



# Direct As(III) removal from brackish groundwater by vacuum membrane distillation: Effect of organic matter and salts on membrane fouling



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## ABSTRACT

Arsenic contamination of shallow ground water is being one of the biggest health threats in the world, particularly in Asia. In this work, vacuum membrane distillation (VMD) was proposed as an advanced solution for arsenic removal. The case study concerned arsenic contamination in Vietnam, where drinking water resources present not only high arsenic concentration (1–3050 ppb) but also high salinity (5–15 g/L). For that reason, synthetic brackish solutions containing NaCl (10 g/L) and As(III) (at different concentrations – between 300 and 2000 ppb) were used as feed solution in this study. As a result, VMD was capable to satisfy As(III) rejection to meet the required standard in the permeate (MCL = 10 ppb). Salt rejection was also very high (>99.5%). The As(III) rejection rate was always stable at high level (>98.5%), irrespective of high feed As(III) concentrations (up to 2000 ppb). With VMD, a pre-oxidation step was not necessary to convert As(III) into As(V), as it is the case for other conventional treatment processes. Furthermore, no effect of organic matter (humic acid) and calcium on membrane scaling and fouling phenomena was observed at the given concentration in this study.

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## 1. Introduction

The 21st century has witnessed big affords of environmentalists in facing with deficiency of clean water through removing arsenic contamination in drinking water and desalinating from the sea water. As an ubiquitous element in the earth's crust, arsenic can be found everywhere in the world at different levels, depending on regional geological structure or its originating sources. Unfortunately, this element is very difficult to be detected in water as it is a tasteless, invisibility and odourless element. As a consequence, more than 150 million people over 105 countries around the world, especially Bangladesh, Argentina, India, Mexico, Mongolia, Thailand and Taiwan are being infected by acute and chronic exposure of arsenic via drinking water [22,4]. Long-term of inorganic arsenic exposure can cause severe health problems to human including skin lesions, such as: hyperkeratosis and pigmentation changes, diabetes, circulatory disorders and cancer of bladder, lung, kidney and skin [16]. For that reason, recently, authorities have taken a more stringent attitude to arsenic in the environ-

ment; World Health Organization (WHO) and US Environmental Protection Agency (USEPA) guidelines fixed the new standard limit for arsenic in drinking water to 10 ppb [26,24].

Parallel to this sanitary environmental issue, desalination from seawater to produce drinking water became quite popular in many countries. Under impacts of climate change, saline intrusion of seawater to surface and groundwater is becoming an urgent issue in many countries in the world, especially in South-east Asia countries. For example in particular case of Vietnam, more than one million of water wells with high concentration of arsenic are in use at both MeKong Delta ([As] = 1–1610 ppb) and Red River Delta ([As] = 1–2050 ppb), equivalent to 13.5% of Vietnamese population are in hazard poisoned by arsenic [21,8]. These water sources are being intruded by seawater, leading to both arsenic contamination and high salinity. It can be foreseen that desalination and arsenic removal in the brackish groundwater is going to be one of the main issues for the environmentalists in the near future. This is also the main objective of this study to find out an innovative, advanced treatment technology to satisfy both these requirements.

In nature, arsenic exists in both organic and inorganic forms. The organic species (monomethylarsenic and dimethylarsenic) are abundant in seafood, less harmful to the human health and readily eliminated by the body while the inorganic forms are more

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### Nomenclature

$J_{H_2O}$	molar flux of water, mol s <sup>-1</sup> m <sup>-2</sup>	$M_{H_2O}$	molar mass of water, kg mol <sup>-1</sup>
$P_m^*$	vapour pressure of pure water at the membrane's conditions, Pa	$K_M$	Knudsen permeability, s mol <sup>1/2</sup> m <sup>-1</sup> kg <sup>-1/2</sup>
$P_p$	partial pressure of water in the permeate side, Pa	$\chi_{H_2O}$	mole fraction of water
$T_f$	temperature in the feed bulk, K	$\alpha_{H_2O}$	activity coefficient of water
$T_m$	temperature at the membrane surface, K		

prevalent in water and considered more toxic. Therefore, most of studies normally focus on inorganic arsenic form. Inorganic arsenic has several valence states (-3, 0, +3 and +5), the +3 and +5 states are the most abundant found in a variety of minerals and natural waters. Depending on local redox condition, arsenic is found as arsenite As(III) form in anoxic condition or as arsenate form As(V) in aerobic condition, respectively [2].

Several detailed reviews of arsenic removal technologies have been published by Jekel [10], Murcott [15], US Environmental Protection Agency [25] and recently by Jain and Singh [9], Kowalski [12] or Pal [17]. In these reviews, a variety of removal techniques of arsenic from aqueous media applied in both laboratory and field conditions have been listed. Generally, these various technologies available for removal of arsenic from contaminated water are based mainly on six principles, including: oxidation and filtration, biological oxidation, co-precipitation, adsorption, ion exchange and membrane technology. Depending on particular conditions, these mentioned technologies can be applied to remove As to meet the standard. Adsorption and precipitation processes are often applied in rural area owing to its applicability. However, the main drawback of adsorption processes is the disposal of both spent media and the wastewater produced during regeneration/cleaning of the column and adsorbed media. Meanwhile, disadvantages of coagulation/precipitation processes are sludge disposal and post-treatment. Pressure-driven membrane filtration can be a promising option for both As removal and desalination. The negatively charged membranes used in most of the reported studies allowed a higher rejection obtained with anionic components, like As(V), rather than with fully protonated As(III). As a result, oxidation of As(III) to convert into As(V) is also required. However, oxidizing agents, like chlorine, could damage the membrane material. Besides that, typical disadvantages of using membrane processes are the higher cost of the treatment plants due to the membrane itself. Finally, membrane fouling remains the major drawback of such system while handling of its retentate is still a question.

With the aim at reducing the chemical cost for pre-oxidation to convert As(III) into As(V) as observed in most of published studies, this study will mainly focus on direct treatment of As(III) in order to find out an advanced solution, which can be applied to treat directly arsenic in the water, irrespective of its forms.

In this study, vacuum membrane distillation (VMD) technology – one of four variants of membrane distillation (MD) process – was introduced as the key possible process to solve the problem. MD is a thermally-driven separation process, in which only volatile compounds are able to pass through a porous hydrophobic membrane owing to partial pressure difference through the membrane. Then, water could be purified not only from arsenic, but also from salt as they are both non-volatile compounds. VMD technology has been recognised as an alternative for seawater desalination and bench-scale tests have successfully demonstrated the potential of this technology [27,20,13]. The main drawback of this process is the relatively high energy requirement linked to the need to heat the feed water. This can be solved by the use of a renewable source such as solar energy to provide the heat energy required.

Concerning arsenic removal by membrane distillation, few studies have been performed in recent years mainly with direct contact membrane distillation (DCMD) [19,18]; [28]. By using the self-made PVDF membranes (0.15 µm) for removing As(III) and As(V) in synthetic groundwaters, [19] obtained permeate fluxes of 7.60 and 7.50 kg m<sup>-2</sup> h<sup>-1</sup>, irrespective of increase of feed concentration. Permeate arsenic concentration was always under the maximum contaminant limit (MCL = 10 ppb) until the feed As(III) and As(V) concentrations reached 40 ppm and 2000 ppm, respectively with over 99.95% of removal efficiencies for both types of arsenic. A 250 h experiment for As(III) rejection showed this DCMD configuration (with PVDF membrane) had a stable As(III) rejection. SEM analysis of the membrane before and after experiment showed that membrane morphology has been changed slightly, but not for its permeability and salt rejection. Another study with same DCMD configuration with different types of membranes (PTFE 0.22 µm and PP 0.22 µm) was applied for treatment of real arsenic-contaminated groundwaters collected from some arsenic-affected areas in India by [18]. The highest permeate flux obtained was 49.80 kg m<sup>-2</sup> h<sup>-1</sup> at a circulation velocity  $v = 0.052$  m s<sup>-1</sup>;  $T_f = 60$  °C and  $T_p = 21$  °C, respectively. No arsenic was detected in the permeate after 4 days of operation (12 h per day). An average flux decline of 12% was observed for all membranes when arsenic concentration was increased from 0 to 1200 ppb without distinction between As(III) and As(V). However, no experiment on membrane fouling was mentioned regardless of presence of Fe<sup>total</sup> (1.2–1.8 ppm), Ca<sup>2+</sup> (102.5–110.3 ppm), Na<sup>+</sup> (26–32 ppm), in the feed solution. Higher permeate achievement with same DCMD configuration for recovery potable water from As-contaminated saline ground water was obtained by [28]. The highest permeate flux was 90–95 kg m<sup>-2</sup> h<sup>-1</sup> for both PP 0.45 µm and PTFE 0.22 µm, respectively at the operating conditions ( $T_f = 80$  °C,  $T_p = 20$  °C, salt concentration = 1000–10,000 ppm, [As] = 10–400 ppb). No identification was given of what kind of salt was present in the groundwater. The results showed influence of salt concentration to the permeate flux with 5% reduction of permeate flux when salt concentration was increased from 1000 to 10,000 ppm (rejection efficiency of 99%). A minimal decrease of flux could be observed also when As concentration increased from 10 ppb to 350 ppb, but no distinction between As(III) and As(V) was mentioned. The highest arsenic concentration in permeate was 0.17 ppb. A 12-h operation was carried out to evaluate the effect of inorganic fouling on the membranes; but neither fouling nor flux decline was observed during this time.

Concerning vacuum membrane distillation (VMD), this configuration was applied by Criscuoli et al. [1] for treating pure water containing arsenic, both in the trivalent and pentavalent forms by using both PP (0.2–0.45 µm) and PVDF (0.2 µm) commercial membranes. Operating conditions were fixed at low feed temperature (20–40 °C) with  $P_p = 10$  mbar and  $Re = 1700$ . For synthetic feed solutions containing a maximal concentration of 1 ppm for each arsenic form, no arsenic was detected in the permeate. Trans-membrane flux was strongly affected by the feed temperature and neither depended on the arsenic contents nor on the arsenic forms. The highest fluxes ranged between 3 and 12.5 kg m<sup>-2</sup> h<sup>-1</sup>

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