

## Arsenic removal by conventional and membrane technology: An overview

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Presently used arsenic removal technology has been reviewed, pointing especially to the promise of membrane technologies as a practical means of purification. The membrane technologies include reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF). Among them, the applications of the first two have proved to be reliable in removing arsenic from water. The influence of membrane materials, membrane type, operating conditions such as temperature, pressure, pH of the feed solution and feed concentration on arsenic removal efficiency by membrane technologies are discussed. This paper also provides a comparison between conventional technologies and membrane technologies for arsenic removal and concludes that membrane technology is preferred for water treatments to meet the maximum contaminant limit (MCL) standard.

**Keywords:** Arsenic removal, Adsorption, Membrane technology, Reverse osmosis, Nanofiltration, Ultrafiltration, Microfiltration

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Arsenic poisoning has become one of the major environmental problem in the world as millions of human beings are exposed to excessive arsenic through contaminated drinking water. Acute and chronic arsenic exposure via drinking water has been reported in many countries, especially Argentina, Bangladesh, India, Mexico, Mongolia, Thailand, and Taiwan, where a large proportion of ground (well) water is contaminated with a high concentration of arsenic, in some cases exceeding 2,000  $\mu\text{g/L}$ <sup>1</sup>. In Bangladesh<sup>2</sup> ground water, the arsenic concentration ranges from less than 0.25  $\mu\text{g/L}$  to more than 1600  $\mu\text{g/L}$  and in West Bengal (India)<sup>3,4</sup> — from less than 10 to 3200  $\mu\text{g/L}$ . Thousands of people in both Bangladesh and India have already shown the symptoms of arsenic poisoning and several millions are at risk of arsenic contamination from drinking tube well water. Arsenic toxicity has no known effective treatment, but drinking of arsenic free water could help the people of arsenic affected area to get rid of the arsenic poisoning. Hence, provision of arsenic free water is urgently needed to mitigate arsenic toxicity and protection of health and well being of rural people living in acute arsenic problem area in Bangladesh and India. There is, therefore, a great need to develop efficient methods for arsenic removal from drinking water. Researchers all over the

world have been working for years on the development of arsenic removal technology. Due to the importance of the issue, systemization of the available and new information should be done on regular basis. Ming-Cheng Shih<sup>5</sup> has recently made overview of arsenic removal methods by pressure-driven membrane processes. Meenakshi and Maheshwari<sup>6</sup> have overviewed the conventional method for arsenic removal. The purpose of this paper is to present a broad overview of the presently used arsenic removal processes, both conventional and membrane processes, for the removal of arsenic from drinking water. A comparison between conventional processes and membrane processes for the removal of arsenic has also been discussed.

### Arsenic removal technologies

There are several methods available for the removal of arsenic from water. The most commonly used technologies include coagulation, adsorption, ion exchange, bacterial treatment and membrane processes<sup>7-12</sup>.

Arsenic is available in groundwater in As(III) and As(V) forms in different proportions. It has been observed that all types of arsenic removal technologies show better performance when arsenic is present in pentavalent form. For this reason, oxidation

of As(III) appears as a pre-treatment step in most treatment technologies. As (III) is oxidized to As(V) by the treatment with air, oxygen, ozone, free chlorine, hypochlorite, permanganate, hydrogen peroxide/ $\text{Fe}^+$  (Fenton's reagent). Air oxidation of arsenic is very slow and can take weeks for oxidation<sup>13</sup>.

### Conventional technologies for arsenic removal

The conventional technologies work more or less on adsorption principle. In classical adsorption process, arsenic is adsorbed on the active site of the adsorbents. In coagulation, coagulants form flocs and arsenic species are adsorbed on the flocs, which can be easily filtered or settled under the influence of gravity. In ion exchange process, arsenic ions are adsorbed on the active sites of the resin containing fixed counter ions. In the following section, the results obtained using some conventional technologies and techniques presently used are briefly reviewed. The technological processes include oxidation (active or passive) and adsorption on classical adsorbents, ion-exchange resins or colloidal particles formed *in situ*.

Aeration of water containing iron and subsequent storage can reduce arsenic concentration. Ahmed *et al.*<sup>14,15</sup> showed that more than 50% arsenic reduction could be achieved by sedimentation of tube-well water containing 380-480 mg/L  $\text{CaCO}_3$  and 8-12 mg/L of iron. A pilot study showed that the method was effective in reducing arsenic content of ground water to below 0.05 mg/L for influent water with arsenic concentration below 0.10 mg/L<sup>16,17</sup>. But, this process failed to reduce arsenic to the desired level in well-water treatment<sup>17</sup>.

Water treatment with coagulants such as alum, ferric chloride and ferric sulphate is effective in removing arsenic from water. Ferric salts have been more effective than alum for a wide range of pH. Irrespective of the coagulating agent, the pentavalent arsenic could be removed more effectively than trivalent one. Logsdon *et al.*<sup>18</sup> showed that at an influent concentration of 0.3 mg/L, over 95 percent of As(V) was removed with ferric sulphate coagulation and 83-90% with alum coagulation.

Many adsorptive media have been reported to be effective in arsenic removal. These are activated red mud<sup>19</sup>, hydrous ferric oxide<sup>20</sup>, iron oxide coated sand<sup>21</sup>, natural iron ore<sup>22</sup>, activated carbon<sup>23,24</sup>, zero-valent iron<sup>25</sup> and ferruginous manganese ore<sup>26</sup>. The iron coated sand is also an effective adsorbent for the

removal of arsenic from water. Iron coated sand containing 25 mg iron/g sand reduced arsenic concentration to zero from an influent of 300  $\mu\text{g/L}$  of arsenic<sup>27</sup>. Metal oxide based adsorbent has also proven to be good adsorbent, for the removal of arsenic by adsorption mechanism. Hybrid aluminas and composite metal oxides were able to treat 200-500 Bed Volume (BV) of water containing 550  $\mu\text{g/L}$  arsenic and 14 mg/L of iron<sup>14</sup>. Inorganic granular metal oxide based media consisting of aluminium oxide and manganese oxide removed arsenic to a level less than 10  $\mu\text{g/L}$ <sup>28</sup>. These adsorption capacity of activated alumina for the removal of As(III) was 4.08 mg As(III)/g activated alumina<sup>24</sup>.

Ion exchange process is similar to adsorption process. The ion exchange resins perform the function of adsorbent. The arsenic removal capacity of an ion exchange resin depends largely on the sulphate and nitrate contents of raw water as these species have high affinity to the basic anion resins<sup>29</sup>.

Bacteria capable of oxidizing ferrous ions can also reduce arsenic in water by adsorption of arsenic onto the colloidal iron hydroxide. These bacteria can oxidize As(III) to As(V) and thus all the inorganic forms of arsenic could be efficiently removed with biological iron oxidation<sup>30,31</sup>.

### Membrane technology for arsenic removal

Membrane technology is a promising one to remove arsenic from water. 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. Different types of membrane technologies, such as reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), microfiltration (MF), are employed in the removal of arsenic from water. In the next sections, different types of membrane processes used in the removal of arsenic from water are discussed.

### Reverse osmosis (RO)

Reverse osmosis is a well-established technology used for many years in water desalination. In this process, a very high (often close to 100%) rejection of low-molecular mass compounds and ions (total desalination) can be achieved. Moreover, the process

can easily be automated and controlled. Several studies have been performed on the removal of arsenic by RO technology.

Brandhuber and Amy<sup>32</sup> studied 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). The 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<sup>33</sup>, used the TFC-ULP RO membrane from Koch Membrane Systems. The total arsenic removal efficiency was around 99% for an influent concentration of 60 µg/L. Kang *et al.*<sup>34</sup> investigated the removal of arsenic from water by using two types of RO membranes, ES-10 and NTR-729HF (Nitro 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. Amy *et al.*<sup>35</sup> 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.*<sup>36</sup> studied arsenic removal from synthetic water and from surface water sources. Both As(V) and As(III) were effectively removed from water by RO. 99% rejection of As(V) was obtained with RO membrane. In another pilot scale study, the removal efficiencies was 96-99% for As(V) and 46-84% for As(III)<sup>37</sup>.

Considering the developing countries' situation, such as low annual income and low electric popularization, Oh *et al.*<sup>38</sup> studied the rejection of arsenic by using HR3155 membrane (Toyobo Co.,

Ltd) made of cellulose triacetate coupled with a bicycle 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<sup>38</sup>. Removal of arsenic by some commercial reverse osmosis membranes is summarized in Table 1.

### Nanofiltration (NF)

Nanofiltration (NF) membranes are usually applied to separate multivalent ions from monovalent ones. However, it is also possible to achieve a certain separation of ions of the same valence by selecting the proper membrane and operating conditions<sup>39</sup>. NF membranes are sometimes designated as "loose" RO membranes, since they provide higher water fluxes at lower trans-membrane pressures. These membranes are usually asymmetric and negatively charged at neutral and alkaline media. Therefore, 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<sup>40</sup>. 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 but at higher water flux through the membrane. A number of studies dealing with the removal of arsenic from drinking water have been performed by NF membranes and most of them have showed promising results.

Brandhuber and Amy<sup>32</sup> studied the rejection of arsenic by using three NF membranes-NF70 4040-B (Film Tec), HL-4040F1550 (DESAL) and 4040-

Table 1 — Rejection (%) of arsenic by RO membranes

Model	Supplier	Water source	Rejection (%)		Reference
			As(III)	As(V)	
TFC 4921	Fluid Systems	Ground water	63	95	[31]
TFC 4820-ULPT	Fluid Systems	Ground water	77	99	[31]
AG 4040	Desal	Ground water	70	99	[31]
4040 LSA-CPA2	Hydranautics	Ground water	85	99	[31]
TFC-ULP RO	Koch Membrane Systems	Ground water (USA)	Total As rejection 99%		[32]
ES-10	Nitto Electric Industrial Co. (Japan)	Distilled water	75%	95%	[32]
NTR-729HH	Nitto Electric Industrial Co. (Japan)	Distilled water	20-43%	80-95	[32]
DK2540F RO	Desal	Lake water	5	96	[34]
HR3155	Toyobo Co., Ltd	Ground water	55	95	[37]

UHA-ESNA (Hydranautics). The As(V) rejection was more than 95%, whereas the As(III) rejection was 20-53% for all the three membranes. In another experiment<sup>41</sup>, 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%.

Urase *et al.*<sup>42</sup> 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 [As(V)] and arsenite [As(III)]. ES-10 was able to reject 50-89% of As(III) and 87-93% of As(V).

Sato *et al.*<sup>43</sup> used three types of NF membranes to investigate the arsenic removal efficiency from synthetic and ground water. These commercial membranes are SE-10 (aromatic polyamide), NTR-7250 (polyvinyl alcohol), and NTR-729HF (polyvinyl alcohol), all 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.

Saitúa *et al.*<sup>44</sup> studied the removal of As(V) from synthetic solution and surface water with a spiral wound thin film composite polyamide membrane, 192-NF300 (Osmonic Inc). The rejection of As(V) by the NF-300 module was found to be 93-99% for retentate arsenic concentration in the range of 100-382 µg/L. Application of NF-300 membrane to surface water showed that the co-occurrence of

dissolved inorganic salts do not significantly influence the rejection of As(V). The mean rejection of As(V) was 95% while that of sulphate was 97%.

Arsenic removal from synthetic fresh water was also studied by Waypa *et al.*<sup>36</sup>. Both As(V) and As(III) were effectively removed from the water by tight NF membrane, NF-70 (Filmtec), over a wide range of operating conditions. 99% rejection could easily be obtained with NF 70 membrane.

The removal of arsenic from natural ground water with two types of nanofiltration membrane, NF270 and NFc (Filmtec), was also investigated<sup>45</sup>. The result showed that the As(V) rejections by NF270 and NFc were 99 and 96%, respectively. The rejection of arsenic by some commercial nanofiltration membranes is summarized in Table 2.

### Ultrafiltration (UF)

Ultrafiltration is a low pressure driven membrane operation in which macromolecules, colloids and solutes with molecular weight higher than few thousand Daltons are retained. 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 the UF with only pore size dependent sieving.

A negatively charged, thin film, composite sulphonated polysulphone UF membrane (Osmonics GM) was studied to investigate the removal of arsenic from water<sup>46</sup>. The influence of co-occurring divalent ions and natural organic matter (NOM) on arsenic rejection by charged membrane was explored. The presence of anions in feed solution resulted in a decrease in As(V) rejection — the divalent anions

Table 2 — Rejection (%) of arsenic by NF membranes

Model	Supplier	Water origin	Rejection (%)		Reference
			As(III)	As(V)	
NF70 4040-B	Film Tec (Dow Chemical)	Colorado River & State project water (USA)	53	99	[31]
HL-4040F1550	DESAL	Idem	21	99	[31]
4040-UHA-ESNA	Hydranautics	Idem	30	97	[31]
NF-45	Film Tec	Synthetic water	10	90	[40]
ES-10	Nitto-Denko Co. Ltd.	Ground water	50-89	87-93	[41]
ES-10	Nitto Electric Industrial Co., Japan	Synthetic water	80	97	[42]
NTR-729HF	Nitto Electric Industrial Co., Japan	Synthetic water	21	94	[42]
NTR-7250	Nitto Electric Industrial Co., Japan	Ground water	10	86	[42]
192-NF300	Osmonics Inc.	Pure water	-	93-99	[43]
		Volcan River	-	95	
NF70	Film Tec	Fresh water	99	99	[43]
NF270	Film Tec	Ground water(Osijek)	-	99	[44]
NFc	Film Tec	Ground water(Osijek)	-	96	[44]

showing greater influence than the monovalent ones. In presence of divalent cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ), As(V) rejection reduced almost to zero. This reduction in As(V) rejection was due to interaction of solutes and the membrane. These interactions may include the formation of ion pairs between counter ions and the fixed charge group in the membrane matrix, which locally neutralises the membrane charges. The result also showed that the presence of NOM may play an intriguing role in the rejection of As(V) by charged membranes. It is believed that high concentration of organics matter may improve arsenic rejection through the complexation of divalent ions whose presence in solution tends to reduce As(V) rejection. The effect of co-occurring inorganic solutes ( $\text{HCO}_3^-$ ,  $\text{HPO}_4^{2-}$ ,  $\text{H}_4\text{SiO}_4$  and  $\text{SO}_4^{2-}$ ) in feed water on the removal of As(V) and permeate flux was investigated by using a cationic surfactant cetylpyridinium chloride (CPC) and a flat sheet hydrophilic polyethersulfone (PES) ultrafiltration membrane (Millipore, Bedford, MA)<sup>47</sup>. 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  $\mu\text{g/L}$ . The results also showed that the presence of  $\text{HCO}_3^-$ ,  $\text{HPO}_4^{2-}$ , and  $\text{H}_4\text{SiO}_4$  species in feed water did not affect the arsenic removal efficiency (100%).

AWWARF<sup>35</sup> performed a series of bench-scale tests to investigate the effect of membrane charge on the removal of arsenic by using uncharged FV2540F UF membrane and negatively charged GM2540F UF membrane. The results indicated that the rejection of As(V) by FV2540F membrane was lower than that of As(V) by GM2540F membrane. As(III) rejection by both GM2540F and FV2540F was very low at neutral pH, but a better rejection was obtained by GM2540F at basic pH due to electrostatic interaction between arsenic ions and negatively charged membrane surface.

The effect of electrostatic force on arsenic removal efficiency by UF membrane was also investigated by using groundwater with low dissolved organic carbon (LDOC) and high dissolved organic carbon (HDOC)<sup>35</sup>. The arsenic removal efficiency was around 70% for HDOC groundwater, while for LDOC groundwater, it was around 30%. The natural organic matter (NOM) is adsorbed to the membrane surface

forming a negatively charged layer. The charge density of the layer depends on the concentration of the NOM in the solution. Higher NOM concentration leads to higher charge density in the layer adjacent to the membrane surface. Consequently, the negatively charged arsenic species are more effectively excluded.

### Microfiltration (MF)

Microfiltration is a pressure driven membrane operation in which particles are separated in the solvated size range of 0.02 to 10  $\mu\text{m}$  from a fluid mixture. The retention of particles and solutes occurs either by sieving or adsorption in the membrane matrix. The pore size of MF membrane is too large to remove dissolved or colloid arsenic species effectively. But MF membrane can remove particulate form of arsenic. Only a small part of arsenic in water is present in particulate form. Removal efficiency of arsenic by MF membrane can be increased by increasing the particle size of arsenic bearing particles.

Coagulation and flocculation processes increase the particle size of arsenic species and thus increase the removal efficiency of arsenic from water by MF membrane. Flocculation using ferric chloride or ferric sulphate followed by microfiltration using mixed esters of cellulose acetate and cellulose nitrate membrane with pore size of 0.22 and 1.2  $\mu\text{m}$  was used to investigate the removal efficiency of arsenic<sup>48</sup>. The results showed that flocculation followed by microfiltration was better than the flocculation followed by sedimentation for arsenic removal. The removal of arsenic by flocculation and microfiltration depends on the effectiveness of arsenic adsorption onto the ferric complexes present and on the rejection of the formed arsenic containing flocs by the membrane.

### Removal of arsenic as a function of operating parameters

The removal of arsenic by membranes is greatly influenced by operating parameters such as pressure, pH, solute concentration and temperature of the feed solution. Particularly, the rejection of arsenic is highly dependent on the pH of the feed water. Membranes with charged surface can remove arsenic effectively if the arsenic in water remains as anions. The charge of arsenic in water is greatly influenced by pH. In the following section, the effect of operating parameters on the rejection of arsenic from water is discussed.

### Removal of arsenic as a function of pressure

Very few researchers have studied the effect of applied pressure on the removal of arsenic by membrane process. However, it is expected that arsenic rejection increases with pressure due to higher water flux which results in lower arsenic concentration in the permeate water.

Sato *et al.*<sup>43</sup> investigated the effect of applied pressure on the performance of three NF membranes, ES-10, NTR-729HF and NTR-7250. The results showed that arsenic removal efficiency increased slightly with the increasing the applied pressure — an effect attributed to slightly higher increment rate in solvent flux than solute flux. The effect is much more pronounced for As(III) than for As(V). As(III) removal by ES-10, NTR-729HF and NTR-7250 increased from 62, 14 and 7% at 0.3 MPa to 82, 25 and 11% at 1.1 MPa, respectively. The increase of As(V) removal efficiency by all membranes was below 4.0% as the pressure increases from 0.3 to 1.1 MPa. Saitúa *et al.*<sup>44</sup> also showed that the rejection of As(V) by NF-300 membrane was not much sensitive to change in operating pressure between 310 and 724 kPa. Removal efficiency was around 94% over the transmembrane pressure of 310-724 kPa.

Removal efficiency of both As(V) and As(III), under low pressure range of 0.2-0.6 MPa, was investigated by using ES-10 nanofiltration membrane<sup>36</sup>. The removal efficiency was around 99% for arsenate and 55% for arsenite and the efficiency of arsenic increased slightly with the increase in applied pressure.

### Removal of arsenic as a function of pH

Both As(III) and As(V) occur in several protonated forms at neutral pH, the predominant species are  $\text{H}_3\text{AsO}_3$  for As(III), and  $\text{H}_2\text{AsO}_4^-$  and  $\text{HAsO}_4^{2-}$  for As(V). It means that As(V) exists as an anion at typical pH in natural water (pH 5-8), while As(III) remains as a neutral molecule at the same pH region. As(V) has negative charge at  $\text{pH} > 9.2$  (ref. 49). Hence, rejection of arsenic from water by charged membrane is largely affected by the pH of the feed solution. Figures 1 and 2 show the effect of pH on arsenic species and the rejection of As(V) and As(III) by a negatively charged membrane. Data are collected from elsewhere<sup>42</sup>. Kang *et al.*<sup>34</sup> investigated the effect of pH on the removal of arsenic using two types of reverse osmosis membranes, ES-10 and NTR-729HF. It was observed that the removal of arsenite was lower than that of arsenate over the pH range of 3-10.

The removal of arsenite by ES-10 and NTR-729HF was around 77% and 20% at pH 3, 5, 7 and 90% and more than 40% at pH 10. The removal of arsenate by ES-10 was more than 95% at all the pH range investigated while that of arsenate by NTR-729HF increased from 80% at pH 3 to around 90% at pH 5-10.

In nanofiltration, the separation of both the As(V) and As(III) species as a function of pH was reported by Vrijenhoek *et al.*<sup>41</sup>. The separation of As(III) species by the NF-45 was unaffected by change in pH over the range of 4-8, because the As(III) species remain uncharged at these pH values (i.e., in  $\text{H}_3\text{AsO}_3$  form). The rejection of As(III) at this pH range was less than 10%. The rejection of As(V) species, however, increased significantly from 25% at pH 4 to more than 80% at pH 9 due to change of As(V) species from monovalent ions ( $\text{H}_2\text{AsO}_4^-$ ) to divalent ions ( $\text{HAsO}_4^{2-}$ ) with increasing pH. Divalent ions were rejected by the NF-45 membrane at a much higher rate as compared to monovalent ions due to larger hydrated radii of divalent ions as well as due to greater Donnan exclusion. The effect of arsenic removal as a function of pH by using nanofiltration membrane, ES-10 was also investigated by Urase

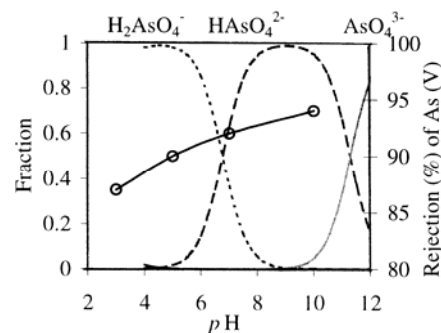


Fig. 1 — Fraction of  $\text{H}_2\text{AsO}_4^-$  species and the rejection of As(V) as a function of pH

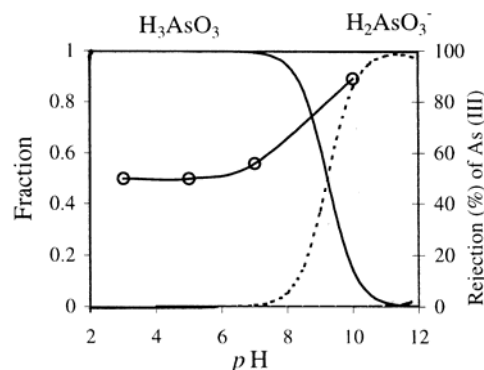


Fig. 2 — Fraction of  $\text{H}_3\text{AsO}_3$  species and the rejection of As(III) as a function of pH

*et al.*<sup>42</sup>. With the decrease of *pH* from 10 to 3, the rejection of arsenite As(III) decreased from 89 to 50%, the rejection of arsenate, on the other hand, decreased from 93 to 87%. The rejection characteristics of both As(III) and As(V) as a function of *pH* using three low pressure nanofiltration membranes, ES-10, NTR-729HF and NTR-7250, was investigated by Oh *et al.*<sup>50</sup>. The *pH* dependence of the removal efficiency of arsenite and arsenate observed for ES-10 was similar to that done by Urase *et al.*<sup>42</sup> for the same membrane. The rejection of arsenite by NTR-729 and NTR-7250 increased from 50% and 30% at *pH* 7 to 80% and 60% at *pH* 10, respectively. The removal of arsenate by both NTR-729 and NTR-7250 was over 90% at *pH* 10 while at *pH* 3 the removal efficiency was 60% and 44%, respectively. Košutić *et al.*<sup>45</sup> studied the rejection of arsenic as a function of *pH* using two types of nanofiltration membranes, NF-270 and NFc. The results indicated only the minor increase in the rejection with the increase of *pH* values.

Brandhuber and Amy<sup>46</sup> found that the removal of As(V) by a negatively charged ultrafiltration membrane exceeded the removal of As(III) at all investigated *pH* values (*pH* 2-10). The magnitude of arsenic rejection by the membrane increased with the increase in the *pH*. The trend of increasing rejection corresponds to the degree of deprotonation of the arsenic anion; the divalent As(V) anion, ( $\text{HAsO}_4^{2-}$ ), being better rejected than the monovalent As(V) anion, ( $\text{H}_2\text{AsO}_4^-$ ), and the monovalent As(III)  $\text{H}_2\text{AsO}_3^-$  being better rejected than the uncharged  $\text{H}_3\text{AsO}_3$  molecule. The rejection of As(V) increased with the increase of *pH* from around 13% at *pH* 2 to more than 80% at *pH* 10 while the rejection of As(III) was less than 10% at *pH* 3 and was around 15% over the *pH* range 4, 6, 8 and 10. The rejection of As(III) increased sharply from 15 to 40% as the *pH* increased from 10 to 11.

#### Removal of arsenic as a function of solution concentration

Using nanofiltration membranes, Vrijenhoek and Waypa<sup>41</sup> observed that the rejection of As(V) by NF-45 increased from 45 to 85% as the feed arsenic concentration increased from 10  $\mu\text{g/L}$  to more than 100  $\mu\text{g/L}$ . The separation of As(III), however, decreased with increasing feed concentration. The increase in As(V) separation and the decrease in As(III) separation could be explained as follows: At the *pH*  $\approx 8$ , the As(III) species were uncharged (in the

form of  $\text{H}_3\text{AsO}_3$ ). Transport of uncharged species through a porous membrane follows diffusion and convection mechanism as described by the Spiegler and Kedem membrane transport model<sup>51</sup>. The Donnan exclusion is typical for NF membranes and does not work for uncharged species. As in the bulk solution As(III) concentration increases, both diffusion and convection of the As(III) species increase. Consequently, the separation of As(III) decreases with increasing bulk solution concentration. Saitúa *et al.*<sup>44</sup> showed that the rejection of As(V) increased as the retentate arsenic concentration increased. The transport of As(V) through the membrane did not increase proportionally with increase in retentate concentration and consequently an increase in rejection was observed.

Brandhuber and Amy<sup>46</sup> also studied the removal of As(V) as a function of solution concentration in ultrafiltration. The rejection of As(V) by UF membrane was reduced when the bulk As(V) concentration was increased. The arsenic removal efficiency by UF membrane was found to be dependent on the feed water arsenic concentration.

#### Removal of arsenic as a function of temperature

Saitúa *et al.*<sup>44</sup> showed that the rejection of As(V) by a nanofiltration membrane was not much sensitive to the change in operating temperature. Brandhuber and Amy<sup>46</sup> reported that a temperature variation of 20°C had little effect on the rejection of arsenic by ultrafiltration membrane. The rejection of arsenic was 82% at a temperature of 20°C and 76% at the temperature of 40°C. The decrease in rejection was expected to occur since the diffusivity of arsenic increases with temperature which in turn increases the diffusive transport of arsenic across the membrane.

#### Comparative discussion on the conventional and membrane technologies

Arsenic(V) is more effectively removed than As(III) by both the conventional and membrane technologies. Arsenic in the ground water exists mostly as As(III). Therefore, for both the technologies, pre-oxidation of As(III) to As(V) is an essential step for better removal. Most of the arsenic removal system based on the conventional processes requires secondary treatment system in order to reduce the arsenic concentration to the MCL standard. Large amount of chemical reagents such as alum or ferric chloride/ferric sulphate is used for coagulation processes. These processes produce a large volume of

sludge, which requires further treatment before disposal. When the sludge is piled, a large amount of leachate with high concentration of arsenic is produced. To the contrary, removal of arsenic by microfiltration and ultrafiltration requires less amounts of coagulants and flocculants to increase the particle size of the arsenic and hence the sludge produced is not so large in volume compare to conventional method. But, the removal of arsenic by NF membranes and RO membranes requires no chemical reagent and no sludge is produced in these processes (Table 3).

Adsorption is a process in which materials having strong affinity for arsenic are used as adsorbents. The adsorbent are very expensive, but not selective to arsenic only. Other anions present in the water are also adsorbed on the adsorbent. As a result, the capacity of the adsorbents is exhausted ineffectively. Ion exchange resin also exchanges other competing anions ( $\text{SO}_4^{2-}$ ,  $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$  etc.), which are present in large amount in water. These anions have almost no effect on removal of arsenic by membrane technologies. Anions are not adsorbed on the surface of the membrane. Membranes remove anions by size exclusion and Donnan exclusion. So, the efficiency of membrane process is not significantly affected by the anions present in the water. The adsorption process requires chemical for regeneration and the regenerative fluid contains high concentration of arsenic. In membrane process, chemical for regeneration is generally not required. Membrane is usually back washed to prevent clogging or fouling.

Water obtained from the conventional technologies contain fine particle that cannot be removed by sedimentation or gravity settling. Therefore, conventional filter is required to remove the fine particles from the water. The membrane technology removes not only arsenic but also portions of other dissolved minerals. It also retains microorganism and

thus diminishes the harmful disease. So, disinfection is not a separate step in membrane technology. Conventional technologies cannot remove pathogens and they require disinfection of the treated water.

The conventional treatment technology requires large area for the building up of a plant, whereas membrane technology based plants are highly compact. The fouled membranes are easily replaced by new membranes. The membranes process can be easily scaled up only by adding more membrane modules and the performance of the membranes is maintained by controlling the parameters of the feed solution. Therefore, the maintenance of membrane technology is simple. The membrane technology (RO, NF, UF, and MF) is a promising method to remove arsenic from water. This technology completely satisfies environmental requirements. RO and NF membrane processes can treat dissolved arsenic better than UF and MF membrane processes.

Operating conditions, such as membrane material, water source, temperature, pressure, pH value of solution and chemical compatibility must be considered during operation of a membrane plant. This is because, these parameters affect the arsenic removal efficiency by membrane technology. No one-membrane material, membrane type and membrane process can be used in all the possible applications. Therefore, different membrane materials type and membrane processes should be evaluated to select the optimum for each situation. A comparison between conventional technology and membrane technology for arsenic removal is given in Table 3.

The water cost depends on capital cost and operating cost of the system. There are four main components that will be considered in capital cost calculation, which are civil cost, mechanical engineering cost, membrane installation cost and electro technical cost. The cost of the first three components decreases as the pressure increases. The

Table 3 — A comparison between conventional technology and membrane technology

Conventional technology	Membrane technology
Pre-oxidation of arsenic is required for better removal	Pre-treatment is required for better removal
Huge amount of chemical reagents is	Almost no chemical reagent is necessary
High volume of sludge is produced	No sludge is produced
Arsenic leached out from sludge	Arsenic is concentrated in the retentate
Adsorption process requires chemical for regeneration and the regenerative fluid contains As	No chemical for regeneration is required
Require high area	Require low area
Maintenance cost is high	Maintenance cost is low
Not easy automation and control	Easy automation and control
Require conventional filter for the removal of fine particles	Require no conventional filter



decrease in pressure will cause a decrease in flux rate across each membrane module. Therefore, the number of modules required to achieve a production target will be increased. Civil cost (such as building for a membrane system), mechanical engineering cost (related to pump installation, valve, filter and plumbing system) and membrane installation cost (related to the number of membrane modules installed) are proportional to the installation size of a membrane system. It means that the greater the number of modules required, the greater the installation system size is. Therefore, all of the above three cost components increased with the decrease of operating pressure. On the other hand, electro technical cost increases as the pressure increases. A total investment cost is a total of all components related to installation size and system power. Therefore, there is one offset point, which balances all four capital cost components. The offset point gives a pressure to the optimal separation system. Operating cost required to run an operation of a separation system consists of six main components as depreciation, energy, chemical, maintenance, quality control and operation of installation costs. All cost components except energy cost are a function of a number of membrane modules. Therefore, these costs are decreased as operating pressure increases. As for the capital cost, there is one cost component that increases with pressure — this is energy usage cost. Energy usage cost is proportional to operating pressure, that is, the increasing pressure will need more energy in order to run an operation system. The balance between these components will contribute to an optimal condition. Bruggen *et al.*<sup>52</sup> studied the economic side of the implementation of nanofiltration membrane for the treatment of groundwater and found that nanofiltration is a valuable option for groundwater treatment. The total cost of nanofiltration treatment per cubic meter water was 0.17 €.

### Conclusion

On the basis of the reports detailed above, the following conclusions can be drawn:

- (i) Both the conventional and membrane technology could be applied for arsenic removal up to the MCL.
- (ii) RO and NF membrane technology is found to be highly effective for the purpose, while UF and MF membrane technology require a preparatory step as colloid formation for effective removal.
- (iii) Disinfection is an essential step for the conventional technology, whereas in membrane technology, the membrane itself acts as a barrier to microorganisms.
- (iv) Easy maintenance and easy handling make the membrane technology a very promising one from small scale to large scale operation.

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### Reference

- 1 Tchounwou P B, Wilson B & Ishaque A, *Rev Environ Health*, 14(4)(1999) 211.
- 2 Kinniburgh D G & Smedley P L, *British Geological Survey*, 2 (2001).
- 3 [http://www.who.int/water\\_sanitation\\_health/dwq/arsenic3/en/](http://www.who.int/water_sanitation_health/dwq/arsenic3/en/).
- 4 United Nations Synthesis Report on Arsenic in drinking water, WHO, (2003).
- 5 Shih M C, *Desalination*, 172 (2005) 85.
- 6 Meenaskshi & Maheshwari R C, *Asian J Water Environ Poll*, 3(1) (2006) 133
- 7 Cheng C R, Liang S, Wang H C & Beuhler M D, *AWWA*, 86(9) (1994) 79.
- 8 Hering J G, Chen P Y, Wilkie J A, Elimelech M & Liang S, *AWWA*, 88(4) (1996) 155.
- 9 Hering J G, Chen P Y, Wilkie J A & Elimelech M, *J Env Eng ASCE*, 123(8) (1997) 800.
- 10 Kartinen E O & Martin C J, *Desalination*, 103 (1995) 79.
- 11 Shen Y S, *AWWA*, 65(8) (1973) 543.
- 12 Joshi A & Chaudhury M, *J Env Eng ASCE*, 122(8) (1996) 769.
- 13 Pierce M L & Moore C B, *Water Res*, 16 (1982) 1247.
- 14 Ahmed M F & Rahaman M M, Water supply and sanitation – low income urban Communities (International Training Network (ITN) Centre, BUET), 2000.
- 15 Ahmed M F, Jalil M A, Ali M A, Hossain M D & Badruzzaman A B M, in *Bangladesh Environment-2000, Bangladesh Poribesh Andolon*, edited by M F Ahmed (Bangladesh University of Engineering & Technology and United Nation University), 2000, 177.
- 16 Sarker A R & Rahman O T, in *Technologies for Arsenic Removal from Drinking Water*, edited by M F Ahmed (Bangladesh University of Engineering & Technology and United Nation University), 2001, 201.
- 17 BAMWSP, DFID and Water Aid Bangladesh, Rapid assessment of household level arsenic removal technologies, phase-I and phase-II, Final report (WS Atkins International Limited), 2001.
- 18 Logsdon G S, Sorg T J & Symons J M, *Removal of heavy metals by conventional treatment*, paper presented at the 16th Water Quality Conference- Trace Metals in Water Supplies: Occurrence, Significance, and Control, University of Illinois, Urbana, 12-13 Feb, 1974.
- 19 Altundogan H S, Altundogan S, Tumen F & Bildik M, *Waste Management*, 22(3) (2002) 357.

- 20 Wilkie J A & Hering J G, *Colloids Surfaces A*, 107 (1996) 97.
- 21 Gupta V K, Saini V K & Jain N, *J Coll Int Sci*, 288 (2005) 55.
- 22 Zhang W, Singh P, Paling E & Delides S, *Miner Eng*, 17 (2004) 517.
- 23 Lorenzen L, van Deventer J S J & Landi W M, *Miner Eng*, 8(4-5) (1995) 557.
- 24 Manjare S D, Sadique M H & Ghosal A K, *Environ Technol*, 26 (2005) 1403.
- 25 Bang S B, Korfiatis G P & Meng X G, *J Hazardous Mater*, 121 (2005) 61.
- 26 Chakravarty S, Dureja V, Bhattacharyya G, Maity S & Bhattacharjee S, *Water Res*, 36 (2002) 625.
- 27 Ali M A, Badruzzaman A B M, Jalil M A, Hossain M D, Hussainuzzaman M M, Badruzzama M, Mohammad O I & Akter N, in *Technologies for Arsenic Removal from Drinking Water*, edited by M F Ahmed (Bangladesh University of Engineering & Technology and United Nation University), 2001, 99.
- 28 Senapati K & Alam I, in *Technologies for Arsenic Removal from Drinking Water*, edited by M F Ahmed (Bangladesh University of Engineering & Technology and United Nation University), 2001, 146.
- 29 Ahmed M A, in *Arsenic Contamination: Bangladesh Perspective*, edited by M F Ahmed (ITN-Bangladesh: BUET, Dhaka-1000, Bangladesh), 2003, 357.
- 30 Katsoyiannis I A & Zouboulis A I, *Water Res*, 38(1) (2004) 17.
- 31 Katsoyiannis I A, Zouboulis A I, Althoff H & Bartel H, *Chemosphere*, 47 (2002) 325.
- 32 Brandhuber P & Amy G, *Desalination*, 117 (1998) 1.
- 33 Koch Membrane Systems Inc. *Membrane Technol*, 140 (2001) 10.
- 34 Kang M, Kawasaki M, Tamada S, Kamei T & Magara Y, *Desalination*, 131 (2000) 293.
- 35 Amy G L, Edwards M, Benjamin M, Carlson K, Chwirka J, Brandhuber P & McNeill L & Vagliasindi F, *AWWARF*, 1998.
- 36 Waypa J J, Elimelech M & Hering J G, *AWWA*, 89(10) (1997) 102.
- 37 Ning R Y, *Desalination*, 143 (2002) 237.
- 38 Oh J I, Yamamoto K, Kitawaki H, Nakao S, Sugawara T, Rahman M M & Rahman M H, *Desalination*, 132 (2000) 307.
- 39 Lhassani A, Rumeau M, Benjelloun D & Pontie M, *Water Res*, 35 (2001) 3260.
- 40 Levenstein R, Hasson D & Semiat R, *J Membrane Sci*, 116 (1996) 77.
- 41 Vrijenhoek E M & Waypa J J, *Desalination*, 130 (2000) 265.
- 42 Urase T, Oh J & Yamamoto K, *Desalination*, 117 (1998) 11.
- 43 Sato Y, Kang M, Kamei T & Magara Y, *Water Res*, 36 (2002) 3371.
- 44 Saitúa H, Campderrós M, Cerutti S & Padilla A P, *Desalination*, 172 (2005) 173.
- 45 Košutić K, Furač L, Sipos L & Kunst B, *Sep Puri Technol*, 42 (2005) 137.
- 46 Brandhuber P & Amy G, *Desalination*, 140 (2001) 1.
- 47 Ergican E, Gecol H & Fuchs A, *Desalination*, 181 (2005) 9.
- 48 Han B, Runnells T, Zimbron J & Wickramasinghe R, *Desalination*, 145 (2002) 293.
- 49 Ferguson J F & Gavis J, *Water Res*, 6 (1972) 1259.
- 50 Oh J I, Lee S H & Yamamoto K J, *Membrane Sci*, 234 (2004) 167.
- 51 Spiegler K S & Kedem O, *Desalination*, 1 (1966) 311.
- 52 Van der Bruggen B, Everaert K, Wilms D & Vandecasteele C, *J Membrane Sci*, 193 (2001) 239.