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Gray water treatment by air-gap multi-effect membrane distillation with pre-treatment by electrocoagulation

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Abstract

The gap between supply and demand of fresh water is widening due to increase in global population. Hence, an alternative source of water such as gray water could potentially save a significant amount of precious fresh water. Gray water is distinct from black water by the amount and composition of its chemical and biological contaminants. In this paper, an air-gap Multi-effect membrane distillation (MEMD) module performance for gray water treatment is described. Surfactant present in the gray water was wetting the membrane pores. Hence, electrocoagulation was used as a pre-treatment of gray water feed. About 99.14% surfactants were removed by electrocoagulation and the experiment shows excellent performance of the MEMD module. The 4-stage MEMD module offered permeate fluxes nearly to 50.12 l/m^2 h at 80 °C. It was also found that a high thermal efficiency and output gain ratio was possible with lower specific energy and no cooling water. Hence, it is apparent that the air gap MEMD technology could be used for gray water treatment. Pre-treatment by electrocoagulation (EC) operation seems an important step in the overall treatment process when large amounts of surfactants are present in the treatment water.

Key words: electrocoagulation, gray water, MEMD, membrane distillation, water treatment

Highlights

- The air-gap MEMD module is developed for gray water treatment.
- The electrocoagulation is used as a pre-treatment and achieved 99.8% removal of surfactant.
- The effects of feed temperature, flow rate and operating time on permeate flux of MEMD module is performed.
- High thermal efficiency and GOR found of air gap MEMD module.
- Low consumption of specific heat was found in MEMD module.

INTRODUCTION

Recently, use of gray water as an alternative water resource has drawn attention. It is non-industrial waste water generated from domestic processes such as washing dishes, laundry and bathing. Gray water is distinct from black water in the amount and composition of its chemical and biological contaminates (from feces or toxic chemicals). Dish, shower, sink, and laundry water comprise 50–80% of residential waste water (Emerson 1998; Intizar *et al.* 2002). But the government of Western Australia has not recommended the gray water from kitchen for reuse due to high levels of organic materials such as oils and fats. Gray water contains higher level of salts, boron, surfactants and oils and

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hence it has harmful effects on soil, plants and underground water (Faisal 2011). There are various methods currently used for gray water treatment such as: (i) Reverse osmosis, (ii) Slow sand filter, (iii) Membrane separation, (iv) Micro filtration, (v) Water softening, (vi) Coagulation and Filtration method (Peprah *et al.* 2018), and (vii) Constructed wetland (Maria *et al.* 2000).

Lately, the membrane thermal driven process, known as membrane distillation (MD) is based on hydrophobic micro-porous membrane material similar to ultrafiltration. It has gained popularity in the academics and scientific communities due to its excellent ability to retain the non-volatile solutes while still producing high quality treated water. Literature shows an excellent performance of MD in desalination and waste water treatment processes (Bonyadi *et al.* 2009; Wang *et al.* 2009; Prince *et al.* 2012). With the MD technology, pollutants or impurities in water are separated through evaporation by thermal energy. The evaporated feed is then passed through hydrophobic micro porous membrane, and the vapors are condensed, in permeates side, by a cooling process. The driving force from feed to permeate is driven by the vapor pressure difference created due to temperature differences. Due to the capillary action in membrane pores, direct mixing of the vapor and liquid phases are prevented (Khayet & Matsuura 2011; Pangarkar *et al.* 2016a). The hydrophobic nature of membrane also helps in preventing the penetration of any aqueous solution into the pores, hence forming the vapor-liquid interface that is responsible for the separation mechanism (Lindsey & Miller 2002; Meindersma *et al.* 2006).

MD shows the following limitations (Schofield *et al.* 1990; Alkhudhiri *et al.* 2012): (i) Low permeate flux due to temperature and concentration polarization in MD, (ii) increased heat loss due to conduction in MD, (iii) less availability of commercial membranes as well as correct design of MD module, (iv) wetting of membrane pore risk is high, (v) high energy consumption as compared to RO, (vi) membranes used in MD are expensive, (vii) higher cooling water consumption.

To overcome the above limitations requires the implementation of the multi-effect concept in a traditional MD module. As referred by Liu *et al.* (2012), Zhao *et al.* (2013), Lu *et al.* (2012), Pangarkar & Deshmukh (2015), and Pangarkar *et al.* (2016b), the following are the advantages of MEMD over traditional MD: (i) Less energy is required for heating the feed water due to internal latent heat recovery, (ii) low grade waste heat or solar energy can be used efficiently, (iii) high rate of water production due to the number of stages, (iv) less cooling water consumption, (v) the MEMD process has a high thermal efficiency, gain output ratio (GOR), and is stable and reliable, (vii) it has lower maintenance cost and 24 hours continuous operation with minimum supervision.

In this study, the air-gap MEMD process was used for gray water treatment, while electrocoagulation (EC) was included as a pretreatment to reduce membrane fouling due to the presence of surfactants in the water feed.

MATERIALS AND METHODS

Gray water and characteristics

The composition of gray water varies with the type and choice of chemicals used for cleaning, laundry, bathing and it is mostly a reflection of the lifestyle. Also, the chemical and microbial quality of gray water is dependent upon source types, place, time of discharge and quantity. A total of six samples of gray water were collected on Monday at 7.00 am and 7.00 pm for three weeks from bathrooms and basins of a residential college campus located in Sinnar rural area in Nashik city, India. A typical characterization of gray water samples is summarized in Table 1. A water analysis kit (Systronics, Type-371) was used for analysis of the characteristics of samples such as pH, total dissolved solids (TDS), total hardness, total soluble solids (TSS). All other parameters were measured in the industrial R&D laboratory located in Nashik city (India).

Table 1 | Average chemical composition of bathrooms, basins of gray water from a residential area in Nashik city, India

Sr. No.	Parameters	Unit	Raw water (average value)
1	pH	–	8.04
2	Total hardness	mg/l	428
3	COD	mg/l	334
4	TDS	mg/l	741
5	TSS	mg/l	172
6	Surfactant	mg/l	36.3
7	Chlorine	mg/l	46.3
8	Nitrites	mg/l	0.11
9	Nitrates	mg/l	0.63
10	Sulphates	mg/l	23.1
11	Sodium	mg/l	59.42
12	Potassium	mg/l	2.92
13	Magnesium	mg/l	0.11
14	Ammonia-nitrogen	mg/l	0.79
15	Calcium	mg/l	0.14

Electrocoagulation setup

A plastic electrocoagulation cell (1.5 liter capacity) with dimensions of 200 × 100 × 150 mm was used for the experiment. Aluminum sheets (200 × 75 × 2 mm) as electrodes were employed in an electrocoagulator unit and DC power supply was used for magnetic stirring at nearly 400 rpm.

Membrane material

Polytetrafluroethylene (PTFE) type of flat sheet membrane was used in this experiment. The detailed characteristics of membrane are shown in Table 2.

MEMD experimental setup and procedure

A MEMD module was constructed by using acrylic material and detailed assembly is shown in Figure 1(a)–1(h). In the module, three feed channels, two cooling channels and four permeate channels were used and the detailed dimensions of each channel are shown in Table 3. The membrane sheet was inserted between the feed flow channels and permeate channel. Aluminum foil and rubber gasket (2 mm thick) was used for the purpose of condensation of water vapor and creating

Table 2 | PTFE membrane characteristics

Particulars	Membrane characteristic
Manufacturer	GmbH, Germany
Pore size	0.45 μm
Porosity	70%
Thickness of sheet	175 μm
Tortuosity	2
Effective membrane area for single stage	80 cm^2
Effective membrane area for 4-stage	320 cm^2

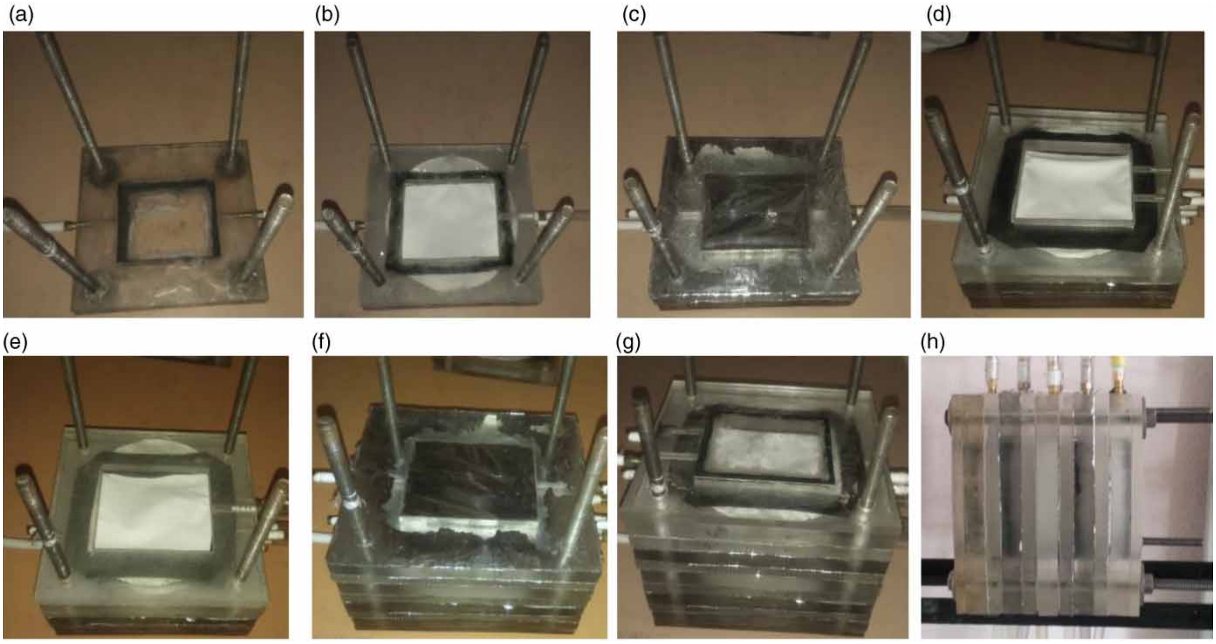

Figure 1 | (a–h) Construction procedure of 4-stage MEMD module.

Table 3 | Dimensions of MEMD flow channels

Channel in module	Size (mm)
Feed	100 × 80 × 5
Cooling	100 × 80 × 5
Permeate	100 × 80 × 5

air gap thickness respectively. In the MEMD module, the arrangement of the plates and membranes of up to 4 stages is shown in Figure 2. The constructed 4-stage MEMD module pump of 0.5 hp was used for circulating the water from feed tank to the module, with dimensions of nearly 180 × 160 × 60 mm, which was used in the experimental setup, as shown in Figure 3. Two feed tanks of 20 liter capacity had the cooling and heating system used in the experimental setup. Circulating Pt100 sensors were used in thermocouples for measuring the temperature of water at various locations in this setup.

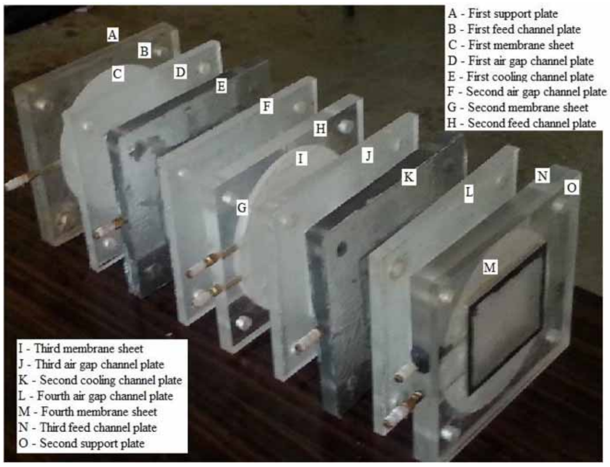

Figure 2 | Plates and membrane sheets arrangement in 4-stage MEMD module.

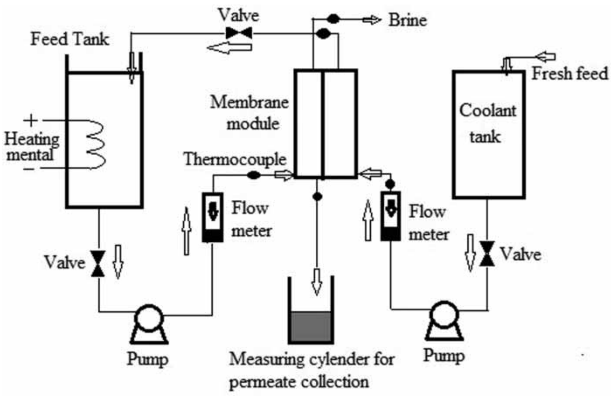

Figure 3 | MEMD experimental setup.

Performance parameters such as permeate flux, specific energy consumption for heating of the feed, gain output ratio (GOR) and thermal efficiency were evaluated by means of Equations (1)–(5), below.

$$\text{Permeate flux } (J_D) \text{ in L/m}^2\text{h of MEMD,} \quad J_D = \frac{V}{A\,t} \tag{1}$$

$$\text{Separation factor (SF) of MEMD,} \quad \%\,SF = \frac{C_f - C_p}{C_f} \times 100 \tag{2}$$

$$\text{Specific energy consumption (E) in kWh/kg,} \quad E = \frac{m_f\, C_{pf}\, (T_f - T_o)}{m_D} \tag{3}$$

$$\text{Gain Output Ratio (GOR) of MEMD,} \quad (GOR) = \frac{m_D\, \Delta H_v}{m_f\, C_{pf}\, (T_f - T_o)} \tag{4}$$

$$\text{Thermal efficiency, } \eta\ (\%), \quad \eta\% = \frac{m_D\, \Delta H_v}{m_f\, C_{pf}\, (T_f - T_{B3})} \tag{5}$$

where, V (L) is volume of permeate collection at time t (h); A (m^2) is membrane area; C_f and C_p represent concentrations in the feed and permeate respectively; m_f and m_D (Kg/s) are mass flow rate of feed and permeate, in that order; T_f, T_O & T_{B3} correspond to feed temperatures during circulation through the 1st feed channel, water after recovery of hot brine heat and output brine water, respectively. C_{pf} (KJ/kg °C) is the specific heat capacity of water, while ΔH_v (KJ/kg) constitutes heat of vaporization of water.

RESULTS AND DISCUSSION

Electrocoagulation: pretreatment operating parameters

A high removal rate was achieved for surfactants through coagulation/flocculation by using aluminum (Al) electrodes. In order to treat gray water with a certain pH by EC, appropriate current density and operating time need to be selected (Paur 2014). In the electrocoagulation cell, the gap between the electrodes was maintained at 5 mm. Current density and operating time were varied from 5–60 A/m^2 and 0–40 min respectively. Feed water circulation rate was maintained at 0.5 l/min. Samples were analyzed for the removal of detergent at each interval of the time and different current density. Figure 4 reveals that higher surfactant removal (99.8%) could be achieved by

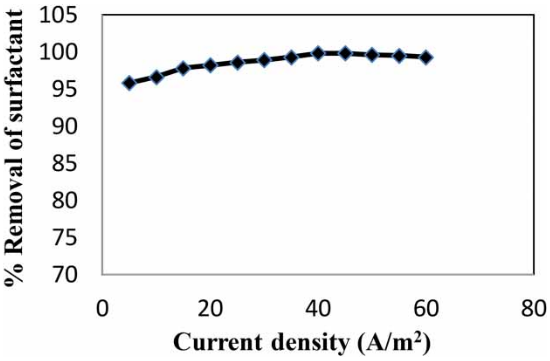

Figure 4 | Effect of current density on removal of surfactant from gray water at 0.5 l/min of feed water circulation rate.

increasing current density up to 40 A/m^2. But the removal rate remains unchanged at higher values of current densities. A similar result was found by Batoul *et al.* (2017) for anionic surfactant removal. Figure 5 shows the effect of time on surfactant removal efficiency. During 10 min of treatment time, removal efficiency reached 99.6% due to increasing the amount of metal hydroxide flocs, which increases the removal efficiency via coagulation followed by precipitation. Further increase of the time had no effect on process efficiency. Hence, the optimum operating conditions such as 40 A/m^2 current density, 10 min operating time and 0.5 l/min circulation rate were used in the experimentation for pre-treatment of gray water.

Effect of temperatures and flow rate on the MEMD permeate flux

Figure 6 shows permeate flux enhancement due to the increase of temperature in the feed, as higher temperatures in feed water show positive effects in the MD process. This seems to be a response of the temperature difference between the feed and coolant water, generating vapor pressure across the membrane, thus increasing the driving force of the permeate flux. During the experiment, the feed temperature was increased from 40 to 80 °C and coolant water temperature was kept constant about 27 °C. The feed water flow rate was kept at 0.5 l/min. The coolant water flow rate was kept at 0.25 l/min for each coolant channel. Thickness of the air gap and feed channel depth were kept constant at 2 and 5 mm respectively. Increasing the feed temperature from 40 to 80 °C, led to increase in the permeate flux from 10.22 to 50.12 l/m^2 h.

The permeate flux of MD also depends on the feed flow rate. The heat transfer coefficient increases in hot feed streams and reduces the temperature, resulting in the concentration polarization effect by increasing feed rate (Guijt *et al.* 2005). Increase in feed rate means the Reynolds number, which increases the turbulence in the feed channel. Hence, the permeate flux increases with the feed flow

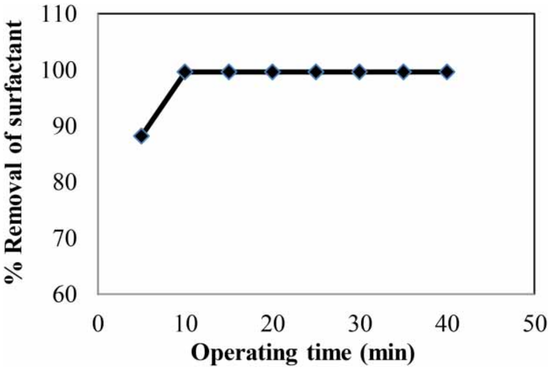

Figure 5 | Removal of surfactant with time at 40 A/m^2 current density and 0.5 l/min feed water circulation rate.

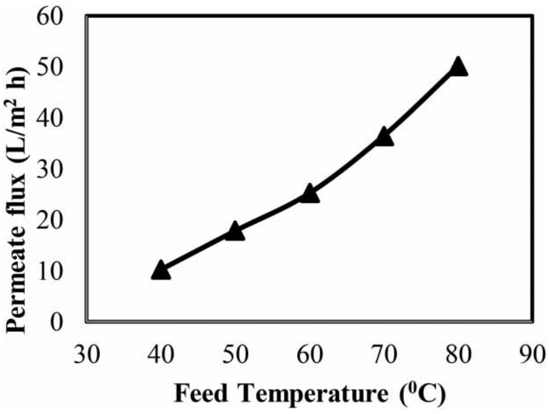

Figure 6 | Effect of feed temperature on MEMD permeate flux.

rate. If feed rate is higher than the needed rate, then energy consumption for preheating the feed will be greater. This energy is unnecessarily wasted and it also creates extra unnecessarily high temperature brine. At lower feed rate, production of over-concentrated feed and crystallization of chemicals could occur inside the module. Hence, the life span of the module might reduce (Zhao *et al.* 2013). Hence it is necessary to determine the optimum feed rate.

Flux production of the MEMD module was observed at various feed rates ranging from 0.3 to 0.5 l/min at a constant feed temperature of 80 °C, while keeping a coolant flow rate of 0.25 l/min and temperature of 27 °C on every coolant channel. Figure 7 shows that enhanced feed rates resulted in boosting flux from 24.9 to 50.1 l/m^2 h. Further rising feed flow rates (i.e. 0.8 l/min) caused a marginal increase in permeate flux from 50.1 to 53.2 l/m^2 h. Hence, it was concluded that the module optimum operation was observed at feed flow rates of 0.5 L/min, feed water temperature of 80 °C and coolant flows of 0.25 l/min at 27 °C.

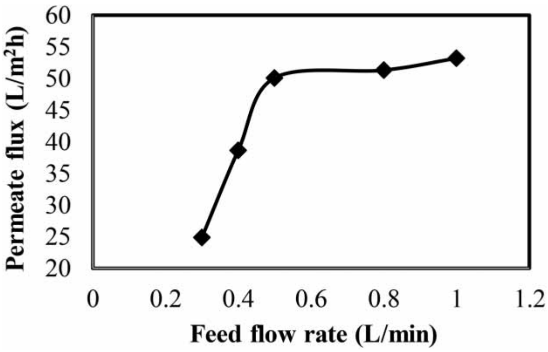

Figure 7 | Effect of feed flow rate on MEMD flux.

Effect of time on MEMD performance

Membrane fouling in MD is recognized as a result of deposition of soluble salts (Gryta 2008; He *et al.* 2008), biological compounds like protein (Goh *et al.* 2013), and carbohydrates (Mokthar *et al.* 2015) on the membrane surface and membrane wetting by the feed water. Fouling of the membrane was measured by measuring the permeate flux with time and is shown in Figure 8. The experiment was carried out for about 90 hours, continuously. Fresh feed water at 28 °C was used as a coolant passed through the coolant channels. The feed flow rate was kept at 0.5 l/min. Initial permeate flux was recorded at about 50.12 l/m^2 h, with a noticeable decrease of about 14% within the first initial

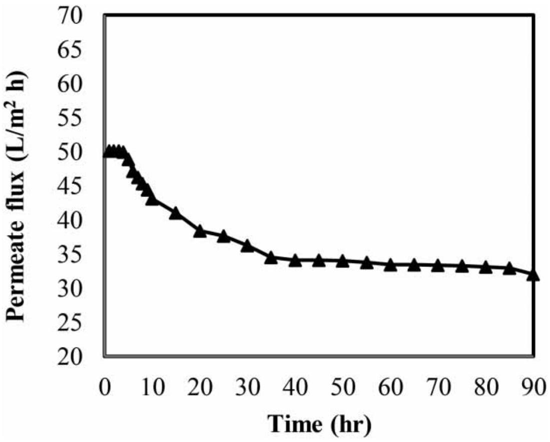

Figure 8 | Effect of operating time on MEMD flux with pre-treatment of gray water by electrocoagulation.

period of 10 hours followed by a less step flux decline and distinctively a more consistent flux from 30 to 90 hours of operation, where 32.02 l/m^2 h developed. Total flux decline was recorded around 36.12%.

Separation efficiency of module

Figure 9 shows the removal efficiency of the pretreated gray water by electrocoagulation and the MEMD module. Electrocoagulation removal efficiencies of surfactants were close to 99.14%, with overall EC efficiencies ranging from 60 to 88%. On the other hand, the removal efficiency of the MEMD module was higher than 99.2%. Permeate conductivity was found to be 1.5 μs/cm. The distillate rendered by the 4-stage MEMD module was of high quality, and potentially for household and industrial uses.

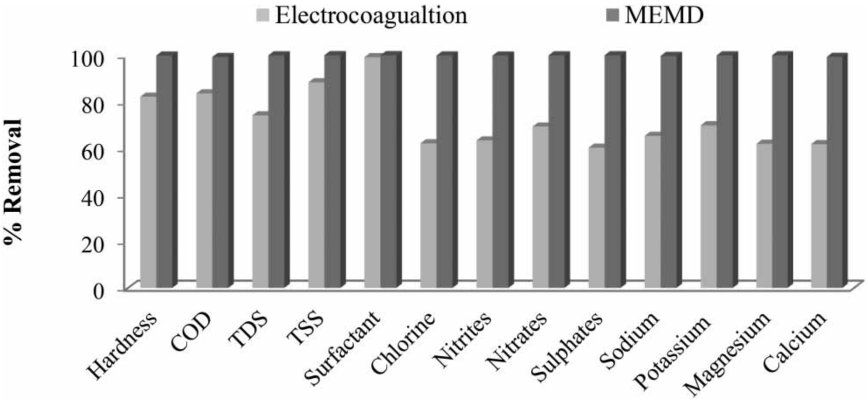

Figure 9 | % removal of gray water characteristics by electrocoagulation and MEMD module.

Energy and economic efficiency of the module

The number of stages in the MEMD module is a critical parameter because it has an effect on the performance parameters of MEMD such as thermal efficiency, energy consumption, GOR, product rate, size and the investment cost of the whole system. In the investigation of performance parameters, fresh feed is used as a cooling water and latent heat was recovered during the condensation of permeate vapor. The performance of the MEMD module for 1st, 2nd and 4th stages are shown in Table 4. On increasing the number of stages 1–4, the flux was reduced due to increase in the membrane area; however, it increases the production rate. Hence the product rate was dependent on the membrane

Table 4 | Performance comparison of the MEMD system with different numbers of stages

Particulars/stages	1st	2nd	4th
Membrane area (m^2)	0.008	0.016	0.032
Flux (L/m^2 h)	53.56	53.28	50.12
Specific energy consumption (kWh/kg)	3.17	1.63	0.57
GOR	0.21	0.43	1.12
Thermal efficiency (%)	154.36	246.65	341.22

area. The specific energy consumption in 1st – stage was about 3.17 kWh/kg, which decreased with increase in the number of stages and it was found to be nearly 0.57 kWh/kg in the 4-stage MEMD. It was also found that the GOR was increased with increase in the number of stages and the maximum GOR reached about 1.12 in the 4-stage MEMD. Most of the experimental MD systems have GOR values less than unity indicating low MD performance because these systems are operated in a single stage (Khayet 2013).

In the economic performance, the water production cost (WPC) of various MD system varies in large magnitude from 220 Rs/m^3 to 8,934 Rs/m^3. This is due to change in the different MD modules, configuration and membrane used. The WPC of the MEMD pilot plant was calculated as about 790 Rs/m^3 (0.79 Rs/L) water production. If the waste heat is available in the industry, the thermal cost required for heating the feed water will be reduced.

CONCLUSION

Membrane pore wetting due to the presence of surfactant in the gray water was one of the biggest problems. The appropriate pretreatment system of electrocoagulation was used, which resulted in removal of nearly 99.14% surfactants. The internal heat recovery was achieved in the design of the MEMD module. The obtained result shows the excellent performance of the MEMD module in terms of the separation efficiency and energy efficiency of the system. Hence, the air gap MEMD process, with electrocoagulation as a pre-treatment, represents good potential for gray water reclamation.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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