of heavy metals in real water treatment to the level required by law or are prohibitively expensive or require post treatment steps. The use of membrane separation process in the treatment of polluted water containing toxic heavy metals is an attractive and appropriate method and are being applied more and more frequently (Danış, 2005).

1.2 Heavy Metals Removal based on Membrane Technology

Membrane technology offers a flexible method for meeting multiple water quality objectives and is applied in a wide range of uses. The first recorded study of the membrane process and innovation of osmosis dates back to middle of 18^{th} century when Nollet showed that a pig's bladder was able to pass preferentially water and ethanol (Glater, 1998). This technology in addition of having ability for removing many contaminants such as bacteria and salts is attractive for heavy metals decontamination for small water systems. The membrane technology can address number of water quality problems whereas being comparatively easy to control. The main property which makes it being utilized in separation process is the capability of a membrane to control the permeability of chemical species through the membrane. Membrane separation processes are classified according to the driving force and pore size that cause the flow of permeate through the membrane.

They include microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Intensive investigations have been done for heavy metals

decontamination by RO and NF (Fu and Wang, 2011). UF and MF technologies are becoming widely applied for water and wastewater treatment However, generally the pore sizes of these membranes are bigger than the size of dissolved heavy metal ions, heavy metals passing simply through these membranes. While only UF process especially in the form of micellar and polymer enhanced, is capable for effective removal some of heavy metal ions such as As(V) and Pb(II) (Ferella *et al.*, 2007).

1.3 Membrane Fouling and its Mitigations

Another, important issue in the application of membrane technology in water and waste water treatment is fouling. Membrane fouling can be defined as the increasing accumulation of contaminants on the membrane that causes a growth in the trans membrane pressure (TMP) requirement for the constant permeates flux or a decrease in the water flux through the membrane in constant-pressure operation. It can happen at the surface which is called macro-fouling or inside the pore or pore fouling or micro-fouling.

Membrane fouling can be mitigated by increasing membranes hydophilicity properties and membrane surface modification; this is generally valuable if proteins are the foulant, because proteins have a tendency to adsorb more intensely on hydrophobic membrane surfaces (Wilf and Alt, 2000). Membrane surface modification intensively has also been studied to change its properties to decrease fouling (Rana and Matsuura, 2010). This can be done by several methods like physical and chemical modifications such as ultraviolet irradiation (Nyström and Järvinen, 1987; Zhang *et al.*, 2002; Taniguchi *et al.*, 2003; Wei *et al.*, 2006), graft polymerization (Ulbricht and Belfort, 1996; Wavhal and Fisher, 2002; Liu *et al.*, 2008), micro-patterning (Lee *et al.*, 2013) and/o nanoimprint lithography (NIL) on the membrane surface (Maruf *et al.*, 2013). Recently surface hydrophilization of the polymeric membranes for the decreasing fouling property has also been widely investigated by dispersing metal oxide nanoparticles into dope solutions by many metal nanoparticles such as titanium dioxide, aluminum oxide, silicon dioxide and zirconium oxide (Ng *et al.*, 2013).

1.4 Problem Statements

Nowadays, the toxicity of hazardous heavy metals has been well known and many organizations around the world adjusted the maximum acceptable concentration of heavy metals in contaminated-water at very low concentration. Stringent drinking water regulations are made in order to lower the MCL of heavy metal concentration. For instance, since 2006, USEPA and WHO have decided to reduce the maximum arsenic concentration in drinking water from 50 part per billion (ppb) to 10 ppb (Mohan and Pittman Jr, 2007). The stiffening of regulations generates strong demands to improve methods for removing pollutants from the water and controlling water-treatment residuals.

Conventionally, many treatment methods such as chemical precipitation (Harper and Kingham, 1992), coagulation and flocculation (Bilici Baskan and Pala, 2010) and ion exchange (Kartinen Jr and Martin, 1995) could be employed for heavy metals decontamination, but they are found to have inconsistent and/or incomplete elimination of heavy metals. In order to meet the MCL required by law, additional post-treatment process always is required to complete the treatment process, which indirectly would increase the overall cost of treatment. Although membrane technology is reported to be used for heavy metals removal when it is operated in NF or RO mode (Oh *et al.*, 2004; Chan and Dudeney, 2008), the relatively high energy consumption resulted from high operating pressure remains as a concern to many. Low pressure driven membranes like MF and UF on the other hand are not effective in removing heavy metals, mainly due to their porous structure which offers minimal/none resistance against arsenic (Brandhuber and Amy, 1998).

Adsorption is now recognized as an effective and economic method for heavy metal wastewater treatment. The adsorption process offers flexibility in design and operation and in many cases will produce high-quality treated effluent. In addition, because adsorption is sometimes reversible, adsorbents can be regenerated by suitable desorption process (Harper and Kingham, 1992; Fu and Wang, 2011). Recent investigations show many of metal oxide nanoparticles have high adsorption capacity and selectivity for removing some of hazardous heavy metals from

contaminated water (Deliyanni *et al.*, 2007; Hua *et al.*, 2012). This partially can be attributed to their high surface areas and activities because of their size-qualification effect (Henglein, 1989). However, as the size of metal oxides reduces from micrometer to nanometer levels, the increased surface energy inevitably leads to their poor stability. Consequently, nano sized metal oxides are prone to agglomeration due to Van der Waals forces or other interactions (Pradeep and Anshup, 2009; Hua *et al.*, 2012). Their high adsorption capacity and selectivity would be significantly reduced or even lost. Furthermore, metal oxide nanoparticles are not capable for using in fixed beds or any other flow-through system due to excessive pressure drops or difficult separation from aqueous solutions and poor mechanical strength, on the other hand, unfortunately, it may be extremely difficult to remove 100 % of these metal oxides from aqueous solution, even if they have unique properties (Li *et al.*, 2012)

Another problem for using of metal oxide nanoparticles in real application for water treatment is their nontoxicity effect, while, they can enter through the skin and can be translocated to lymph nodes, and if they enter blood circulation, they can be distributed throughout the body and taken up into the liver, spleen, bone marrow, heart and other organs (Mishra *et al.*, 1996). So, not only can nanoparticles have adverse effects on their own, if they are associated with or comprised of toxic metal ions they may release them into the body once they are exposed to the various and complicated chemistries.

To overcome these problems and promote the applicability of metal oxide adsorbent nanoparticles in real water and wastewater treatment and heavy metal decontamination, researchers in recent years have focused on impregnating nanoparticles into porous host media such as Bentonite (Ranđelović *et al.*, 2012), alginate (Guo and Chen, 2005), zeolite (Li *et al.*, 2011b), diatomite (Jang *et al.*, 2006; Jang *et al.*, 2007), cellulose (Guo *et al.*, 2007) and porous polymer (Pan *et al.*, 2009; Su *et al.*, 2009). But compared to other host materials, porous polymeric hosts are a particularly attractive option partly because of their controllable pore size and surface chemistry as well as their excellent mechanical strength for long-term use (Hua *et al.*, 2012).

In the development and practical use of UF membranes, one of the main problems that should be addressed is fouling, while, fabricating of low fouling membrane is significant and attractive subjects. Therefore, in this study to be practically useful, current proposed investigation is to fabricate low fouling membrane via impregnating hydrophilic metal oxide particles in the polymeric structure of the membranes and also surface modification must further be improved with respect to fouling resistance, stability and the membrane performance.

1.5 Objectives of the Study

Based on the problem statement identified, the major objective of this research was to fabricate and characterize novel nanocomposite UF MMMs, to remove selected heavy metals from aqueous system. Therefore the objectives of this research are:

1) To fabricate and characterize polyethersulfone (PES)/FMBO mixed matrix membranes (MMMs) for removing arsenite from contaminated water samples.

2) To prepare and characterize PES/HMO MMMs for removing lead from polluted waters.

3) To study the effect of impregnating inorganic metal oxide particles in the PES matrix in terms of surface pattern formation and BSA fouling mitigation.

1.6 Scopes of the Study

In order to achieve the above mentioned objectives, the following scopes of works have been identified:

1) Synthesizing FMBO through chemical precipitation process using low cost chemical such as potassium permanganate, ferrous sulphate and sodium hydroxide as precipitant factor.

2) Preparing HMO nanoparticles by chemical precipitation process using very cheap materials, i.e. potassium permanganate,manganese sulphate and sodium hydroxide.

3) Characterizing the FMBO and HMO particles using, Fourier Transform Infrared spectroscope (FTIR), X-ray diffractometer (XRD) and Transmission Electron Microscope (TEM).

4) Fabricating and characterizing the MMMs in the form of flat sheet by dispersion metal oxide particles into PES dope solution. The MMMs formation process was carried out based on dry-wet phase inversion process.

5) Investigating the performance of MMM embedded with different loadings of inorganic metal oxides nanoparticles for heavy metals removal under different process conditions.

6) Evaluating sustainability of membranes in terms of BSA fouling mitigation and adsorbents regeneration.

1.7 The Significance of Research

The significance of this current research is the development of novel, efficient and environmental friendly water treatment technology with low cost and low energy consumption for hazardous heavy metal decontamination. The hydrophilic nanosized metal oxide adsorbents with high adsorption capacity for As(III) and Pb(II) removal has been synthesized and was used as inorganic filler for mixed matrix membranes preparation. Besides showing promising results in eliminating selected heavy metal ions from contaminated drinking water, the resultant mixed matrix membranes also exhibited excellent antifouling properties against proton desorption, mainly due to the nano-sized pattern formed on membrane surface coupled with improved surface hydrophilicity. As high as 97.5% of the original adsorption capacity of PES/HMO-2.0 MMM for Pb(II) adsorption, and 87.5% of the original adsorption capacity of PES/FMBO-1.5 MMM for As(III) adsorption, could be easily recoverd after subjecting the membranes to a simple desorption process using acidic and alkaline solution.

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