Trina Dutta<sup>a</sup>, Chiranjib Bhattacherjee<sup>b</sup>, Sangita Bhattacherjee<sup>c</sup>

<sup>a</sup> Asst. Professor, Chemistry Dept, JIS College of Engineering, Kalyani, Nadia - 741235

<sup>b</sup> Professor, Chemical Engineering Dept., Jadavpur University, Kolkata - 700032

<sup>c</sup> Asst. Professor, Chemical Engineering Department, Heritage Institute of Technology, Kolkata – 700107

**Abstract:** 

Arsenic poisoning has become one of the major environmental problems in the world as

millions of human beings have been exposed to excessive arsenic through contaminated

ground water used for drinking. Arsenic is classified as Group 1 carcinogenic substance

to humans based on powerful epidemiological evidence. In 2001 USEPA promulgated a

rule for lowering the Maximum Contaminant Level (MCL) from 50µg/L to 10µg/L.

Hence a continuous investigation of the available arsenic removal technology is essential

to develop an economical yet effective method for removing arsenic from water. This

paper offers an overview of the application of membrane technology in the water

treatment research that have already been realized or that are suggested on the basis of

bench scale or lab scale research. The performance of various membrane processes such

as Reverse Osmosis (RO), Nanofiltration (NF), Ultrafiltration (UF), Microfiltration (MF)

and Membrane Distillation (MD) in removal of arsenic from water have been presented.

The influence of pH, membrane material, membrane types etc. on arsenic removal

efficiency using membrane technologies have also been explored.

Keywords: Arsenic removal, Reverse Osmosis, Nanofiltration, Ultrafiltration,

Microfiltration

#### 1.0 Introduction

Drinking arsenic contaminated water is a major threat to mankind. Arsenic is the twentieth most abundant element in the earth's crust, fourteenth in the seawater and the twelfth most abundant element in the human body (Mandal and Suzuki, 2002). Though it is necessary as a nutrient in small quantities, it is known to be highly toxic if ingested in large dose. Acute and chronic Arsenic exposure through drinking water has been reported in many countries. Its elevated concentrations are found in groundwater in some areas of India, Bangladesh, China, Chile, Argentina, Mexico, Hungary, Taiwan, Vietnam, Japan, New Zealand, Germany and the United States due to naturally occurring arsenic in the aquifer sediment (Bang *et al.*,2005). As per the guideline recommended by World Health Organization, the maximum limit of arsenic in drinking water should not exceed 10 ppb. Numerous investigations (Bhattacharjee *et al.*,2005; Harvey *et al.*,2005; Pokhrel *et al.* 2009) were carried out some of which focus on West Bengal (India) and Bangladesh where the largest affected population lives on the Bengal Delta Basin.

Arsenic, atomic number 33, is in Group 15 of the periodic table, directly below phosphorus. In the natural environment, Arsenic is rarely encountered as the free element; however it can occur in four oxidation states (-3, 0, +3, +5), but the two predominated oxidation states common in drinking water are oxyanions of trivalent Arsenic(As [III]) and pentavalent Arsenic (Hem, 1992). Technologies those have been used across the globe to remove arsenic below permissible limit include adsorption, precipitation, ion-exchange and membrane technology etc. The first three methods require huge chemicals and either generate high volumetric sludge or release noxious chemical reagents into the

environment during regeneration. Membrane technology is superior to those as membrane

itself does not accumulate arsenic or generate sludge which would cause disposal

problems. Instead it prevents other dissolved impurities and harmful microorganisms to

pass through the membrane. This review paper briefly summarizes various membrane

processes that have been used earlier for the removal of arsenic and their merits and

demerits.

2.0 Arsenic Removal using Membrane Separation

Membranes are typically synthetic materials with billions of pores or microscopic holes

that act as a selective barrier; the structure of the membrane allows some constituents to

pass through, while others are excluded or rejected. The movement of molecules across

the membranes needs a driving force, such as pressure difference between the two sides

of the membrane. Various types of pressure driven membrane technologies such as

Reverse osmosis (RO), Nano-filtration (NF), Ultra-filtration (UF), microfiltration (MF),

membrane distillation (MD) have been explored by researchers for the removal of arsenic

from contaminated water.

2.1 Arsenic Removal using RO

Reverse osmosis is the oldest and has been identified as the best available technology to

help small water treatment systems to remove arsenic from water. RO membrane

contains extremely small pores (< 0.001 µm) (Schneiter, 1983) and a very high (often

close to 100%) rejection of low-molecular mass compounds and ions can be achieved.

Moreover, the process can easily be automated and controlled. With the invent of

cellulose acetate RO membrane in the 1980's, arsenate removal efficiency of above 90%

have been achieved with the RO system operated at high-pressure around 400 psi (Clifford *et al.*, 1986; Fox, 1989). However, arsenite removal efficiency is less than 70%.

Brandhuber and Amy (1998) conducted experiments on the removal of arsenic by using four types of RO membranes-TFC 4921 (Fluid Systems), TFC 4820-ULPT (Fluid Systems), AG 4040 (DESAL) and 4040LSA-CPA2 (Hydranautics). It has been reported that As (V) rejection (95%) was significantly greater than As (III) rejection (65-85%) by all RO membranes.

The Environmental Technology Verification Program operated by the USEPA, used the TFC-ULP RO membrane from Koch Membrane Systems in which total arsenic removal efficiency was reported around 99% for an influent concentration of 60 μg/L (Koch membrane Sys.,2001). Kang *et al.*(2000) investigated the removal of arsenic from water by using two types of RO membranes, ES-10 (polyamide) and NTR- 729HF (polyvinyl alcohol) (Nitto Electric Industrial Co, Japan). The removal of arsenate by ES-10 was more than 95% and that of arsenite was between 75 and 90% while the removal of arsenate and arsenite by NTR-729HF was 80-90% and 20-43%, respectively. From the investigation it was also observed that solution pH played a significant role in removal of both arsenate and arsenite from water. The removal of arsenate using NTR-729HF from around 80% at pH 3 suddenly increased to 95% with the increase of pH from 5 to 10. The removal efficiency of arsenite achieved by using ES-10 was around 75% at a pH 3 and that was increased to 90% at pH10. This could be explained that the membrane developed negative charge at higher pH which would lead to the formation of electrical

double layer adjacent to membrane surface thus caused hindrance to the passage of

Amy *et al.* (1999) performed bench scale single element and flat sheet RO testing by using a RO membrane, DK2540F (DESAL). For single element testing, the removal efficiency of arsenate was 96% while that of arsenite was around 5%. The removal efficiency of arsenate was around 88% for the flat sheet testing. Waypa *et al.* (1997) studied arsenic removal from synthetic water and from surface water sources using thin-film composite type membrane. Both As (V) and As (III) were effectively removed from water by RO - 99% rejection of As (V) was obtained with RO membrane. Thin-film composite type membrane showed higher selectivity and required lower driven pressure and gave higher flow rate of permeate.

Considering the developing countries' situation, such as low annual income and low electric popularization, Oh *et al.*(2000) studied the rejection of arsenic by using HR3155 membrane (Toyobo Co., Ltd) made of cellulose triacetate coupled with a bi- cycle pumping system. The result showed that the removal efficiency of arsenate was over 95% and that of arsenite was around 55%. Thus, RO process coupled with a bicycle pump device could be used for the removal of arsenic in areas where the electricity supply is not feasible or available (Lhassani *et al.*,2001). Removal of arsenic by some commercial reverse osmosis membranes is summarized in table-1(Uddin *et al.*, 2007).

## 2.2 Arsenic Removal using NF

arsenate and arsenite anions.

Nanofiltration (NF), a cross-flow filtration technology, ranges somewhere between UF and RO and is operated under a transmembrane pressure not exceeding 3MPa and thus

reduces the operating cost significantly compared to RO. The nominal pore size of the membrane is typically about 1 nm and MWCO is typically less than 1000Da. However, NF membranes are still subject to scaling and fouling and often modifiers such as antiscalants are required for use.

NF membranes are usually applied to separate multivalent ions from monovalent ions. These membranes have slightly charged surfaces. Charge interaction plays a dominant role in separation of molecules using this membrane. NF membranes are usually asymmetric and negatively charged at neutral and alkaline media but lose their charge in acidic pH. This type of membrane can remove both dissolved arsenate and arsenite due to size exclusion according to the report of EPA in 1999 (EPA draft, 1999). Brandhuber and Amy (1999) studied the rejection of arsenic by using three NF membranes-NF70 4040-B (Film Tec), HL-4040F1550 (DESAL) and 4040-UHA-ESNA (Hydranautics). The As(V) rejection was more than 95%, whereas the As(III) rejection was 20-53% for all the three membranes. This could be due to the fact that As (V) exists as an anion at the typical pH (5-8) in natural water while As(III) remains as a neutral molecule at the same pH region. Due to electrostatic repulsion between arsenate molecules and the charged NF membrane at neutral and alkaline pH, the rejection of arsenate was substantially much higher than that of arsenite. In another experiment (Levenstein et al., 1996), a commercial loose, porous polyamide thin film composite membranes, NF-45 (Film Tec, Minnetonka, MN) showed 90% removal of As(V). The removal of As(III), however, was 10-20%. The mechanism of transport and rejection characteristics of NF membranes are quite complex. The separation of anions is based not only on different

rates of their diffusion through the membrane (at low pressure), convection (at high pressure), but also on repulsion (Donnan exclusion) between anions in solution and the surface groups, which is obviously higher for multi-valent anions. The advantage of introducing this additional mechanism of ion exclusion (in addition to the size-based exclusion) is that high ion rejections similar to those in RO can be achieved at higher water flux through the membrane. Donnan exclusion compared to other pressure driven membrane process has a pronounced effect on the separation in NF. Due to slightly charged nature of the membrane, solutes with an opposite charge compared to the membrane (counter-ions) are attracted, while solutes with a similar charge (co-ions) are repelled. At the membrane surface a distribution of co- and counter-ions takes place, thereby causing an additional separation (Nath, 2008).

Urase *et al.*(1998) carried out experiment on the rejection of different arsenic compounds by low pressure aromatic polyamide NF membrane, ES-10 (Nitto-Denko Co. Ltd). In this study, ground water collected from a shallow well near the University of Tokyo was spiked with arsenate and arsenite. In the experiment, 50-89% rejection of As(III) and 87-93% rejection of As(V) was observed.

A set of NF study (Vrijenhoek, 2000) for Arsenic removal of ground water and shallow water were carried out using three types of commercial membrane ES-10 (aromatic polyamide), NTR- 7250 (polyvinyl alcohol), and NTR-729HF (polyvinyl alcohol) obtained from Nitto Electric Industrial Co., Japan. The removal efficiency of As(V) and As (III) was found to be almost the same for both the synthetic and ground water, showing no significant effect of the ionic composition of the water source on arsenic

removal. In another research (Sato *et al.*, 2002; Saitúa H *et al.*, 2005) using these ES-10, NTR-7250 and NTR-729HF membranes, the removal efficiency of arsenate exceeded 85% and increased with the increase of pressure for all three membranes.

Another study (Seidel et al., 2001) revealed that much higher removal efficiency of arsenic was obtained in presence of sodium chloride, especially at low arsenic content in water. However, the rejection of arsenic was reported to decrease by about 5% in the when the porous polyamide thin-film composite membrane (NF-45) was replaced by sulfonated polysulfone thin-film composite membrane denoted BQ01 by the manufacturer (Osmonics, San Diego, CA). These results showed the effect of sodium chloride on arsenic removal depends on the characteristics of the membranes. The performance of NF membrane was found to be affected by the solution pH. It was reported by Vrijenhoek and Waypa (2000) that the rejection of As(V) species increased significantly from 25% at pH 4 to more than 80% at pH 9 due to change of As(V) species from monovalent ions (H<sub>2</sub>AsO<sub>4</sub><sup>-</sup>) to divalent ions with increasing pH. At pH 4-8, the charged sites of NF-45 membrane were negatively charged. Hence higher Donnan exclusion could be one reason for obtaining higher rejection. Larger hydrated radii of divalent ion compared to monovalent ions were another possible cause for increased rejection of As(V) species. The separation of uncharged As(III) remained unaffected with change in pH over the range 4-8.

In a very recent work (Zhao *et al.*, 2012), arsenic removal was investigated with a simulated aqueous solution of arsenic salt (Na<sub>2</sub>HAsO<sub>4</sub>.7H<sub>2</sub>O) by employing self-made PMIA (Poly m–phenylene isophthalamide) nanofiltration membrane and more than 90%

rejection of arsenate was reported. As(V) rejection increased from 83% at pH 3 to 99% at

pH 9. Presence of NaCl increased As(V) rejection in the feed range studied and was

reported to be 99% with a feed concentration of 100µg/L. However presence of Na<sub>2</sub>SO<sub>4</sub>

decreased the rejection by about 8%.

Rejection of Arsenic species by various NF membranes is presented in Table 2 (Uddin et

al., 2007).

2.3 Arsenic Removal using UF

Ultrafiltration (UF) is primarily a size exclusion-based pressure-driven membrane

separation process. UF membranes typically have pore sizes in the range from 10 to

1000<sup>o</sup>A and are capable of retaining species in the molecular weight range of 300 to

500,000 dalton. Typical rejected species include sugars, bio-molecules, polymers, and

colloidal particles.

UF may not be viable technique for the removal of arsenic due to the large pore size of

the membrane. UF with electric repulsion may have higher arsenic removal efficiency

compared to UF with only pore size dependent sieving. A series of bench-scale tests

(Amy et al., 1998) were performed to investigate the influence of membrane charge on

the arsenic removal efficiency. Negatively charged GM2540F UF membrane and

uncharged FV2450F UF membrane were used in the investigation. From the results it

was found that GM2540F membrane gave better rejection of As(V) at neutral pH

compared to acidic pH whereas FV2540F membrane gave poor rejection for both of AsV

and AsIII species. So, it can be stated that electrostatic interaction between arsenic ions

and negatively charged membrane surface could be a reason for the higher removal rates of As(V).

Another study (Brandhuber and Amy, 2001) was conducted to investigate the removal of arsenic from water using negatively charged, thin film, composite sulphonated polysulphone UF membrane (Osmonics GM). The influence of co-occurring divalent ions and natural organic matter (NOM) on arsenic rejection by charged membrane was explored. In presence of divalent cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>), As(V) rejection reduced almost to zero. This reduction in As (V) rejection could be attributed due to formation of ion pairs between counter ions and the fixed charge group in the membrane matrix, which locally neutralizes the membrane charges. The presence of NOM improved As(V) rejection in presence of divalent cations. This might be due to the complexion of divalent ions whose presence in solution tends to reduce As (V) rejection. Another possibility might be caused due to adsorption of Natural organic matter (NOM) onto the membrane surface to form a negatively charged layer. Higher NOM concentration in solution led to higher charge density in the adjacent membrane layer causing more rejection of negatively charged arsenic species.

In another study (Iqbal *et al.*, 2007) the effect of co-occurring inorganic solutes (HCO<sub>3</sub>, HPO<sub>4</sub><sup>2-</sup>, H<sub>4</sub>SiO<sub>4</sub> and SO<sub>4</sub><sup>2-</sup>) in feed water on the removal of As(V) and permeate flux were investigated by using a cationic surfactant cetlylpyridinium chloride (CPC) and a flat sheet hydrophilic polyethersulfone (PES) ultrafiltration membrane (Millipore, Bedford, MA). PES membrane without surfactant micelles was found to be ineffective for arsenic removal while the addition of surfactant significantly increased the arsenic

removal efficiency. Arsenic removal with surfactant was found to be 78-100% while arsenic removal in the presence of inorganic solutes was only 25%, corresponding to a permeate water arsenic concentration of 30  $\mu$ g/L.

Brandhuber and Amy (2001) had also studied arsenic removal using negatively charged ultrafiltration membrane in the pH range 2-10. It was found that the degree of arsenic rejection increased with increase in pH. The rejection of As(V) increased with increase of pH from around 13% at pH 2 to more than 80% at pH 10. The rejection of arsenite ions was much lower than that of arsenate ion. The rejection of As(III) was reported around 15% in the pH range of 4-10 and about 40% with the increase of pH from 10 to 11. They also studied the effect of temperature on efficiency of arsenic removal. It was reported that an increase of 20°C temperature had caused slight decrease of As(V) rejection from 82% (at 20°C) to 76% at 40°C. This was due to higher diffusive transport of arsenic species at higher temperature.

Iqbal *et.al.* (2007) had investigated the removal characteristics of arsenate with ground water as feed using micellar-enhanced ultrafiltration (MEUF). Among four different cationic surfactants used, the highest removal efficiency of arsenic (96%) was obtained in case of hexadecylpyridinium chloride (CPC) and the removal efficiency with hexadecyltrimethylammonium bromide (CTAB) was 94%. But the removal efficiency with benzalkonium chloride (BC) was the lowest (57%) due to higher critical micelle concentration (CMC) of BC compared to those of other surfactants. During MEUF, over 80% of arsenic was removed with octadecylamine acetate (ODA).

## 2.4 Arsenic Removal using MF

Microfiltration is a low pressure membrane process for separating colloidal and suspended particles in the range 0.1 to 10 µm. It closely resembles conventional coarse filtration or sieving. This type of membrane is porous enough to pass molecules of true solution. Microfiltration alone cannot remove the dissolved arsenate and arsenite species from arsenic contaminated water. Removal of arsenic by MF membrane can only be achievable by increasing particle size of arsenic bearing species. Coagulation (Ghurye et al.,2004; Chwirka, 2004; Han et.al., 2002; Meng et. al., 2000) and flocculation processes prior to MF could increase the particle size of arsenic bearing species effectively and were found to remove arsenic species from arsenic contaminated water. MF membranes made of mixed esters cellulose acetate and cellulose nitrate with pore size 0.22 to 1.2 µm and combined with ferric chloride or ferric sulphate and cationic polymeric flocculants were used to investigate the arsenic removal efficiency (Chwirka, 2000). The results showed that the arsenic removal efficiency by the combination of flocculation and MF technique are higher than MF only and depend on the effectiveness of arsenic adsorption onto the ferric complexes present and on the rejection of the arsenic containing flocs formed by the MF membrane (Han et al., 2002). During the coagulation process, ferric chloride hydrolyzed in water to form ferric hydroxides precipitate bearing a net positive charge on its surface. In the pH range 4-10, negatively charged arsenate anions got effectively adsorbed by forming surface complex (Shih, 2005). Removal of arsenite was poor as it remained as neutral species in the above pH range and could not get adsorbed. Complete removal of arsenic from water could have been achieved by completely oxidising arsenite to arsenate prior to coagulation-microfiltration process. Coagulation-

flocculation process increases the effective size of arsenic containing particles and thus makes it possible to remove arsenic species using low-pressure membrane technology

like MF.

In some recent study (Ghosh et.al., 2011), electrocoagulation (EC) followed by

microfiltration using a ceramic membrane was found to be quite effective in removal of

arsenic from a concentration of 200 µg/L in feed solution in presence of fluoride and iron

contaminant to a arsenic content of 8.7 µg/L. EC experiment consisted of a bath with

four aluminium sheets of 0.15 m X 0.05 m X 0.002 m. EC experiments were continued

upto 45 minutes with a current density of 625 Am<sup>-2</sup>.

2.5 Arsenic Removal using MD

Membrane distillation is a non-isothermal membrane separation process which employs a

microporous hydrophobic membrane with pore size ranging from 0.01 µm to 1 µm. It is

known since 1963and is still being developed in desalination testing stages and not fully

implemented in industry. The main requirements for MD process are that the membrane

should not be wetted and only vapor and non condensable gases should be present within

its pores. Such hydrophobic, microporous membranes made of polytetrafluoroethylene

(PTFE), polypropylene (PP), polyethylene (PE), and polyvinylidenefluoride (PVDF) are

now commercially available. Recently Qu et.al.(2009) had studied arsenate and arsenite

removal using self-made polyvinylidene fluoride (PVDF) membrane in a direct contact

membrane distillation unit.

The experimental results indicated that the permeate As(III) and As(V) were under the

maximum contaminant limit (10 µg/L) until the feed As(III) and As(V) concentration

achieved 40 and 2000 mg/L, respectively. Islam (2005) studied arsenic separation by air-

gap membrane-distillation (AGMD) using a small-scale commercial prototype MD

module and reported successful treatment of arsenic-contaminated water. In another work

(Macedonio and Drioli, 2008), RO followed by membrane distillation was found to be

effective in reducing arsenic concentration below 10µg/L when 36% RO permeate had

been recycled to MD unit.

Manna et.al.(2010) had performed experimental investigation for arsenic removal from

contaminated ground water using solar-driven MD process by employing a flat sheet

cross flow membrane module fitted with a hydrophobic PVDF membrane. This direct

contact MD unit with 0.13µm PVDF membrane produced high flux of 95 kg/m<sup>2</sup>.h at a

feed water temperature of 60°C for a feed flow rate of 0.120m<sup>3</sup>/h.

3.0 Conclusion

Arsenic level can be brought down effectively by using membrane technology. However

the removal efficiency varies considerably depending upon the technology and process

parameters as well. Effects of pH, temperature, initial concentration, were found to affect

arsenic removal efficiency substantially. In all types of removal methods, pentavalent

arsenic was found to be more effectively removed than arsenties (AsIII). Due to the ionic

charge, arsenate (V) is more easily removed from source waters than arsenite (III).

The membrane technologies, RO, NF, UF, coagulation followed by MF and MD have

been demonstrated to be quite effective to remove arsenic from water and meet the

arsenic MCL standard. However, the effectiveness of membrane technologies is

dependent to a variety of parameters like source water characteristics, other water

contaminants, arsenic species, membrane characteristics, membrane materials, morphology

etc. Among membrane technologies, high pressure membrane processes such as RO and

NF are more effective in removing arsenic species than UF and MF. Negatively charged

UF membrane were found to be effective in rejecting arsenic species. Miceller-enhanced-

ultrafiltration using suitable surfactant proved to increase arsenic removal significantly.

In case of NF membranes, Donnan exclusion plays significant role in separation of

anionic species of arsenic along with convection and diffusion. Microfiltration followed

by coagulation-flocculation was found to be very effective in removal of arsenic. This

hybrid treatment technology has proved to be an efficient as well as less energy

consuming, low cost technology. Recently MD processes are found to be useful for this

purpose. From the encouraging results obtained from solar driven direct contact

membrane distillation module, it may be stated that MD process could be an effective

alternative for removal of arsenic from water in the vast arsenic affected rural areas of

south-east Asian countries.

Though a number of arsenic removal methods are available, a more cost-effective,

environment friendly technology is yet to be developed so as to further improve the

efficiency of arsenic removal as well as to resolve sludge and arsenic concentrates

management problems.

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# **List of Tables:**

- 1. Percent Rejection of arsenic by RO membrane.
- 2. Percent Rejection of arsenic by NF membrane.



Table 1: Rejection (%) of arsenic by RO membrane

Model	Supplier	Water	Rejection%	
			As(III)	As(V)
TFC 4921	Fluid Systems	Ground water	63	95
TFC 4820- ULPT	Fluid Systems	Ground water	77	99
AG 4040	Desal	Ground water	70	99
4040 LSA CPA2	Hydranautics	Ground water	85	99
TFC ULP	Koch Membrane Systems	Ground water (USA)	99	100
ES 10	Nitto Electric Industrial Co. (Japan)	Distilled water	75	95
NTR-729HF	Nitto Electric Industrial Co. (Japan)	Distilled water	20-43	80-95
DK2540F	Desal	Lake water	5	96
HR3155	Toyobo Co., Ltd.	Ground water	55	95

Table :2 Rejection (%) of Arsenic by NF membrane

Model	Supplier	Water Origin	Rejection	
			As(III)	As(V)
NF70 4040-B	Film Tec (Dow Chemical)	Colordo River	53	99
HL- 4040F1550	DESAL	Idem	21	99
4040-UHA- ESNA	Hydranautics	Idem	30	97
NF-45	Film Tec	Synthetic water	10	90
ES-10	Nitto-Denko Co. Ltd.	Ground water	50-89	87-93
ES-10	Nitto Electric Industrial Co., Japan	Synthetic water	80	97
NTR-729HF	Nitto Electric Industrial Co., Japan	Synthetic water	21	94
NTR- 7250	Nitto Electric Industrial Co., Japan	Ground	10	86
NF 70	Film Tec	Fresh water	99	99
NF 270	Film Tec	Ground water(Osijek)	-	99
NFc	Film Tec	Ground water (Osijek)	-	96