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## Removal of arsenic from contaminated groundwater by solar-driven membrane distillation

Ajay K. Manna<sup>a</sup>, Mou Sen<sup>a</sup>, Andrew R. Martin<sup>b</sup>, Parimal Pal<sup>a,\*</sup><sup>a</sup> Environment and Membrane Technology Laboratory, Department of Chemical Engineering, National Institute of Technology, Durgapur 713209 India<sup>b</sup> Department of Energy Technology, The Royal Institute of Technology (KTH), Stockholm, Sweden*Solar-driven membrane distillation has the potential of removing arsenic from contaminated groundwater.*

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## ABSTRACT

Experimental investigations were carried out on removal of arsenic from contaminated groundwater by employing a new flat-sheet cross flow membrane module fitted with a hydrophobic polyvinylidene-fluoride (PVDF) microfiltration membrane. The new design of the solar-driven membrane module in direct contact membrane distillation (DCMD) configuration successfully produced almost 100 per cent arsenic-free water from contaminated groundwater in a largely fouling-free operation while permitting high fluxes under reduced temperature polarization. For a feed flow rate of 0.120 m<sup>3</sup>/h, the 0.13 μm PVDF membrane yielded a high flux of 74 kg/(m<sup>2</sup> h) at a feed water temperature of 40 °C and, 95 kg/m<sup>2</sup> h at a feed water temperature of 60 °C. The encouraging results show that the design could be effectively exploited in the vast arsenic-affected rural areas of South-East Asian countries blessed with abundant sunlight particularly during the critical dry season.

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### 1. Introduction

Groundwater contamination by arsenic leachate has now affected millions of people from South-East America to Argentina, Taiwan, China, Nepal, Bangladesh and India. Most of the affected parts belong to the developing countries in South-East Asia where the problem assumes an alarming proportion particularly during the dry seasons (January–June) when groundwater levels fall sharply. Arsenic contamination and its mitigation is a priority area in drinking water quality within the World Health Organization (WHO) and other national and international agencies, and a multitude of studies (Schreiber et al., 2000; Panthi et al., 2006; Pokhrel et al., 2009; Chen et al., 2009; Harvey et al., 2006; Chowdhury et al., 2000; Chakraborti et al., 2003; Bhattacharjee et al., 2005; Acharya, 2002) have been commissioned many of which focus on West Bengal (India) and Bangladesh where the largest affected population lives on the Bengal Delta Basin. While the problem is relatively well known, challenges remain in the widespread implementation of low-cost and effective arsenic removal strategies in such regions. Through extensive studies (Pal et al., 2007a,b; Wickramasinghe et al., 2004; Pagana et al., 2008;

Hsieh et al., 2008; Xia et al., 2007; Hering and Elimelech, 1996; Greenleaf et al., 2006; Brandhuber and Amy, 2001; Fagarassy et al., 2009; Nguyen et al., 2009) carried out over the last few decades on removal of arsenic from drinking water, adsorption, chemical coagulation–precipitation, ion-exchange and membrane separation have been established as the broad technology options of purification. Though arsenic removal efficiencies of these processes are largely established as shown in the review paper (Shih, 2005) and presented in Table 1. Wide variations are observed in designs, separation results, applicability and viability of the reported membrane-based techniques which are relatively new. Socio-politico-economic conditions (Roy et al., 2008) also vary from country to country and treatment options are not equally available in all the affected countries. Thus permissible maximum contaminant level (MCL) of arsenic in drinking water varies across the affected countries as compiled by Choong et al. (2007) in Table 2. Moreover, in majority of membrane-based separation studies, simulated water instead of actually contaminated groundwater has been used in the backdrop of high sensitivities of membranes to feed water characteristics. Pal et al. (2007a) have shown that for large scale treatment, physico-chemical separation technique is possibly the best for the developing South-East Asian countries. For small-scale treatment, solar-driven membrane distillation (SDMD) could be an ideal technology option as almost 100% arsenic can be separated out from groundwater using the low grade solar energy

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**Table 1**  
Relative removal efficiencies for As(V) by the major existing techniques.

Treatment process	Maximum removal, %
Alum precipitation process	90
Iron precipitation	95
Lime softening process (pH > 10.5)	90
Combined with iron (and manganese) removal	>90
Ion exchange (sulfate 50 ppm)	95
Activated alumina	95
Reverse osmosis, nanofiltration, electrodialysis	>95
Membrane distillation	>99.9%

which is abundant in these affected countries, particularly during the crisis months of the dry season. Fouling which is the major disadvantage of a membrane-based separation process is almost absent here as there is little chance of clogging of the pores of the hydrophobic microfiltration membrane used in MD. Moreover, necessity of high transmembrane pressure of reverse osmosis is redundant in this SDMD. Using microporous membranes as a support for vapor–liquid interfaces at the entrance of the pores, SDMD can operate on the principle on vapor liquid equilibrium as a basis for molecule separation. In MD process, a hot aqueous feed solution is brought in contact with one side of a hydrophobic, microporous membrane. After the evaporation of volatile molecules, to be separated from the feed, at the hot feed side, transport of vapor through dry pores of hydrophobic membranes occurs due to a vapor pressure difference across the membrane, which is the driving force. As membrane material is water repellent, liquid water cannot enter the pores unless a hydrostatic pressure exceeding the liquid entry pressure of water is applied. In the absence of such a pressure differential between the two sides of the membrane, a liquid–vapor interface is formed on either side of the membrane pores and a vapor pressure difference can result between these two interfaces from a temperature difference that has to be maintained. Evaporation can take place on the hot interface producing vapor to be subsequently transported to the other relatively cold side through the pores. Transport of such vapor can take place following Knudsen model, Poiseuille model, transition-Knudsen model or pore diffusion model depending on the magnitude of Knudsen number ( $K_n$ ) which is defined as the ratio ( $\lambda/d$ ) of the mean free path ( $\lambda$ ) of the transported molecules and the diameter of the pores ( $d$ ). When  $K_n > 1$ , Knudsen model dominates and for  $K_n < 0.01$ , molecular diffusion model can better explain the transport process. Details of such models can be found in Schofield et al. (1987), Kimura et al. (1987) and Phattaranawik et al. (1998).

Membranes with pore sizes ranging from 0.01  $\mu\text{m}$  to 1  $\mu\text{m}$  should be generally used. MD can be compared with pressure-driven microfiltration, which is characterized by a membrane pore size between 0.05  $\mu\text{m}$  and 2  $\mu\text{m}$  and operating pressure above 2 bars. The main requirements for MD process are that the membrane should not be wetted and only vapor and noncondensable gases should be present within its pores. Such hydrophobic, microporous membranes made of polytetrafluoroethylene (PTFE), polypropylene (PP), polyethylene (PE), and polyvinylidene fluoride (PVDF) are now commercially available.

Though solar-driven membrane distillation (SDMD) could be an ideal technology solution to groundwater arsenic contamination problem relatively little attention has been drawn to this technique and membrane distillation despite being known since the late 1960s mainly remained confined within desalination for the production of ultra pure water from saline water. However, in some recent studies (Macedonio and Drioli, 2008; Qu et al., 2009) experiments have been conducted utilizing laboratory-scale direct contact membrane distillation (DCMD) modules with tubular geometries for arsenic separation. These studies report high

**Table 2**  
Maximum permissible contaminant level (MCL) of As set by different countries.

Countries/others	Permissible value ( $\mu\text{g/L}$ )
WHO/USPEA/European union	10
Germany	10
Australia	7
France	15
India, Bangladesh, Vietnam, Mexico	50
Malaysia	10–50

removal efficiencies, especially when compared to pressure-driven membrane processes like reverse osmosis (RO) or nanofiltration (NF). Qu et al. (2009) found that the concentration of arsenic in product water could be brought down to 10  $\mu\text{g/L}$  with feed water arsenic concentrations as high as 40 mg/L and 2000 mg/L for As(III) and As(V), respectively. Macedonio and Drioli (2008) reported similar results, although the feed water arsenic concentration was much lower in their studies. Islam (2005) studied arsenic separation by air gap membrane distillation (AGMD) using a small-scale commercial prototype MD module and reports successful treatment of arsenic-contaminated water. However, in the reported investigations, flux still remains low which stands in the way of viability of MD process in arsenic separation and studies on SDMD in arsenic separation are highly inadequate to build up scale up confidence.

The objective of the present work was to study a solar-driven DCMD module using flat plate geometry (Fig. 1) in a cross flow mode for arsenic removal from contaminated groundwater.

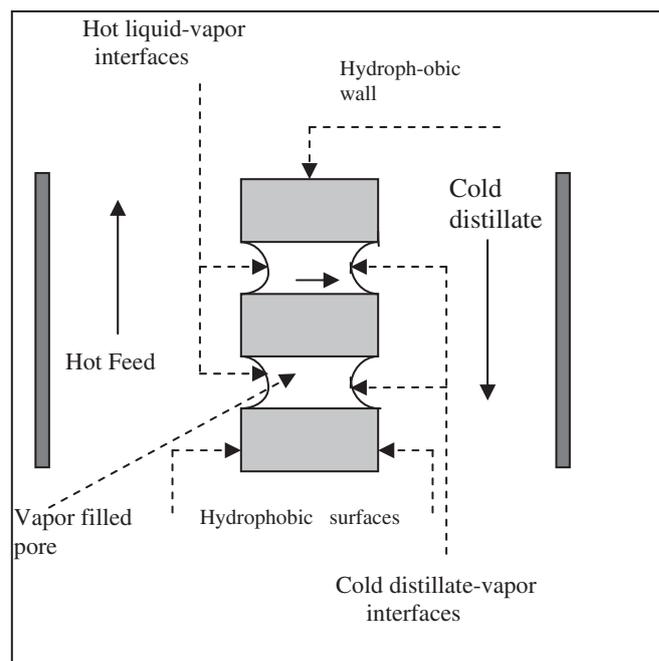
## 2. Materials and methods

### 2.1. Experimental set up

Fig. 2 shows the solar-driven membrane distillation set up used for carrying out the DCMD tests. The solar-driven membrane distillation set up consisted of four major components – a direct contact membrane distillation (MD) module, a solar energy collector and two thermostatic baths. The system works in two loops, namely the solar loop and the arsenic removal loop.

#### 2.1.1. Solar heating loop

An evacuated glass tube type solar energy collector (Bhaskar Solar, India) was used to heat up the feed (arsenic-contaminated water). The schematic diagram of

**Fig. 1.** DCMD configuration.

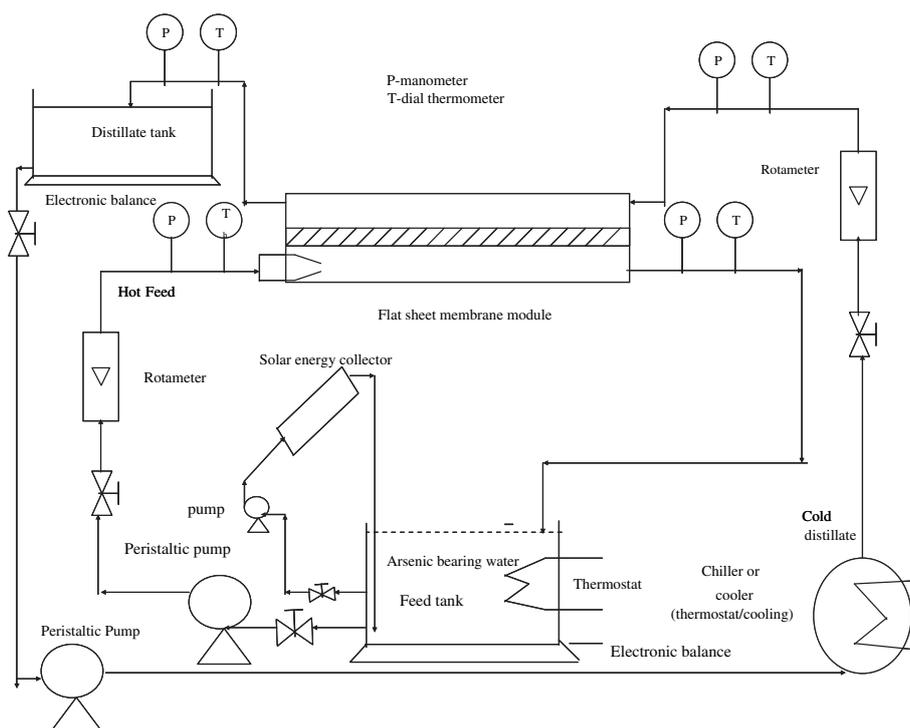


Fig. 2. Process flow diagram for DCMD (Direct Contact Membrane Distillation) experimental set up.

the solar loop is shown in Fig. 3. It had two major components, a solar collector and a 100 L solar collector storage tank. In this type, the solar collector is made of double layer borosilicate glass tubes (47 mm outer diameter, 37 mm inner diameter, 1.5 m length) evacuated (air is removed completely creating vacuum) for providing very good insulation. The outer wall of the inner tube is coated with selective absorbing material (black chrome) that absorbs solar radiation and transfers the heat to the water, which flows through the inner tube. Solar-heated water circulates between storage tank and heating tubes continuously by natural convection and gravity (thermo-siphon process) till the sunset. The brighter and stronger the radiation falling on the collector, the faster is the circulation. Circulation of hot water between the feed tank of the MD module and the solar heating system was maintained by a centrifugal pump. The storage tank permits extended operation of the MD module even after sunset. Feed flow rate was measured using a rotameter in the loop while a manometer registered the pressure in the collector storage tank ( $P_c$ ). Temperatures of feed inlet, outlet and collector storage tank were registered on three attached dial thermometers.

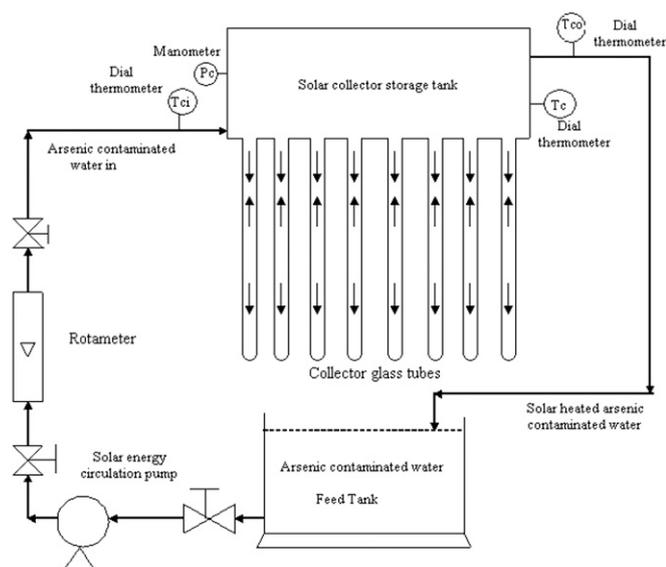


Fig. 3. Schematic flow diagram of solar loop.

### 2.1.2. Arsenic removal loop

The arsenic removal loop consisted of two peristaltic pumps (Enertech Electronics Private Limited, India) for the circulation of the cold (distillate) and hot (feed) streams, two rotameters for measurement of these flow rates and a flat-sheet direct contact membrane distillation module operated in cross flow mode. Flat-sheet hydrophobic PVDF membranes (Sepromembranes) of 0.13  $\mu\text{m}$  pore size were used in the membrane module. Hot feed water was pumped to the lower side of the membrane while cold stream flowed counter-currently over the upper surface of the membrane in the module. The properties of the used PVDF membrane are shown in Table 3. Inlet and outlet pressures of the two streams were monitored through manometers. Dial thermometers registered the module inlet and outlet temperatures of both the streams. Two thermostatic baths (Polyscience, NILES, USA) were used for maintaining constant desired temperatures of the hot and cold streams. The effective membrane surface area in lab-made flat module design was 0.0162  $\text{m}^2$ . The flat membrane module was horizontally oriented.

### 2.1.3. Experimental procedure

Experiments were carried out using mainly arsenic-contaminated groundwater collected from some affected areas (West Bengal, India) and the characteristics of this water are given in Table 4. PVDF membrane sheets cut from a roll were sandwiched between two halves of a rectangular polycarbonate box. A finely perforated stainless steel plate supported the membrane in the module.

The feed temperature in the feed tank was varied between 40  $^{\circ}\text{C}$  and 61  $^{\circ}\text{C}$  with the aid of solar heating loop and the circulation pump at the ground level. Membrane module was run after heating feed water to 40–42  $^{\circ}\text{C}$ . Feed side and distillate side temperatures were controlled by controlling the ground level circulation pump and using thermostatic baths attached to the module. During experiments, the feed was at atmospheric pressure and its flow rate was varied between 50 and 120 kg/h. In DCMD test, the distillate flow rate was maintained at 150 kg/h and the distillate water temperature was maintained at 20–22  $^{\circ}\text{C}$ . Flow rates of the feed and distillate were controlled by controlling the circulating peristaltic pumps. Leakages were checked prior to each run. Inlet feed pressure was controlled to avoid membrane wetting. Data were collected after the system had reached steady state.

Table 3  
Some characteristics of the used PVDF microfiltration membrane.

Material	PVDF (Sepro)
Type	Flat sheet
Nominal pore size ( $\mu\text{m}$ )	0.13
Thickness ( $\mu\text{m}$ )	150
Porosity (%)	70–75

**Table 4**  
Characteristics of arsenic-contaminated groundwater used in the experiment (water from Chakdah West Bengal).

Contaminants	Concentration (mg/L)
Arsenic	0.3–0.5
Iron	4.2–5.0
Ca	102.5–110.3
Na	26–32
TSS	250–312
Magnesium	21–23
Manganese	1–2.2
pH	7.1–7.3

Temperatures, pressures, and flow rates were continuously monitored and controlled. Quantity and temperature of distillate water in the distillate tank were recorded every 15 min. Estimation of arsenic in the collected distillate samples was done with the help of an Atomic Absorption Spectrophotometer (Model AAS-100, Perkin Elmer). The difference in weight of the distillate in the distillate tank over a certain time period was used to calculate the distillate flux through the known membrane surface under the given experimental conditions. Distillate flux was calculate using the relation distillate flux (kg/m<sup>2</sup> h)

$$= \frac{\text{Difference in weight of distillate in the distillate tank (kg)}}{\text{Membrane area (m}^2\text{) \cdot Time (h)}} \quad (1)$$

2.1.4. Analytics

Samples of distillate collected at definite interval were analyzed for measurement of arsenic concentration in an atomic absorption spectrophotometer (AAS-100, Perkin Elmer) following flame-fias technique the details of which are given elsewhere by Pal et al. (2007b). Measurement of concentrations of iron, calcium, magnesium, manganese and sodium in groundwater was also done by the same atomic absorption spectrophotometer following standard procedure (as given in the Analytical Methods, part number 0303-0152, Perkin Elmer, USA, 1996).

3. Results and discussion

3.1. Performance of the solar energy collector and the membrane module

The most important performance parameters of SPMD process include solar collector efficiency ( $\eta_c$ ), evaporation efficiency, gained output ratio (GOR) and thermal recovery ratio (TRR) and the observed values of these performance parameters have been presented in Table 5 for a specific set of operating conditions.

3.1.1. Energy efficiency of the solar collector ( $\eta_c$ ) system

The solar collector is the most important component in the SPMD unit. The energy efficiency of solar collector correlates the thermal energy gained to the solar radiation on the collector, and it is defined as the ratio of useful energy obtained in collector to solar radiation in coming to the collector.  $\eta_c$  is expressed as

$$\eta_c = \frac{\dot{m}_c C_p (T_{co} - T_{ci})}{AI} \quad (2)$$

with  $\dot{m}_c$  mass flow rate of feed (kg/s),  $T_{co}$  and  $T_{ci}$  the feed temperature at the collector outlet and inlet (K),  $C_p$  the feed specific heat (J/kg K),  $A$  the solar collector area (m<sup>2</sup>), and  $I$  the global irradiation (W/m<sup>2</sup>). The schematic representation of solar loop in the study is shown in Fig. 3. It is observed that the value of  $\eta_c$  varies between 22% and 55% for the compact system and between 15% and 35% for the large system (Banat et al., 2007). The average solar collector efficiency as computed using Eq. (2) for the system used was found to be 67% against the average reported range of 30–65%.

There is scope for achieving further energy economy in the present scheme of SDMD through the use of photovoltaic cells where solar energy could be converted into electricity to run the small pump of a water cooling system that in turn could be used in

**Table 5**  
Performance indicators of solar energy collector and membrane module.

$\dot{m}_d$ (L/h)	$\dot{m}_h$ (L/h)	$T_{hi}$ (°C)	$T_{di}$ (°C)	$\dot{m}_{evp}$ (kg/h)	$\eta_c$ (%)	Evaporation efficiency (%)	GOR	TRR
150	120	59.6	21.5	1.46	67	85	0.88	0.40

maintaining low temperature on the permeate side replacing the thermostatic water bath.

3.1.2. Evaporation efficiency of the module

As MD process is based on evaporation to perform required separation, the efficient heat in MD is considered the amount of heat required in evaporation step. One of the efficiency parameters of the MD process is evaporation efficiency that can be defined as the heat required for the evaporation versus the total heat exchanged between the feed and distillate. The evaporation efficiency can be maximized by reducing the internal heat loss by conduction through the membrane, the temperature polarization effect, and the external heat loss to the environment. Heat for evaporation ( $Q_{evp}$ ), heating energy ( $Q_k$ ) and cooling energy ( $Q_c$ ) were computed using the following equations:

$$Q_{evp} = \dot{m}_{evp} \lambda_{evp} \quad (3)$$

$$Q_h = \dot{m}_h C_{ph} (T_{hi} - T_{ho}) \quad (4)$$

$$Q_c = \dot{m}_d C_{pd} (T_{do} - T_{di}) \quad (5)$$

where  $\dot{m}_{evp}$  is the mass flow rate through the membrane (kg/h),  $\lambda_{evp}$ , the latent heat of vaporization (J/kg),  $\dot{m}_h$ , the feed flow rate (kg/h),  $\dot{m}_d$ , the distillate flow rate (kg/h),  $C_{ph}$ , the feed specific heat (J/kg K),  $C_{pd}$ , the distillate specific heat (J/kg K),  $T_{hi}$ ,  $T_{di}$  the feed and distillate temperatures (in K) at the module inlet, and  $T_{ho}$ ,  $T_{do}$  the feed and distillate temperature at the module outlet. The evaporation efficiency was found to increase with the feed temperature and by operating the system at a feed temperature of 60 °C, an evaporation efficiency of almost 85% was achieved due to high water flux against 39% in DCMD and 90% in VMD reported in earlier studies (Criscuoli et al., 2007).

3.1.3. Gained output ratio (GOR) of the system

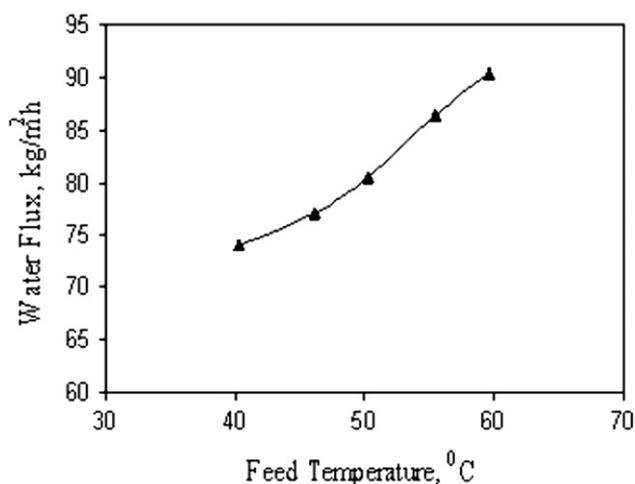
The one of the most important performance parameters used generally in thermal desalination process is the gained output ratio (GOR). It is usually defined as the heat of evaporation ( $Q_{evp}$ ) to the heating energy ( $Q_h$ ) i.e., the ratio of the latent heat of evaporation of the product water to the input thermal energy (i.e., theoretical energy required to produce the distillate divided by the actual thermal energy consumed in the feed side of the module). The GOR of the membrane module was computed using the equation

$$GOR = \frac{\dot{m}_{evp} \lambda_{evp}}{\dot{m}_h C_{ph} (T_{hi} - T_{ho})} \quad (6)$$

GOR was found to be 0.9 for feed temperature of 333 K against the observed value of GOR in the range of 0.3–0.9 in earlier studies (Koschikowski et al., 2003; Banat et al., 2007).

3.1.4. Thermal recovery ratio (TRR) of the system

One of the most important parameters of the SDMD process is how much heat it contributes to produce a specific quantity of vapors (that passing through the membrane) from the total incident radiation. The general definition of the TRR is the theoretical energy needed for distillate produced divided by the total thermal energy input. In the SPMD, the total thermal energy input is the



**Fig. 4.** Effect of feed temperature on flux of water. Experimental conditions: feed and distillate flow rates 120 and 150 L/h respectively; distillate inlet temperature 20–22 °C and arsenic concentration in feed water 396 ppb.

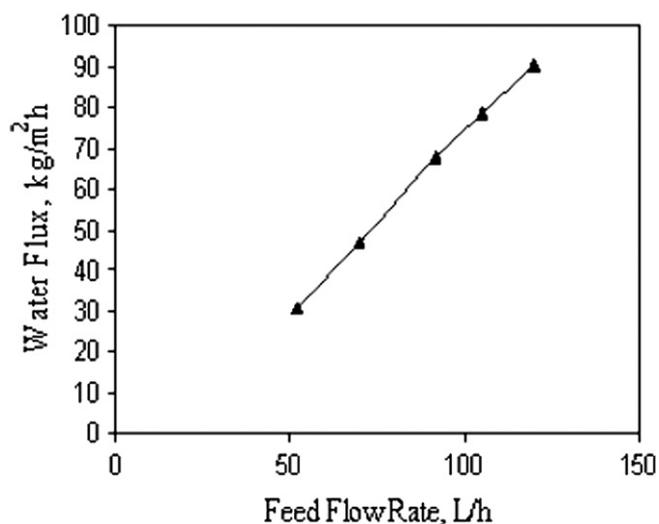
solar energy incident on the solar collector. Thermal recovery ratio (TRR) for the system was computed using the relation

$$\text{TRR} = \frac{\dot{m}_{\text{evp}} \lambda_{\text{evp}}}{AI} \quad (7)$$

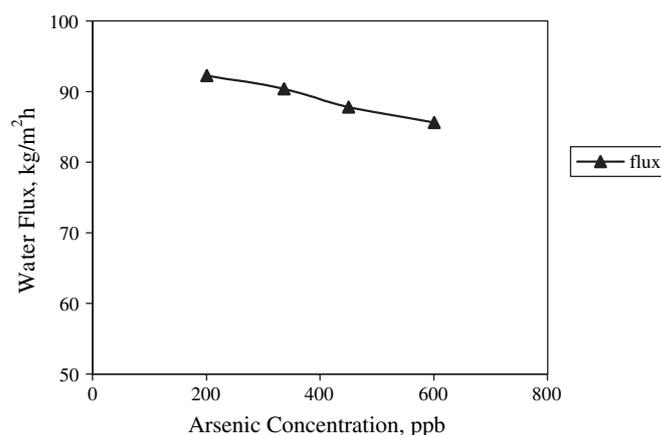
where  $I$  is the global irradiation and  $A$  is the area of solar collector. The TRR of the SPMD plant is measure of its efficiency to produce distillate. As  $T$  varies from sunrise to sunset, an average value was taken for the TRR calculation. In the present study, the TRR value was calculated to be near about 0.4 at feed temperature 60 °C which was comparable to reported range of values (0.1–0.4) in solar desalination study.

### 3.2. Effects of operating parameters on flux and permeate quality

The effects of feed inlet temperature, the feed flow rate inside the module, arsenic concentration in the feed, distillate inlet temperature and operating time on flux and separation capability of the module were studied. The average results of repeated runs on each variable have been presented in Figs. 4–8.



**Fig. 5.** Variation of flux as a function of feed flow rate. Distillate flow rate 150 L/h; feed and distillate inlet temperatures 61 °C and 20 °C, respectively; arsenic concentration in feed water 396 ppb.



**Fig. 6.** Effect of arsenic feed concentration on flux. Feed and distillate flow rates: 120 and 150 L/h, respectively; feed and distillate inlet temperatures 60 °C and 20 °C respectively.

#### 3.2.1. Effect of the hot feed inlet temperature on water flux

Fig. 4 shows that as the feed inlet temperature increased from 40 °C to 60 °C, the water vapor flux went up from 74 kg/m² h to 90 kg/m² h. Such trend of exponential rise of flux with increase of feed temperature has been reported in earlier desalination studies (Criscuoli et al., 2007; Martinez and Florido-Diaz, 2001) also. This flux enhancement following rise in feed temperature may be attributed to vapor pressure rise according to the Antoine equation

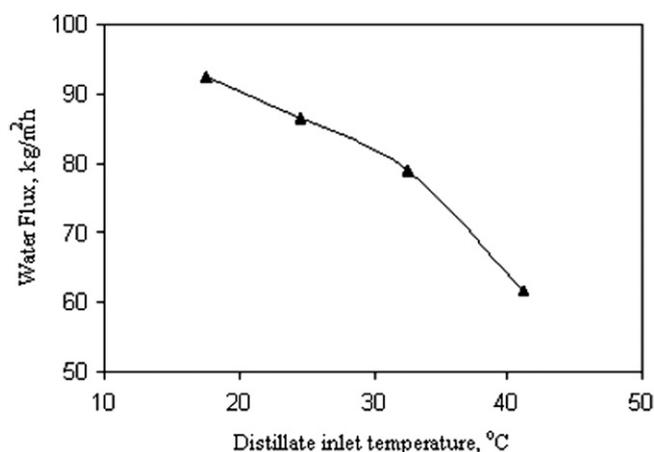
$$p^{\circ} = \exp\{23.238 - 3841/(T - 45)\} \quad (8)$$

where  $p^{\circ}$  is the vapor pressure of water in Pa and  $T$  is the temperature in K. The rise in vapor pressure directly enhances flux ( $J$ ) following the relation

$$J = K\Delta P \quad (9)$$

where the proportionality constant  $K$  is determined by membrane properties such as material and morphology, indicating porosity, tortuosity, pore radius, etc. and the transmembrane vapor pressure gradient  $\Delta P$  (difference of the feed side vapor pressure  $p_1$  and permeate side vapor pressure  $p_2$ ) is the driving force for vapor permeation through the membrane.

The solvent vapor pressure depends on temperature ( $T$ ) and concentration ( $c$ ). Therefore, the transmembrane vapor pressure gradient for isothermal system is



**Fig. 7.** Variation flux as a function of distillate inlet temperature. Feed and distillate flow rates: 120 and 150 L/h, respectively; feed inlet temperature 61 °C, arsenic concentration in feed water 398 ppb.

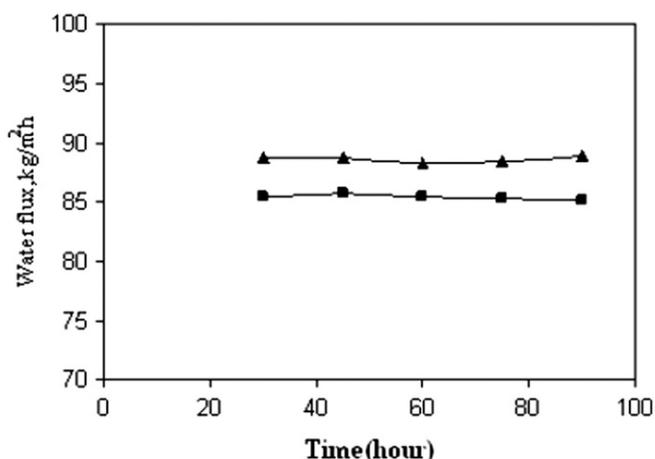


Fig. 8. Time profile of the water vapor flux for the PVDF membrane. Experimental conditions: feed and distillate flow rates 120 and 150 L/h, respectively; feed and distillate inlet temperatures 59–60 °C and 20–22 °C, respectively. Arsenic concentration in feed: 402 ppb (▲) and 605 ppb (■).

$$\Delta P = p_1(c_1, T) - p_2(c_2, T) \quad (10)$$

where the subscripts 1 and 2 represent the feed side and permeate side, respectively.

In membrane distillation process, Raoult's Law of partial pressure can be applied for calculation of partial pressure in dilute solution. Dilute arsenic solution used in the experiment can be assumed to obey colligative property. The addition of a solute (or solutes) to the solvent (pure water) lowers the free energy of the solution and we shall see that the vapor pressure ( $p^*$ ) of pure water is lowered. The colligative properties of dilute solutions primarily stem from the validity of the Raoult's Law. The two major assumptions required to use Raoult's Law are that the liquid phase is an ideal solution, and the vapor phase is an ideal gas. When the solution is dilute the solute molecules are sufficiently separated and the solution is supposed to behave ideally. The second assumption means that Raoult's Law can apply only for low to moderate pressures. According to the Raoult's law predictions, the vapor pressure of water in the solution would drop by slightly more than 2% (Cath et al., 2004).

### 3.2.2. Effect of feed flow rate on flux

Fig. 5 shows that water vapor flux increased from 30 kg/m<sup>2</sup> h to 90 kg/m<sup>2</sup> h following an increase of feed flow rate from 50 L/h to 120 L/h. Increase of feed flow rate through the flow channel means increase of velocity that led to an increase of the Reynolds number. An increased Reynolds number enhances mixing in the flow channel and decreases thickness of the feed side temperature boundary layer. Mass and energy transport enhances with an increase of feed flow rate. Therefore, the feed side boundary layer heat transfer coefficient increased with increase of feed flow rate leading to enhanced flux. The characteristic dimension  $D$  of the definition of Reynolds number ( $Re = Du\rho/\mu$ , where  $D$  is the characteristic dimension,  $u$  is the velocity,  $\rho$  is the density, and  $\mu$  is the dynamic viscosity) is the equivalent diameter of the non-circular channel of the present study. This equivalent diameter is four times the hydraulic radius (defined as the ratio of cross sectional area and wetted perimeter of the channel).

### 3.2.3. Effect of arsenic concentration on flux

In Fig. 6, it is observed that flux decreased from 91 kg/m<sup>2</sup> h to 85 kg/m<sup>2</sup> h following an increase of arsenic concentration in the feed from 200 µg/L to 600 µg/L where the standard error of the

estimate was 0.483 and the value of the adjusted  $R^2$  was 0.96 indicating that 96% of the total variation in the value of flux could be accounted for by the variation in the value of arsenic concentration in the feed. This follows from Eqs. (9) and (10) as decrease in water vapor pressure reduced transmembrane pressure gradient and lead to decrease in flux. The drop in flux here could also be partially attributed to presence of other solutes (like iron) and possible influence of concentration on viscosity that affect the polarization phenomena and transport coefficients in the feed side. Normally higher arsenic concentration in groundwater is almost always associated with higher iron concentration in the Bengal Delta basin. Considering the possible presence of other solutes, the effect of arsenic concentration on flux for the investigation range was relatively low possibly due to its marginal reducing effect on vapor pressure in DCMD as observed in some reported studies (Geucke et al., 2009; Qu et al., 2009) involving other solutes also. The major advantage of DCMD process when compared with reverse osmosis is the relatively minimal effect of feed concentration on the flux and it was well observed in the present investigation. With an increase of feed concentration in RO process, the performance of the system may significantly suffer as increased feed concentration may reduce the driving force for mass transfer across the membrane and increase the salt passage through the membrane and promotes concentration polarization, scaling, and higher osmotic pressure.

### 3.2.4. Effect of distillate inlet temperature on flux

A strong negative correlation between distillate temperature and transmembrane flux was observed. Fig. 7 shows that transmembrane flux decreased from 91 kg/m<sup>2</sup> h to 60 kg/m<sup>2</sup> h as distillate temperature increased from 20 °C to 42 °C. This sharp drop of 33% in flux following a 22 °C decrease in distillate temperature can be traced to net decrease in driving force for water vapor permeation which is well explained by Eqs. (8)–(10).

### 3.2.5. Trend of water vapor flux with time

Time profile of flux for the same membrane fitted in the module is exhibited by Fig. 8. The MD system was run for 4 days without changing membrane. Each day, the system was operated for 12 h continuously. The water vapor flux was observed to drop only by 5–6% over this time period. Arsenic concentration in the distillate was analyzed every day and no arsenic was detected. This indicates that the membrane pores were not wetted by the feed solution and the selected PVDF membrane was suitable for a reasonably long period of operation.

### 3.3. Effect of new design approach on flux

The membrane module was made of polycarbonate material having high thermal insulation property that ensured minimization of heat exchange with surroundings. The design ensured maximization of mixing on the feed side that substantially reduced effects of concentration polarization as well as temperature polarization. Hot feed entered the wide feed side channel (12 mm × 60 mm × 270 mm) through a very narrow circular conduit (4 mm diameter) and underwent flash vaporization on exposure to a low pressure in the wide channel. This ensured not only high rate of evaporation but also promoted heat transfer through better mixing and minimizes temperature and concentration polarization. The evaporation process was also facilitated by placing the feed cell at the bottom side of the module from which evaporated vapors could flow vertically upward through the microporous membrane to the cold distillate on the other side of the membrane. The distillate cell side channel having the same dimension (60 mm × 12 mm × 270 mm) of the feed channel was designed for minimization of pressure drop in the flow channel,

and maximization of fluid mixing that reduced temperature polarization. Therefore, large amount of vapor coming from feed side were easily condensed in the cold distillate. Thus high fluxes of the order of 74.0 and 90.3 kg/(m<sup>2</sup> h) were achieved for 313 K and 333 K feed temperatures respectively through 0.13 μm PVDF membrane against 13.5 and 41.5 kg/m<sup>2</sup> h fluxes reported in earlier studies (Lawson and Lloyd, 1996; Hogan et al., 1991; Hsu et al., 2002; Wirth and Cabassud, 2002) for the same feed temperature and same pore size of the membrane (PTFE). Flux in PVDF membrane of thickness 0.22 mm and 70–75% porosity was reported to be around 8 and 28 kg/(m<sup>2</sup> h) at 40 °C and 60 °C feed temperatures respectively in the investigation of Martinez and Florido-Diaz (2001). Analysis of the collected permeate in all cases has shown almost zero arsenic concentration which is extremely difficult to be achieved in other arsenic removal technologies.

#### 4. Conclusions

The SDMD module resulted in almost 100% arsenic removal from drinking water exploiting low grade solar energy with the help a locally made simple solar panel for raising feed water temperature. The cross flow, flat plate module also yielded high flux compared to earlier investigations. The module permitted easy cleaning and long hours of operation without any flux decline. The selected membrane, geometry and design, materials of cell construction (polycarbonate with poor thermal conductivity), and mode of operation – all contributed to separation efficiency, thermal efficiency and highly enhanced flux. The simplicity of design, commercial availability of membrane and abundance of solar energy of the affected study region make the present SDMD system highly applicable to community-based purification of arsenic-contaminated groundwater in the South-East Asian countries.

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#### References

- Acharya, S.K., 2002. Arsenic contamination in groundwater affecting major parts of southern West Bengal and parts of western Chhattisgarh: source and mobilization process. *Current Science* 82 (6), 740–743.
- Banat, F., Jwaied, N., Rommel, M., 2007. Desalination by a “Compact SMADES” autonomous solar-powered membrane distillation unit. *Desalination* 217, 29–37.
- Bhattacharjee, S., Chakravarty, S., Maity, S.V., Dureja, V., Gupta, K.K., 2005. Metal contents in the ground water of Sahebgunj district, Jharkhand, India, with special reference to arsenic. *Chemosphere* 58, 1203–1217.
- Brandhuber, P., Amy, G., 2001. Arsenic removal by charged ultrafiltration membrane-influences of membrane operating conditions and water quality on arsenic rejection. *Desalination* 140, 1–14.
- Cath, T.Y., Adams, V.D., Childress, A.E., 2004. Experimental study of desalination using direct contact membrane distillation: a new approach to flux enhancement. *Journal of Membrane Science* 228 (230), 5–16.
- Chakraborti, D., Mukherjee, S.C., Pati, S., Sengupta, M.K., Rahman, M.M., Chowdhury, U.K., Lodh, D., Chanda, R.C., Chakraborti, A.K., Basu, G., 2003. Arsenic groundwater contamination of in Middle Ganga Plain, Bihar, India: a future danger? *Environmental Health Perspectives* 111, 1194–1201.
- Chen, Y., Parvez, F., Gamble, M., Islam, T., Ahmed, A., Argos, M., Graziano, J.H., Ashan, H., 2009. Arsenic exposure at low-to-moderate levels and skin lesions, arsenic metabolism, neurological functions, and biomarkers for respiratory and cardiovascular diseases: review of recent findings from the Health Effects of Arsenic Longitudinal Study (HEALS) in Bangladesh. *Toxicology and Applied Pharmacology* Available on-line E pub 2009 Jan 27. PubMed PMID:19371619.6: Coll-Vinent B, Vilardell C, Font C.
- Choong, T., Chuah, T.G., Robiah, Y., Koay, F.L.G., Azni, I., 2007. Arsenic toxicity, health hazards and removal techniques from water: an overview. *Desalination* 217, 139–166.
- Chowdhury, U.K., Biswas, B.K., Roy Chowdhury, T., Samanta, G., Mandal, B.K., Basu, G.C., Chanda, C.R., Lodh, D., Saha, K.C., Mukherjee, S.K., Roy, S., Kabir, S., Quamruzzaman, Q., Chakraborti, D., 2000. Groundwater arsenic contamination in Bangladesh and West Bengal, India. *Environmental Health Perspectives* 109, 393–397.
- Criscuoli, A., Carnevale, M.C., Drioli, E., 2007. Evaluation of energy requirements in membrane distillation. *Chemical Engineering and Processing* 47, 1043–1050.
- Fagarassy, E., Galambos, I., Bekassy-Molnar, E., Gy, Vatai, 2009. Treatment of high arsenic content wastewater by membrane filtration. *Desalination* 240 (1–3), 270–273.
- Greenleaf, J.E., Lin, J., Sengupta, A.K., 2006. Two novel applications of ion exchange fibres: arsenic removal and chemical-free softening of hard water. *Environmental Progress* 25, 300–311.
- Geucke, T., Deowan, S.A., Hoinkis, J., Patzold, Ch., 2009. Performance of a small-scale RO desalinator for arsenic removal. *Desalination* 239, 198–206.
- Harvey, C.F., Ashfaq, K.N., Yu, W., Badruzzaman, A.B.M., Ali, M.A., Oates, P.M., Michael, H.A., Neumann, R.B., Beckie, R., Islam, S., Ahamed, M.F., 2006. Groundwater dynamics and arsenic contamination in Bangladesh. *Chemical Geology* 228, 112–136.
- Hsieh, L.C., Weng, Y., Huang, C.P., Li, K.C., 2008. Removal of arsenic from groundwater by electro-ultrafiltration. *Desalination* 234, 402–408.
- Hering, J.G., Elimelech, M., 1996. Arsenic removal by ferric chloride. *Journal of the American Water Works Association* 88 (4), 155–167.
- Hogan, P., Audio, A., Fane, A.G., Morrison, G.L., 1991. Desalination by solar heated membrane distillation. *Desalination* 81, 81–90.
- Hsu, S.T., Cheng, K.T., Chiou, J.S., 2002. Seawater desalination by direct contact membrane distillation. *Desalination* 143, 279–287.
- Islam, A.M., 2005. Membrane distillation process for pure water and removal of arsenic. M.Sc. thesis, Chalmers University of Technology, Sweden.
- Kimura, S., Nakao, S., Shimatani, S.I., 1987. Transport phenomena in membrane distillation. *Journal of Membrane Science* 33, 285.
- Koschikowski, J., Wiegand, M., Rommel, M., 2003. Solar thermal-driven desalination plants based on membrane distillation. *Desalination* 156, 295–304.
- Lawson, K.W., Lloyd, D.R., 1996. Membrane distillation II: direct contact membrane distillation. *Journal of Membrane Science* 120, 123–133.
- Macedonio, F., Drioli, E., 2008. Pressure-driven membrane operations and membrane distillation technology integration for water purification. *Desalination* 223, 396–409.
- Martinez, L., Florido-Diaz, F.G., 2001. Theoretical and experimental studies on desalination using membrane distillation. *Desalination* 139, 373–379.
- Nguyen, V.T., Vigneswaran, S., Ngo, H.H., Shorn, H.K., Kandasamy, J., 2009. Arsenic removal by a membrane hybrid filtration system. *Desalination* 236, 363–369.
- Pagana, A.E., Sklari, S.D., Kikkinides, E.S., Zaspalis, V.T., 2008. Microporous ceramic membrane technology for the removal of arsenic and chromium ions from contaminated water. *Microporous and Mesoporous Materials* 110, 150–156.
- Panthi, S.R., Sharma, S., Mishra, A.K., 2006. Recent status of arsenic contamination in groundwater of the Terai region of Nepal. *Kathmandu University Journal of Science, Engineering, and Technology* 1, 1–11.
- Pal, P., Ahamad, Z., Pattanayak, A., Bhattacharya, P., 2007b. Removal of arsenic from drinking water by chemical precipitation – a modeling and simulation study of the physical-chemical processes. *Water Environment Research* 79 (4), 357–366.
- Pal, P., Ahamad, Z., Bhattacharya, P., 2007a. ARSEPPA: visual basic software tool for arsenic separation plant performance analysis. *Chemical Engineering Journal* 129, 113–122.
- Phattaranawik, J., Jiratananon, R., Fane, A.G., 1998. Heat transport and membrane distillation coefficients in direct contact membrane distillation. *Journal of Membrane Science* 144, 211–222.
- Pokhrel, D., Bhandari, B.S., Viraraghavan, T., 2009. Arsenic contamination of groundwater in the Terai region of Nepal: an overview of health concerns and treatment options. *Environmental International* 35 (1), 157–161.
- Qu, D., Wang, J., Hou, D., Luan, Z., Fan, B., Zhao, C., 2009. Experimental study of arsenic removal by direct contact membrane distillation. *Journal of Hazardous Materials* 163 (2–3), 874–879.
- Roy, M., Nilsson, L., Pal, P., 2008. Development of groundwater resources in a region with high population density: a study of environmental sustainability. *Environmental Sciences* 5 (4), 251–267.
- Schreiber, M.E., Simo, J.A., Freiberg, P.G., 2000. Stratigraphic and geochemical controls on naturally occurring arsenic in groundwater, eastern Wisconsin, USA. *Hydrogeology Journal* 8, 161–176.
- Schofield, R.W., Fane, A.G., Fell, C.J.D., 1987. Heat and mass transfer in membrane distillation. *Journal of Membrane Science* 33, 299.
- Shih, M.C., 2005. An overview of arsenic removal by pressure-driven membrane processes. *Desalination* 172, 85–97.
- Wickramasinghe, S.R., Han, B., Zimbron, J., Shen, Z., Karim, M.N., 2004. Arsenic removal by coagulation and filtration: comparison of ground waters from the United States and Bangladesh. *Desalination* 169, 231–244.
- Wirth, D., Cabassud, C., 2002. Water desalination using membrane distillation: comparison between inside/out and outside/in permeation. *Desalination* 147, 139–145.
- World Health Organization, United nations synthesis report on arsenic in drinking water, [http://www.who.int/water\\_sanitation\\_health/dwq/arsenic3/en/](http://www.who.int/water_sanitation_health/dwq/arsenic3/en/).
- Xia, S., Dong, B., Zhang, Q., Xu, B., Gao, N., Causseranda, C., 2007. Study of arsenic removal by nanofiltration and its application in China. *Desalination* 204, 374–379.