



Potable water recovery from As, U, and F contaminated ground waters by direct contact membrane distillation process

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ABSTRACT

In this study, the feasibility of the direct contact membrane distillation (DCMD) process to recover arsenic, uranium and fluoride contaminated saline ground waters was investigated. Two types of membranes (polypropylene, PP; and polytetrafluoroethylene, PTFE) were tested to compare the permeate production rates and contaminant removal efficiencies. Several experiments were conducted to study the effect of salts, arsenic, fluoride and uranium concentrations (synthetic brackish water with salts: 1000–10,000 ppm; arsenic and uranium: 10–400 ppb; fluoride: 1–30 ppm) on the desalination efficiency. The effect of process variables such as feed flow rate, feed temperature and pore size was studied. The experimental results proved that the DCMD process is able to achieve over 99% rejection of the salts, arsenic, fluoride and uranium contaminants and produced a high quality permeate suitable for many beneficial uses. The ability to utilize the low grade heat sources makes the DCMD process a viable option to recover potable water from a variety of impaired ground waters.

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1. Introduction

The need for freshwater can never be overstressed. Water is an essential natural source which is often considered to be available abundantly and free of cost [1]. Increasing withdrawals of freshwater due to population explosion and industrialization from current dwindling surface water sources have led the populations around the world to depend on the ground water sources for domestic and agricultural uses. Groundwater, in many parts of the world, is not suitable for direct consumption due to high salt concentrations and in some cases due to naturally occurring hazardous contaminants such as arsenic, fluoride and uranium [1–3]. Excess quantities of these chemicals in the groundwater are toxic and often accompanied by high salt concentrations requiring severe treatment before they are suitable for human consumption and most other uses. For instance: two-thirds of the continental United States including New Mexico has large volumes of saline water sources. The total volume of ground water in aquifers in New Mexico is estimated to be 20 billion acre-feet, however, 75% of the groundwater is too saline (10,000–35,000 ppm) for most uses and the remaining 25% of the ground water contains dissolved concentrations of lower than 2000 mg/L. The groundwater quality is easily affected by human activities especially in the areas with shallow aquifers. The potential contributors for the groundwater contamination are oil and

gas producing and refining areas in southeastern and northwestern parts of the state; uranium and copper mining and milling sites [4]. Concentrations of dissolved arsenic in ground-water in alluvial basins of Arizona commonly exceed 50 $\mu\text{g/L}$ and reach values as large as 1300 $\mu\text{g/L}$ [3]. In the United States, elevated concentrations of naturally occurring uranium are found in ground water in the Colorado Plateau, Western Central Plateau, Rocky Mountain System Basin, and Range, and Pacific Mountain System [5]. There are over 20 developed and developing countries including USA which have unacceptable concentrations of arsenic and fluorides in the groundwater [1,6–8].

Arsenic originates from the abundant natural source of earth's crust and is a toxic chemical which can cause lung and skin cancer, gastrointestinal disorders, muscular weakness, and loss of appetite. The EPA revised maximum contaminant level (MCL) for arsenic is 0.01 mg/L [8,9]. Fluoride in ground water is the result of aluminum production, phosphate fertilizers, which may contain up to 4% fluorine. Allowable limit for fluoride concentration in drinking water is 1.5 mg/L [10]. Fluorides in excess concentrations can lead to dental and skeletal fluorosis, and finally resulting in paralysis or crippling fluorosis [1]. Uranium originates from rocks and mineral deposits and found in most drinking water sources in the form of three isotopes: U-238 (over 99% by weight), U-235, and U-234. Due to varying amounts of each isotope in the water, the ratio of uranium concentration ($\mu\text{g/L}$) to activity (pCi/L) varies with drinking water sources from region to region. Based on considerations of kidney toxicity and carcinogenicity, EPA proposed a uranium MCL of 20 $\mu\text{g/L}$ in 1991 (corresponding to 30 pCi/L based on a mass/activity

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ratio of 1.5 pCi/ μg); the final rule was set at 30 $\mu\text{g}/\text{L}$ in December 2000 after the conversion factor was revised to 1 pCi/ μg [11,12]. Uranium concentrations in excess of 20 $\mu\text{g}/\text{L}$ have been reported in groundwater from parts of New Mexico, USA [13], and central Australia [14].

Currently available technologies for removal of arsenic, fluoride and uranium are based on physical and chemical mechanisms. The techniques include (a) coagulation with lime, alum, ferric hydroxide, ferric sulfate, sodium sulfate followed by flocculation, sedimentation and filtration; (b) adsorption on activated carbon; and (c) ion exchange, and (d) reverse osmosis [9,15,16]. These technologies have the following contaminant removal efficiencies: *arsenic*: coagulation/filtration (95%); lime softening (>90% at pH=10.5); ion exchange (95%); reverse osmosis (>95%); and activated alumina (95%) [17]; *fluoride*: adsorption (90%), coagulation/filtration followed by lime softening (18–33%); ion exchange (90–95%); and reverse osmosis (>95%) [1]. *Uranium*: coagulation/filtration (80 to >95%) depending on the pH; lime softening (85–99%); anion exchange (90–100%); reverse osmosis (90–99%); and activated alumina (~90%) [12,18,19].

Although the above mentioned processes are efficient in treating variety of impaired waters, they cannot offer chemical saving, energy and cost efficient solution to treat brackish waters contaminated with arsenic, fluoride and uranium and create sludge or solid wastes concentrated with contaminants. Membrane distillation process, a non-conventional technology, can be a feasible alternative to treat the contaminated high saline ground waters. The process operates in the temperature range of 40–80 °C and can be powered by waste process heat or low grade renewable heat sources. This process can be particularly attractive if the ground water is saline and reasonably at high temperatures, as the geothermal waters serve both purposes of feed and heat source. The concentrated brine along with the contaminants can be returned to the groundwater source after the recovery of freshwater without causing any adverse surface environmental effects. Concentrations as high as 10,000 mg/L total dissolved solids (TDS) may be re-injected to the groundwater wells but this number varies according to the federal and state regulations.

The objective of this study is to demonstrate the feasibility of membrane distillation process application for treating arsenic, fluoride and uranium contaminated high saline ground waters. Some research has been conducted on the feasibility of membrane distillation technology to remove arsenic contaminated salt water in the past [20–22]. However, fluoride and uranium removal associated with saline ground waters has not been reported to our knowledge. Here, we present the results of the uranium and fluoride removal efficiencies of the membrane distillation process in addition to arsenic removal. A flat sheet direct contact membrane (DCMD) module was operated with a low grade heat source and synthetic brackish water with arsenic, fluoride and uranium compounds. Thermal analysis of the DCMD process, experimental studies to determine the effect of contaminant concentrations, process variables such as feed flow, temperature and pore size are presented.

2. Membrane distillation (MD) process

Membrane distillation is a separation process resulting from simultaneous mass and heat transfer phenomena across a hydrophobic microporous membrane. Direct contact membrane distillation (DCMD), one of the MD configurations, is the most commonly used process mode due to its simplicity in structure, design and process operation. In a DCMD process, saline water with hazardous contaminants is passed on one side of a hydrophobic porous membrane while a colder water stream flows on the other side to

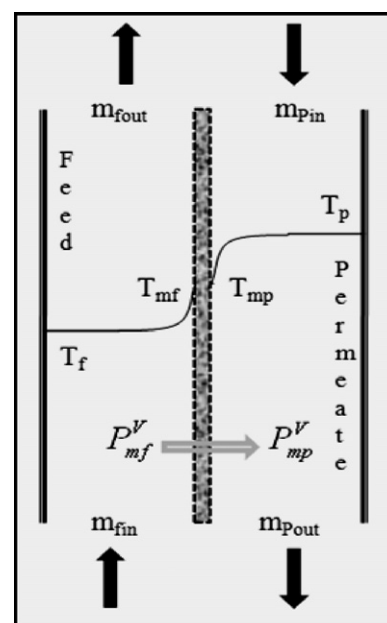


Fig. 1. Principle of direct contact membrane distillation.

directly condense the permeate water as shown in Fig. 1. The mass transfer, also called the flux, across the membrane occurs due to the evolved partial vapor pressure difference between the hot and cold sides of the membrane.

The advantage of membrane distillation process over distillation and other membrane based technologies can be explained as follows: low temperature operation (suitable to utilize local process waste heat and low grade renewable energy sources) and lower heat losses (energy efficient due to smaller surface area of the unit); smaller equipment size due to high surface to volume ratio; less vapor velocity and space compared to distillation units; thermally driven process operating at atmospheric pressures as opposed to reverse osmosis which operates at pressures much higher than atmospheric pressure and consume prime energy; 100% (theoretical) rejection of ions, macromolecules, colloids, cells, and other non-volatile constituents; pressure driven processes such as reverse osmosis, ultra filtration, micro filtration have not shown to achieve such high levels of rejection and permeate flux rates are comparable; less mechanical demands due to lower operating pressures; less fouling and more chemically resistant membranes (polytetrafluoroethylene (PTFE), polypropylene (PP), and polyvinylidenedifluoride (PVDF)) which just serve as a barrier to separate the hot and cold fluids [23,24].

2.1. Thermal analysis on the DCMD process

The mass transfer or permeate flux across the membrane can be expressed as [24,25]:

$$N = C(P_{mf}^v - P_{mp}^v) \quad (1)$$

where N is the vapor permeate flux ($\text{kg m}^{-2} \text{s}^{-1}$), P_{mf}^v and P_{mp}^v ($\text{kg m}^{-1} \text{s}^{-2}$) are vapor partial pressure of water vapor at the hot feed stream side (contaminated brackish water) and the permeate side (freshwater), respectively; and C (s m^{-1}) is the total mass transfer coefficient.

The heat transfer rate across the membrane Q_m is due to the heat of conduction through the membrane, $Q_{m,\text{cond}}$, and to the heat of evaporation of the volatile stream, $Q_{m,\text{M.T.}}$ [26].

$$Q_m = Q_{m,\text{cond.}} + Q_{m,\text{M.T.}} = h_m(T_{mf} - T_{mp}) + N\Delta H_v \quad (2)$$

where h_m (W/m² K) is the heat transfer coefficient of the membrane, T_{mf} , and T_{mp} (K) are the temperature of feed–membrane interface, membrane–permeate interface, respectively; and ΔH_v (J/kg) is the enthalpy of the vapor diffused.

Temperature polarization is the main reason for the heat transfer through the bulk of the solution [27]. The ratio of useful energy for mass transfer of vapors to the total energy invested in the process is the temperature polarization coefficient (TPC) which is commonly defined as [28,29]:

$$\text{TPC} = \frac{T_{mf} - T_{mp}}{T_f - T_p} \quad (3)$$

where T_f , and T_p (K) are temperatures of feed and permeate streams in the bulk side, respectively. T_{mf} , T_{mp} can be calculated using the following equations [27]:

$$T_{mf} = \frac{h_m(T_p + (h_f/h_p)T_f) + h_f T_f - N\Delta H_v}{h_m + h_f(1 + h_m/h_p)} \quad (3.1)$$

$$T_{mp} = \frac{h_m(T_f + (h_p/h_f)T_p) + h_p T_p + N\Delta H_v}{h_m + h_p(1 + h_m/h_f)} \quad (3.2)$$

where h_f , h_m and h_p (W/m² K) are the heat transfer coefficients on the feed, across the membrane and cold streams respectively. TPC is dependent on the feed and permeate velocities and module design and configuration. TPC value indicates if the process is heat transfer limited or mass transfer limited.

The heat transfer coefficient for the membrane can be obtained using the following equation [27,29]:

$$h_m = \frac{k_m}{\delta} = \frac{k_g \varepsilon + k_s(1 - \varepsilon)}{\delta} \quad (4)$$

where k_m (W/m K) is the thermal conductivity of the membrane, k_s and k_g (W/m K) are the thermal conductivities of the hydrophobic membrane material and the air trapped inside the membrane pores, respectively. ε is the porosity of the membrane material and δ is the thickness of the membrane.

The general correlation for the Nusselt number is given by [30]:

$$Nu = ARe^\alpha Pr^\beta \quad (5)$$

where Nu is Nusselt number $Nu = \frac{h_f d}{k_m}$, Re is Reynolds number $Re = \frac{d v \rho}{\mu}$, Pr is Prandtl number $Pr = \frac{C_p \mu}{k_m}$, and A , α , and β are parameters for the working regime and correlation used.

Using Nusselt number equation, h_f and h_p can be calculated for their respective flow conditions.

Thermal efficiency (%) of the DCMD process is given as [27]:

$$\eta = \frac{Q_{diff}}{Q_{diff} + Q_{cond}} = \frac{N\Delta H_v}{N\Delta H_v + h_m(T_{mf} - T_{mp})} \quad (6)$$

where η is the thermal efficiency, Q_{diff} is the vapor diffusive energy flux which is same as latent heat of the vapor passing through the membrane and Q_{cond} is the conductive energy flux. Eqs. (1)–(5) were used to calculate the thermal efficiency of the DCMD process.

2.2. Contaminant rejection efficiency

The contaminant rejection efficiency is calculated using the following equation:

$$R(\%) = \frac{C_f - C_p}{C_f} \times 100\% \quad (7)$$

where C_f is the contaminant concentration in the feed and C_p is the contaminant concentration in the permeate.

Table 1
Membrane characteristics.

Material	PP	PTFE
Type	Flat sheet	Flat sheet
Nominal pore size ((m)	0.22, 0.45	0.22
Thickness, ((m)	110	60
Porosity, (%)	0.70, 0.75	0.80
Thermal conductivity, k_m (W/m K)	0.054	0.073

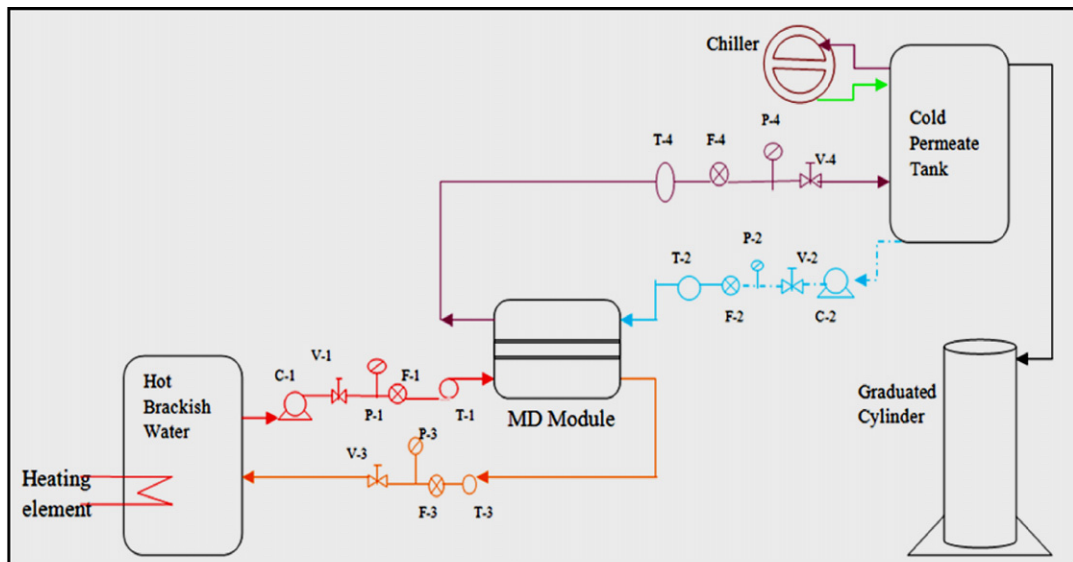
3. Materials and methods

Direct contact membrane distillation (DCMD) experiments were conducted to treat synthetic brackish water containing arsenic, uranium, and fluoride. Synthetic water was prepared using reagent grade chemicals and deionized water (Milli-Q system, resistivity of 18.2 M Ω cm). Two different microporous hydrophobic membranes supplied by General Electric were used, namely polytetrafluoroethylene (PTFE) and polypropylene (PP). A circular acrylic case with an inside diameter of 8.75 cm and a contact surface area of 60.0 cm² was assembled in the lab to hold the flat sheet membranes with same surface area. The characteristics of the two membranes are summarized in Table 1. Salt concentration in the water was varied between 1000 and 10,000 ppm. Arsenic and uranium concentrations were varied in the ranges from 10 to 400 ppb and fluoride concentration from 1 to 30 ppm (Uranium sample solution was prepared by adding uranyl nitrate (UO₂(NO₃)₂·6H₂O) solution to the deionized water to obtain required concentrations (10–400 ppb). Arsenic sample solution was prepared by adding standard Sodium salt heptahydrate (Na₂HAsO₄·7H₂O) solution, purchased from SCP science, to the deionized water sample to obtain required concentrations (10–400 ppb). Fluoride samples were prepared from Sodium Fluoride (NaF) solution, by diluting it to different required concentrations (1–30 ppm) using deionized water. NaF solution was purchased from Aqua Solutions). During the experiments feed brackish water at varying temperature, flow rate and concentration was circulated in counter current with cold distillate water contained in a 20 gallons tank and the permeate produced was collected in a graduated cylinder. Temperature and flow rate of the feed water ranged from 50 to 80 °C and 1–4 L/min, respectively. The feed water was heated with a tank heater (Model CH-OTS) with 6 kW capacity and built-in thermostat. The distilled water (permeate tank) was kept cold with help of a chiller (Isotemp 3006S). The experimental unit included temperature, flow rate and pressure measuring devices for inlet and outlet streams (Fig. 2). The TDS concentration of permeate produced was measured with a conductivity meter (Orion 120). Arsenic and uranium concentrations were analyzed with inductively-coupled plasma mass spectroscopy (ICP-MS) (Elan DRC-e, Perkin Elmer), and fluoride concentration was measured with a combined pH and ion-meter (Accumet Excel XL25, Fischer Scientific). Data collected were used to calculate the permeate flux, thermal efficiency and contaminants removal efficiency. Long term experiments were conducted for 12 hr and permeate was collected to analyze the effect of membrane aging in permeate flux production and quality. Analysis of the structural changes of the membranes was conducted with a scanning electron microscope (Hitachi, S-3400N).

4. Results

4.1. Effect of feed temperature

The effect of temperature gradient across the membrane on the permeate flux and its relation to the vapor pressure are shown in Fig. 3a and b. The vapor pressure was calculated using Antoine's equation [27,28]. It is evident that the mass transfer across the



(C =centrifugal pump, F=flow meter, P= pressure gauge, T= temperature recorder and V=valve; 1.hot water in, 2.cold water in, 3.hot water out, 4.cold water out)

Fig. 2. Schematic of the experimental set up for DCMD process.

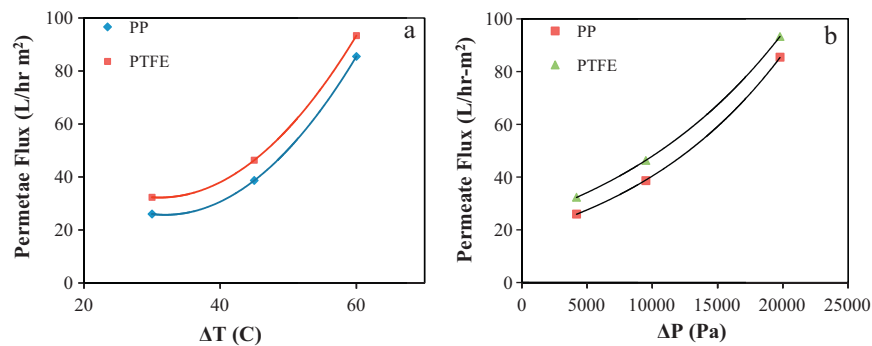


Fig. 3. Effect of temperature gradient (a) and vapor pressure difference (b) on the permeate flux (Process parameters: pore size = 0.22 (m), $T_p = 293$ K, feed and permeate volumetric flows, $(r_f = r_p = 4$ L/min; feed and permeate side pressures, $P_f = P_p = 5.5$ psi; salt concentration: 6000 ppm, $U, A_s = 400$ ppb, $F = 30$ ppm).

membrane is due to the evolved vapor pressure difference between the hot and cold sides of the membrane (Fig. 3b). Increased feed temperature results in higher flux rates due to increased vapor pressure difference. Thermal efficiency increased with increase in feed temperature. For the feed temperature about 50 °C, thermal efficiency measured was 60% and at 80 °C, it was 70% for PP membrane while the values for PTFE membrane were 51% and 61.5% for different temperatures respectively (Fig. 4a and b). This observation

is similar to other previous studies [26,31,32]. At high temperatures the amount of heat transfer by conduction has less effect than the heat transferred by the diffusing species on the thermal efficiency. The increase of the thermal efficiency with high feed temperature is due to the exponential rise in the mass flux with temperature [27,32]. The thermal efficiency is defined as the ratio of the heat transferred due to the mass transfer of the permeate vapor to the overall heat transferred in the DCMD process (Eq. (6)).

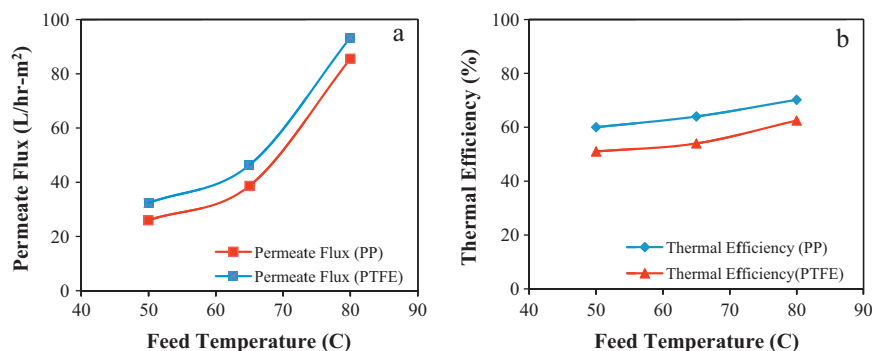


Fig. 4. Effect of the feed temperature on (a) permeate flux and (b) thermal efficiency (Process parameters: pore size = 0.22 (m), $T_p = 293$ K, $(r_f = r_p = 4$ L/min, $P_f = P_p = 5.5$ psi, Salt concentration: 6000 ppm, $U, A_s = 400$ ppb, $F = 30$ ppm).

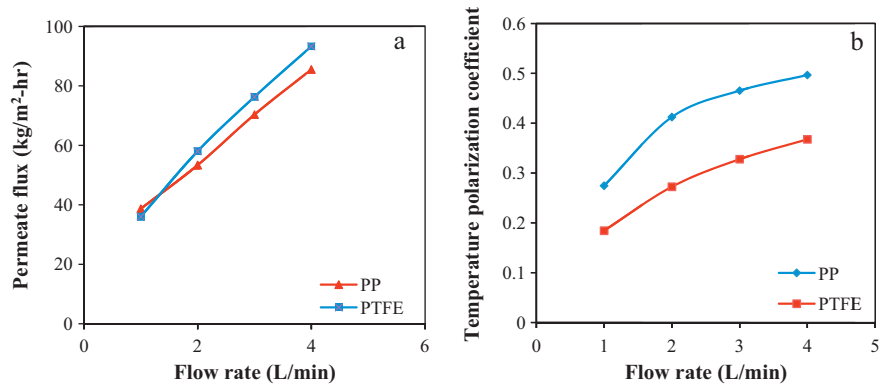


Fig. 5. Effect of the flow rate on (a) permeate flux and (b) temperature polarization coefficient ($(r=(p, T_f=353\text{ K and } T_p=293\text{ K, pore size}=0.22\text{ }\mu\text{m, salt concentration}=6000\text{ ppm, U, As}=400\text{ ppb, F}=30\text{ ppm})$).

4.2. Effect of feed flow rate

The effect of feed flow rate on the permeate flux and temperature polarization coefficient is shown in Fig. 5a and b. The permeate flux increased with increase in the feed velocity as well as the temperature polarization coefficient. However, the permeate flux increased with increase in temperature gradient and temperature polarization coefficient decreased from 0.60 to 0.49 for PP and 0.42 to 0.36 for PTFE membranes. The exponential increase of MD flux with temperature for commercial PTFE membranes is due to the exponential increase of vapor pressure with temperature as per the Antoine equation. The permeate flux for PP membrane is lower than PTFE membrane as mass transfer resistance of the PP membrane was higher due to higher membrane thickness. Thus the effect of the membrane thickness at higher temperature overshadows the effect of the temperature on water flux. The other reasons for the higher flux of PTFE membranes than PP are higher porosity of PTFE membranes and rougher surfaces of PTFE membranes that help mixing at the membrane interfaces [21]. On the other

hand, thermal conductivity of the PP membrane is lower compared to PTFE membrane which results in higher thermal efficiency. The TPC decreased with increase in the temperature gradient due to higher bulk temperature gradient (denominator of Eq. (3)) of feed and permeate side flows. Other reasons could be inefficient mixing (lack of channel design) and small feed and permeate velocities [29,33]. The TPC values for PP membrane were higher than PTFE due to lower heat losses through the membrane (thermal conductivity of the PP membrane is lower). It suggests that the design for DCMD module should include proper channeling to improve the temperature polarization coefficient. If the TPC is less than 0.2, the process is heat transfer limited and would indicate a poor module design. If the TPC is greater than 0.6, the process is mass transfer limited due to a low membrane permeability. Usually, the TPC depends on the membrane characteristics, feed velocity, fluid dynamics and feed concentration. Flux increases with increasing fluid flow over the membrane surface [29,33]. The mass flux increases with increase in the feed temperature and flow velocities on both sides of the membrane. Permeate flux was $36\text{ kg/m}^2\text{ hr}$ at a feed velocity of 0.105 m/s

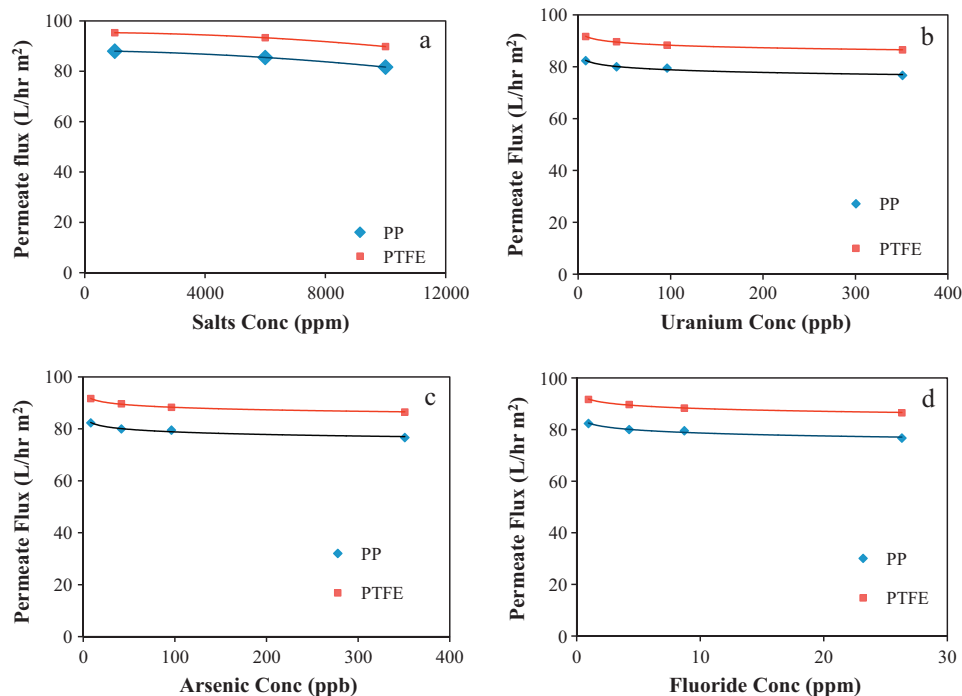


Fig. 6. Effect of (a) salt; (b) uranium; (c) arsenic; and (d) fluoride concentrations on the permeate flux (pore size = $0.22\text{ }\mu\text{m}$, $T_f=353\text{ K}$ and $T_p=293\text{ K}$, $(r=(p=4\text{ L/min, } P_f=P_p=5.5\text{ psi})$).

Table 2
Contaminant removal rates (0.22 (m pore size).

Membrane type	Feed conc. (ppm)	Rejection rate salts	Feed conc. (ppb)	Rejection rate As	Rejection rate U	Feed conc. (ppm)	Rejection rate F
PP	10,000	99.93	10	99.41	98.77	1	95.1
PTFE		99.94		97.78	96.13		90.1
PP		99.94	40	99.64	96.71	5	98.29
PTFE		99.96		99.82	99.83		98.88
PP		99.96	100	98.99	99.73	10	99.24
PTFE		99.9		99.42	99.71		98.94
PP		99.93	400	99.83	99.93	30	99.56
PTFE		99.9		99.85	99.49		99.38

and the flux was 93 kg/m² hr at a feed velocity of 0.42 m/s. The flow velocities were equivalent to a Reynolds number of 240–960 meaning that a laminar flow was kept during the experiments. The difference in permeate flux is due to the reduced temperature polarization and improved heat transfer coefficients due to effective mixing in the bulk fluid at higher feed velocities [34].

4.3. Effect of salt concentration and other contaminants (arsenic, uranium and fluoride)

The effect of salt concentration has been tested in the range of 1000–10,000 ppm. Fig. 6a shows that the higher salt concentrations result in lower permeate flux rates. This result is in agreement with results reported in the literature [24,27]. The permeate flux rate decreased by 5% for the PTFE membrane when the salt concentration was increased from 1000 ppm to 10,000 ppm. Water vapor pressure reduction due to concentration polarization effect is the reason for the decrease in permeate flux rate at higher salt concentrations [28]. Over 99% of salts were rejected through the DCMD process which resulted in high quality permeate suitable for industrial and process applications. Similarly, the arsenic/fluoride/uranium concentrations were increased by 10 times the acceptable drinking water standards and the effect of which was found to be minimal on the permeate flux rates (Fig. 6b–d). The decrease in permeate flux rate is more evident when the contaminants concentration is high, probably because the polarization effect increases. For the highest arsenic, fluoride, and uranium feed concentrations, the resulting concentrations in the permeate were 0.17 ppb, 0.56 ppm, and 0.2 ppb, respectively. The removal efficiency of these contaminants for most of the tests was in the range of 99–99.9% by the DCMD process which is higher than other conventional technologies (Table 2). Scaling and fouling of the membrane might occur when the concentration of the salt is high as reported by He et al. [35–37]. Fouling will cause damage of the membrane and will not prevent the passage of small quantities of arsenic, fluoride, and uranium through the membrane. The results for arsenic removal were in agreement with those obtained in previous studies [20–22]. The effect of salt concentration in the feed water was significant on the permeate flux rate compared to the concentration of the contaminants. This means the DCMD process can be applied to recover impaired waters containing variety of other hazardous compounds. In recent studies by Macedonio and Drioli, membrane distillation process was combined with reverse osmosis process to recover freshwater from arsenic and boron contaminated waters [38,39]. This hybrid process removed arsenic and boron with much lower chemical requirements. From Table 2, it can be concluded that both the PP and PTFE membranes have performed at the same level and had similar contaminant rejection rates except the PTFE membrane produced higher permeate flux compared to PP membrane. In all the tests performed, the PTFE membrane performed at higher desalination efficiency which is mainly a function of vapor flux (Eq. (5)).

4.4. Pore size effect and long run performance

Membrane properties such as porosity, pore size, and membrane thickness play a significant role in dictating the resistance to mass transfer through microporous membranes in membrane distillation. Membrane conditions such as compaction and wetting are important for efficient operation of the DCMD process as well [28]. The pore size effect on the permeate flux is shown in Fig. 7. Permeate flux rate increased with larger pore size, however, at higher feed flow rates the membrane may get wetted and the pores may get plugged with the liquid permeate causing resistance to mass transfer at extended runs. Larger pore size may allow the lower molecular weight contaminants such as fluorides to pass through the membrane thus reducing the quality of the freshwater recovered. Therefore, it is important to develop the membrane materials with high permeability properties. MD Membranes with higher permeability rates may compete with reverse osmosis by using low grade waste or renewable heat to meet the mechanical energy requirements under 2 kWh/m³ [40].

Fouling is one of the main obstacles for membrane distillation irrespective of the configuration. Several types of fouling can occur in the membrane systems, inorganic, organic and biological fouling [41]. 12 hr tests were conducted to evaluate the effect of inorganic fouling on the membranes. The permeate flux remained unchanged more or less during the 12 hr test operation. The SEM images of PTFE membrane before and after 12 hr operation did not show any characteristics of fouling. It has been reported in a recent study that the membrane morphology changed slightly after 250 hr of operation using arsenic contaminated feed water [22]. In another study, almost 100% of arsenic rejection was observed even after 120 hr of operation without wetting the membrane pores [21].

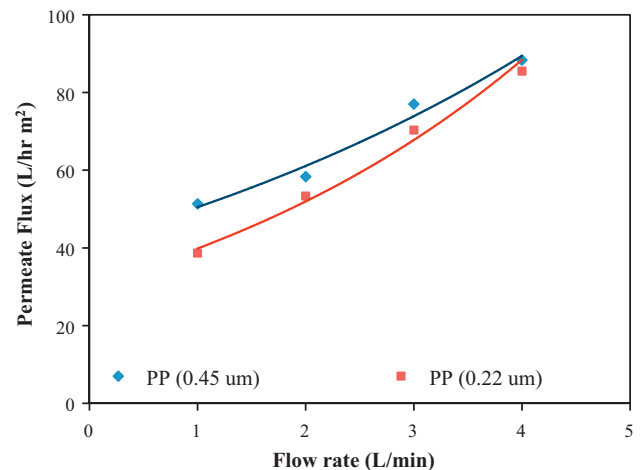


Fig. 7. Effect of pore size on the permeate flux ($T_f=353\text{ K}$ and $T_p=293\text{ K}$, ($r_p=4\text{ L/min}$. Salt concentration: 6000 ppm, U, As=400 ppb, F=30 ppm).

These results indicate that MD membranes are resistant to hazardous and radioactive contaminants present in the feed water.

5. Conclusions

The feasibility of applying membrane distillation process for recovering potable water from arsenic, uranium and fluoride contaminated brackish waters has been demonstrated through experimental studies. The membrane distillation process produced a high quality permeate with dissolved solids concentrations less than 20 ppm (>99% rejection of salts) along with arsenic, fluoride and uranium contaminant reductions in the range of 96.5–99.9%. It was observed that the DCMD process was tolerant of the chemicals present in the feed water and produced constant permeate rates irrespective of the contaminant concentrations, which is uncommon in other separation processes. The high quality permeate recovered from the DCMD process can be diluted with the raw water to produce a resultant drinking water with permissible contaminant concentration levels to improve the economics of the process. Among PP and PTFE membranes employed in this study, the PTFE membrane produced a higher permeate flux rate and desalination efficiency in all cases; however, the contaminant removal rates were comparable between PP and PTFE membranes. The simplicity of the design and operation of the DCMD process enables the technology to be coupled with various types of waste/renewable heat sources or hybrid configurations to recover freshwater from highly contaminated saline groundwaters.

References

- [1] Meenakshi, R.C. Maheshwari, Fluoride in drinking water and its removal, *J. Hazard. Mater.* 137 (2006) 456–463.
- [2] D. Chakraborti, S.C. Mukherjee, S. Pati, M.K. Sengupta, M.M. Rahman, U.K. Chowdhury, D. Lodh, C.R. Chanda, A.K. Chakraborti, G.K. Basu, Arsenic groundwater contamination in Middle Ganga Plain, Bihar, India. A future danger? *Environ. Health Persp.* 111 (2003) 1194–1201.
- [3] Groundwater Contamination, National Academy of Sciences, National academy press, Washington, DC, 1984.
- [4] K. Ong, New Mexico: Groundwater Quality, National Water Summary, U.S. Geological survey water-supply paper 2325, (1986) 377–384.
- [5] N.R. Frederick, Arsenic in ground-water under oxidizing conditions, *Environ. Geochem. Health* 11 (1989) 171–185.
- [6] R. Nickson, J. McArthur, W. Burgess, K.A. Ahmed, P. Ravenscroft, M. Rahman, Arsenic poisoning of Bangladesh groundwater, *Nature* 395 (1998) 338.
- [7] A.H. Smith, P.A. Lopipero, M.N. Bates, C.M. Steinmaus, Arsenic epidemiology and drinking water standards, *Science* 296 (2002) 2145–2146.
- [8] D.K. Nordstrom, Worldwide occurrences of arsenic in groundwater, *Science* 296 (2002) 2143–2145.
- [9] Arsenic and Uranium Removal from Drinking Water by Adsorptive Media U.S. EPA Demonstration Project at Upper Bodfish in Lake Isabella, CA, Interim Evaluation Report, EPA/600/R-08/026, April 2008.
- [10] C.T. Hess, J. Michael, T.R. Horton, H.M. Prichard, W.A. Coniglio, The occurrence of radioactivity in public water supplies in the United States, *Health Phys.* 48 (1985) 553–586.
- [11] USEPA. National Primary Drinking Water Regulations: Radionuclides Final Rule. Federal Register, 40 CFR Parts 9, 141, and 142 (2000).
- [12] S.L. Stover, Removal of uranium from aqueous wastes using electrically charged carbon nanofibers, Master's thesis, West Virginia University, Morgantown, West Virginia, 2000.
- [13] A.C. Hakonson-Hayes, P.R. Fresquez, F.W. Whicker, Assessing potential risks from exposure to natural uranium in well water, *J. Environ. Radioactiv.* 59 (2002) 29–40.
- [14] S. Hostetler, J. Wischusen, G. Jacobson, Groundwater Quality in the Papunya-Kintore Region, Australian Geological Survey Organisation, Northern Territory Canberra, 1998.
- [15] M. Pinon-Miramontes, R.G. Bautista-Margulis, A. Perez-Hernandez, Removal of arsenic and fluoride from drinking water with cake alum and a polymeric anionic flocculent, *Fluoride* 36 (2003) 122–128.
- [16] N. Sanjeev Kumar, S. Goel, Factors influencing arsenic and nitrate removal from drinking water in a continuous flow electrocoagulation (EC) process, *J. Hazard. Mater.* 173 (2010) 528–533.
- [17] S.R. Wickramasinghe, B. Han, J. Zimbron, Z. Shen, M.N. Karim, Arsenic removal by coagulation and filtration: comparison of groundwaters from the United States and Bangladesh, *Desalination* 169 (2004) 231–244.
- [18] E.M. Aieta, Radionuclides in drinking water: an overview, *J. Am. Water. Works Assoc.* 79 (1987) 144–152.
- [19] J.D. Lowry, S.B. Lowry, Radionuclides in drinking water, *J. Am. Water. Works Assoc.* 80 (1988) 51–64.
- [20] A.K. Manna, M. Sen, A.R. Martin, P. Pal, Removal of arsenic from contaminated groundwater by solar-driven membrane distillation, *Environ. Pollut.* 158 (3) (2010) 805–811.
- [21] P. Pal, A.K. Manna, Removal of arsenic from contaminated groundwater by solar-driven membrane distillation using three different commercial membranes, *Water Res.* 44 (2010) 5750–5760.
- [22] D. Qu, J. Wang, D. Hou, Z. Luan, B. Fan, C. Zhao, Experimental study of arsenic removal by direct contact membrane distillation, *J. Hazard. Mater.* 163 (2009) 874–879.
- [23] M.I. Ashiq, Membrane distillation process for pure water and removal of arsenic, Masters thesis, Chalmers University of Technology, Sweden, 2005.
- [24] S. Yarlagadda, L.M. Camacho, V.G. Gude, Z. Wei, S. Deng, Membrane distillation for desalination and other separations, *Recent Pat. Chem. Eng.* 2 (2009) 128–158.
- [25] K.W. Lawson, D.R. Lloyd, Membrane distillation: review, *J. Membr. Sci.* 124 (1997) 1–25.
- [26] A. Criscuoli, M.C. Carnavale, E. Drioli, Evaluation of energy requirements in membrane distillation, *Chem. Eng. Process.* 47 (2008) 1098–1105.
- [27] M. Qtaishat, T. Matsuura, B. Kruczek, M. Khayet, Heat and mass transfer analysis in direct contact membrane distillation, *Desalination* 219 (2008) 272–292.
- [28] T.Y. Cath, V.D. Adams, A.E. Childress, Experimental study of desalination using direct contact membrane distillation: a new approach to flux enhancement, *J. Membr. Sci.* 228 (2004) 5–16.
- [29] L. Martinez-Diez, M.I. Vazquez-Gonzalez, F.J. Florido-Diaz, Temperature polarization coefficients in membrane distillation, *Sep. Sci. Technol.* 33 (1998) 787–799.
- [30] M.A. Izquierdo-Gil, C. Fernandez-Pineda, M.G. Lorenz, Flow rate influence in direct contact membrane distillation experiments: different empirical correlations for Nusselt number, *J. Membr. Sci.* 321 (2008) 356–363.
- [31] A.S. Jonsson, R. Wimmerstedt, A.C. Harrysson, Membrane distillation—a theoretical study of evaporation through microporous membranes, *Desalination* 56 (1985) 237–249.
- [32] F.W. Banat, J. Simandl, Desalination by membrane distillation: a parametric study, *Sep. Sci. Technol.* 33 (1998) 201–226.
- [33] A. Burgoyne, M.M. Vahdati, Direct contact membrane distillation, *Sep. Sci. Technol.* 35 (2000) 1257–1284.
- [34] S. Srisurichan, R. Jiratananon, A.G. Fane, Mass transfer mechanisms and transport resistances in direct contact membrane distillation process, *J. Membr. Sci.* 277 (2006) 186–194.
- [35] F. He, J. Gilron, H. Lee, L. Song, K.K. Sirkar, Potential for scaling by sparingly soluble salts in crossflow DCMD, *J. Membr. Sci.* 311 (2008) 68–80.
- [36] F. He, K.K. Sirkar, J. Gilron, Studies on scaling of membranes in desalination by direct contact membrane distillation: CaCO₃ and mixed CaCO₃/CaSO₄ systems, *Chem. Eng. Sci.* 64 (2009) 1844–1859.
- [37] F. He, K.K. Sirkar, J. Gilron, Effects of antiscalants to mitigate membrane scaling by direct contact membrane distillation, *J. Membr. Sci.* 345 (2009) 53–58.
- [38] F. Macedonio, E. Drioli, Pressure-driven membrane operations and membrane distillation technology integration for water purification, *Desalination* 223 (2008) 396–409.
- [39] F. Macedonio, E. Drioli, Membrane engineering progresses in desalination and water reuse, *Membr. Water Treat.* 1 (1) (2010) 75–81.
- [40] C. Cabassud, D. Wirth, Membrane distillation for water desalination: how to choose an appropriate membrane, *Desalination* 157 (2003) 307–314.
- [41] M. Gryta, Fouling in direct contact membrane distillation process, *J. Membr. Sci.* 325 (2008) 383–394.