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Investigation on the application of polysulfone-based bipolar membrane for desalination of water

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ABSTRACT

The present paper focuses on the application of a novel and green separation technology, (i.e.) bipolar membrane (BPM) electrodialysis process, in a laboratory scale for seawater treatment using polysulfone (PSu) based synthesized membranes and a commercial membrane based on polystyrene divinyl benzene. All the experiments performed for both the groups were under similar conditions. This study was designed to evaluate the efficiency of the synthesized BPMs using pH, TDS, and conductivity changes in the acid and base compartments. From the stack performance, it was observed that the synthesized membranes showed a greater current efficiency (21.5%) and transport number of ions (0.143) than commercial membranes (10.7% and 0.0714). Analysis of the experimental results suggested that the PSu-based system exhibited higher performance efficiency than the commercial-based systems. This was further confirmed by the analysis of acid/base production, water dissociation flux, and other associated parameters.

Keywords: Acid/base production; Bipolar membrane electrodialysis; Conductivity change; Water dissociation efficiency

1. Introduction

Interest in ion exchange membranes (IEMs) is essentially due to their large varied applications, especially in separation processes such as dialysis [1], Donnan dialysis [2], and electrodialysis [3]. The advantages of membrane processes over conventional separation techniques include low energy consumption; minimal requirement of chemical reagents; and importantly, their amenability for automation. Hence, the novel and green separation technology, known as bipolar membrane electrodialysis (BPMED) technology, has garnered lot of interest recently. Electrodialysis is generally considered as the most economical process for the production of potable water from brackish water when the salt concentration is relatively low (less than 5,000 ppm) [4–10]. In the Third World countries, demineralized brackish underground water being a major source of drinking water, the BPMED technology could be exploited to increase the accessibility to potable water.

From the literature, it was evident that many researchers have based their worked on the desalination of seawater using electrodialysis process [11–14]. One significant feature of electrodialysis is that the salt can be concentrated to a comparatively high value (in excess of 18–20% by weight) without severely affecting the economics of the process. BPMED combines both the conventional electrodialysis as well as water dissociation through bipolar membranes (BPMs) such that

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under an applied electric field H_2O (water) splits into H^+ and OH^-. Thus, according to Haerens [15], due to its specific advantages, BPMED is considered as a well-developed membrane separation technology for desalination of aqueous solutions or concentrated industrial streams which can also be used to recycle valuable components from wastes [16–18] by providing acid or base sources *in situ* with high efficiency through simple operational methods. When combined with other membrane separation processes or chemical unit operations, electrodialysis can improve the overall process sustainability. It should be noted that, the efficiency of BPMED process is determined by the quality of bipolar and monopolar (AEM and CEM) membranes which constitute the repeat units in an ED stack.

Among the many methods reported in the literature for the preparation of IEMs (BPM), the paste method [19] has been frequently adopted as the resulting membranes exhibit good physiochemical properties. The important part of a BPM is its intermediate layer. It is known that BPMs have either nil or negligible water dissociation efficiency (WDE) if they are prepared with cation- and anion-exchange layers alone. In order to improve the performance of a BPM, usually a thin intermediate layer, capable of acting as a water dissociation catalyst, is generally introduced between the layers [19]. Different types of intermediate layers, including a matrix material containing quaternary and non-quaternary amine group, weak acid, and its corresponding bases, such as pyridines, carboxylic acid, phenolic, and phosphoric acid groups, have been employed by various investigators [20–22]. In addition, because of their amphoteric characteristics, inorganic substances such as sodium meta silicate, ruthenium trichloride, titanium oxide, manganese oxide, zeolite, chromic nitrate, and indium sulphate have also been tried [19]. The usage of some metallic compounds/heavy metal ions, such as Fe^{2+}, Fe^{3+}, Ti^{4+}, Sn^{2+}, Pd^{2+}, Ru^{3+}, etc. [20], have also been reported. In the present work, the water dissociation catalyst used was polyvinyl alcohol (PVA).

Many research institutes such as the Central Salt and Marine Chemicals Research Institute, Bhavnagar (India), have installed more than 2000 brackish water desalination plants with a total capacity of producing more than 1,000,000 m^3 of potable water per day from brackish water [23]. This is in addition to the setting up of electrodialysis desalination systems powered by photovoltaic cells that were developed and installed between 1985 and 1995 [24,25]. It is worth noting that a few ED or electrodialysis reversal plants were successfully commercialized in the USA, Russia, and China during the 1970s [23,26]. Recent developments include the apparatus for seawater desalination by electrodialysis using IEMs for converting seawater into potable water [27–29].

In the present study, the work was designed to evaluate the desalination efficiency of polysulfone (PSu) based BPM on Chennai seawater (Bay of Bengal) in terms of current efficiency, energy consumption, acid and base concentrations, transport number of the ions, conductivity and pH changes, WDE, and water dissociation fluxes in the acid and base compartments (ACs and BCs). Finally, the efficiency of the laboratory synthesized, relatively inexpensive BPM, made from stable cation-exchange and anion-exchange functional groups with PVA as the intermediate, was evaluated by observing the change in pH, TDS, and conductivity values. The experimental results were compared with those for the commercially available polystyrene divinyl benzene (PSDVB) membranes in a five-compartment electrodialytic cell. In addition the variation in the potential with respect to time was also measured.

2. Experimental

2.1. Materials and methods

A summary of the commercial membranes, together with their properties and top layer composition as specified by the manufacturer, is given in Table 1. Besides the commercial membranes, lab-prepared membranes were also used. These membranes (composed of PSu) were fabricated according to the methodology described in our previous paper [30]. The physicochemical and instrumental characterizations of the prepared membranes such as IEC, water absorption, FTIR, and SEM have also been discussed in our previous reports [30–32]. Commercially available IEMs such as CMI—7000S and AMI—7001S were purchased from Membranes International INC, New Jersey, USA, while the PSDVB (Gel polystyrene cross-linked with divinylbenzene) BPM was procured from Arun electro chemicals, Chennai.

2.2. Principle and characterization of BPM

The water dissociation capabilities of the BPM were evaluated using experiments in a laboratory-scale fabricated two-compartment electrodialytic cells. Since a BPM consists of two (anion and cation exchange) layers joined together using catalytic PVA intermediate, when it is placed in an external electric field, hydrogen and hydroxyl ions are generated, thus resulting in acidification/alkalization of the adherent solution. The water dissociation capabilities were measured in terms

Table 1
Overview of the used commercial and synthesized membranes with specifications

Type of ion-exchange membrane	Membrane properties	Laboratory synthesized membrane	Commercially procured membrane
AEM (anion-exchange membrane)	Anion-exchange membrane functionality	Strong base Anion-exchange Membrane	Strong base Anion-exchange Membrane
	Membrane code	QPSu	AMI—7001S
	Polymer structure	PSu	Gel polystyrene cross linked with divinylbenzene
	Functional group	Quaternary ammonium	Quaternary ammonium
	Ionic form	Chloride	Chloride
	Color	Brown	Light yellow
	Thickness	0.099 ± 0.05 mm	0.45 ± 0.025 mm
	Thermal stability (°C)	>198	90
	Chemical stability range (pH)	1–12	1–10
	Reinforcement	No	Yes
	Total exchange capacity (meq/g)	1.57	1.30
	Water absorption (%)	5.9	3.7
CEM (cation-exchange membrane)	Cation-exchange membrane Functionality	Strong acid Cation-exchange Membrane	Strong acid Cation-exchange Membrane
	Membrane code	SPSu	CMI—7000S
	Polymer structure	PSu	Gel polystyrene cross linked with divinylbenzene
	Functional group	Sulfonic acid	Sulfonic acid
	Ionic form	Sodium	Sodium
	Color	Cloudy white	Brown
	Thickness	0.12 ± 0.05 mm	0.45 ± 0.025 mm
	Thermal stability (°C)	>240	90
	Chemical stability range (pH)	1–12	1–10
	Reinforcement	No	Yes
	Total exchange capacity (meq/g)	1.47	1.60
	Water absorption (%)	7.3	6.5
BPM (bipolar ion-exchange membrane)	BPM functionality	Strong acid cation & base anion-exchange membrane	Strong acid cation & base anion-exchange membrane
	Membrane code	BPMPSu	PSDVB
	Polymer structure	PSu	Gel polystyrene cross linked with divinylbenzene
	Functional group	Sulfonic acid & quaternary ammonium	Sulfonic acid & quaternary ammonium
	Ionic form	Sodium & chloride	Sodium & chloride
	Color	Brownish yellow	Light yellow
	Thickness	0.20 ± 0.05 mm	0.85 ± 0.025 mm
	Thermal stability (°C)	>180	90
	Chemical stability range (pH)	1–12	1–10
	Reinforcement	No	Yes

Table 2
Details of electrodialysis system

Parameter	Values/material used
Number of compartments	2
Volume of each chamber	160 mL
Anode	Titanium base metal coated with Ti–Ru–Pd oxides
Cathode	Stainless steel
Current	0–1.5 A
Voltage	0–50 V
Cell dimension	12 cm × 10 cm
Effective membrane area	120 cm^2

of water dissociation flux (mol m^{-2} s^{-1}), using the following equation (1) [32] which is indicative of the amount of proton/hydroxyl ion present on the surface of the IEM, during the experiments in ACs/BCs, respectively.

$$\text{Water dissociation flux } J_{H^+ \text{or } OH^-} = \frac{V \times \left(\frac{dc_{H^+ \text{or } OH^-}}{dt}\right)}{A} \quad (1)$$

where V is the total volume (cm^3); $J_{H^+ \text{or } OH^-}$ is the water dissociation flux (mol m^{-2} S^{-1}); $dc_{H^+ \text{or } OH^-}$ is the change in acid (H^+) or base (OH^-) concentration in AC and BC with time; t is the operation time (sec); and A is the effective membrane area.

The cell configurations are illustrated in Fig. 1. The two compartments were separated by the PSu-based BPM having an effective membrane area of 120 cm^2 at the center. The membrane was placed in such a way that the anionic exchange layer (AEL) of the BPM faced the anode while the cationic exchange layer (CEL) of the BPM faced the cathode. The neighboring compartments, having a total volume of 160 cm^3, were initially filled with distilled water. However, with increase in the duration of the operation of the instrument, it was observed that the pH of one of the compartments (with the cathode) changed to acidic nature while the other compartment (with anode) changed to basic nature. The experiment was carried out for 5 h to determine the changes in pH, conductivity, and TDS measurements in both ACs as well as BCs.

2.3. Electrodialysis stack

Fig. 2 and Table 2 illustrates the configuration of the BPMED, (Arun electro chemical, Chennai) used for the present study. The stack comprised of (a) cathode (made of stainless steel) and anode (made of Ti coated with TiRu–Pd oxides) electrodes (b) monopolar (cation exchange and anion exchange), and BPMs, each having an effective membrane area of 120 cm^2.

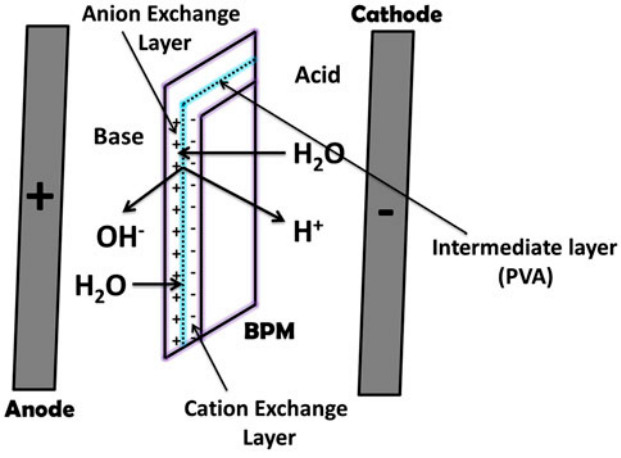


Fig. 1. Two-compartment electrodialytic laboratory-scale equipment for pilot water dissociation test to determine the efficiency of BPMPSu membrane.

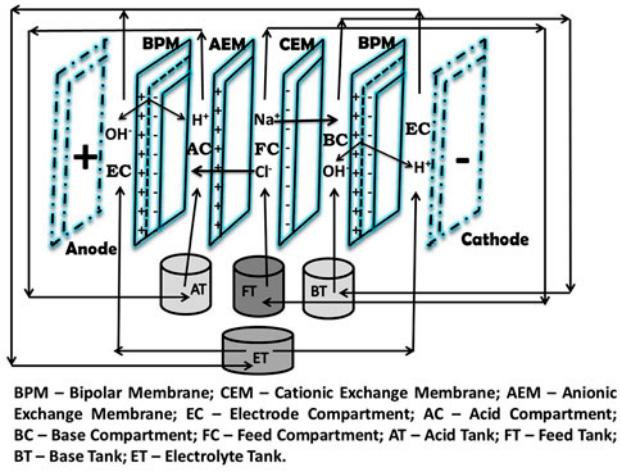


Fig. 2. Technological process like ion transport involved in a five-compartment BPMED stack for acid/base productions.

These membranes were positioned in between the electrodes such that the stack comprised a total of five compartments. The volume of each chamber was about 160 cm^3. All the membranes in the stack were separated by a spacer–gasket arrangement. The configuration of the membrane arrangement was in the order BP-A-C-BP in between the electrodes. The two electrodes were connected to a direct current power supply. Each compartment was connected to a tank of 1 L capacity, allowing for batch mode recirculation of external solutions by submersible pumps.

In this experiment, a NaCl solution (0.05 mol/L) was used as the supporting electrolyte solution in each electrode compartment/tank while seawater from Chennai beach, having minute concentrations of certain elements such as oxygen, hydrogen, sodium, chlorine, bromine, magnesium, calcium, potassium, etc., was taken in the central compartment/feed tank. The membrane arrangement in the electrodialysis cell is shown in Fig. 2. The conductivity of the solution was measured using a conductivity meter (Deep Vision instruments) at specific time intervals. The pH of the solution was measured using Hanna pH meter. Acid/base concentrations of the solution in ACs and BCs were determined by titrimetric method. The chloride ion concentration was measured by argentometric method.

The process parameters such as current efficiency [30], energy consumption [30], transport number of ions [32], WDE, and water dissociation flux [32] were calculated using the following set of equations (2–5). In order to minimize the cell voltage generated in the initial stages of the experiments, dilute HCl (0.01 N) and dilute NaOH (0.01 N) solutions were used in the ACs and BCs, respectively, where acids and bases were produced during the performance study. The feasibility of using PSu-based synthesized membranes in BPMED stack to desalinate seawater was studied by monitoring the ion concentration in all five tanks every 15 min. At the same time, the conductivity value, pH, and TDS were also observed in addition to the amount of current produced, which was measured using a multimeter. The entire experiment was run for 8 h. The BPMED process was then repeated with the commercial PSDVB-based membrane stack in the place of PSu while identically maintaining all the remaining parameters.

$$\text{Current efficiency}\,\eta\,(\%) = \frac{FV\Delta C}{nIt} \times 100 \tag{2}$$

$$\text{Energy consumption}\,(\text{Wh}) = \int_0^t UIdt \tag{3}$$

$$\text{Transport number of ions}\, t_{\pm}^{m} = \frac{nF(C^0V^0 - C^iV^i)}{It} \tag{4}$$

$$\text{WDE}\; r_{\text{diss}}^{\text{BPM}} = 1 - \left(t_{\text{Na}^+}^{\text{BPM}} + t_{\text{Cl}^-}^{\text{BPM}}\right) \tag{5}$$

where η is the current efficiency (%); F is the Faraday constant (96,500 C/mol); V is the total volume (cm^3); ΔC is the change in the concentration of acid or base (mol/L); n is the stoichiometric number ($n = 1$ in this case); I is the applied current (A); t is the operation time (s or h); U is the potential (V); $t_{\pm}^{m}$ is the transport number of anions; C^0 and C^i are the initial and final ion concentration in the anodic/cathodic compartment while V^0 and V^i are the initial and final volume in the anodic/cathodic compartment; $r_{\text{diss}}^{\text{BPM}}$ is the WDE; $t_{\text{Na}^+}^{\text{BPM}}$ is the transport number of sodium ion; and $t_{\text{Cl}^-}^{\text{BPM}}$ is the transport number of chloride ion.

3. Results and discussions

3.1. Characterization of BPM

Confirmation of SPSu-PVA-QPSu membrane possessing BPM characteristics was done by analyzing the acid and base yield in the neighboring compartments as evidenced from the change in pH value and TDS measurements (Fig. 3). From the graph it is obvious that with increments in the performance time, the pH of the solutions in the two compartments changed. The compartment closer to the anode side was found to be basic in nature and increased with time. Similarly, the other compartment, closer to the cathode

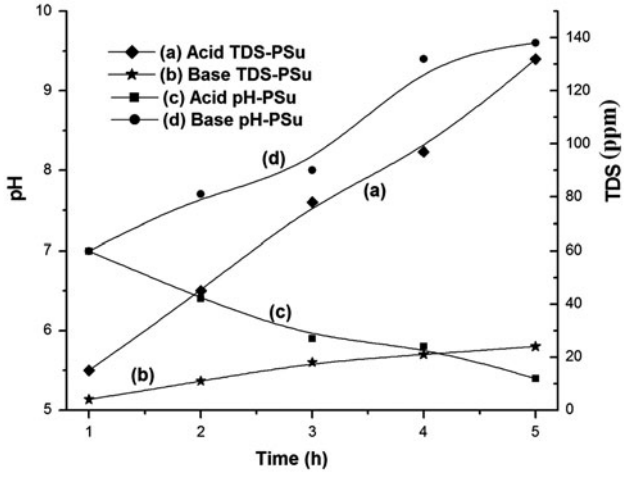


Fig. 3. Variation of pH and TDS measurements with time in turn confirms the BPMPSu efficiency in splitting water into acid and base.

side, was found to be acidic in nature. From the TDS measurement graph, it is clear that both the compartments showed an increase in TDS values which suggested that some ions were introduced during the performance in the neutral compartment. These ions were probably protons and hydroxyl ions which formed on either side during the action of the BPM in splitting water into its ions by making use of an electrical driving force that was applied between the electrodes.

Upon comparing the TDS values of both ACs and BCs, it was clear that the former showed a higher value than the latter which was also proved through conductivity studies. In the case of AC, the conductivity was observed to increase from the initial 0.03 to 0.08 mS/cm, while on the other hand in the BC it was observed finally as 0.05 mS/cm. The minor difference between the AC and BC as observed through TDS and conductivity values suggested that a slight leakage of ions could have occurred through CEL of the BPM than through AEL.

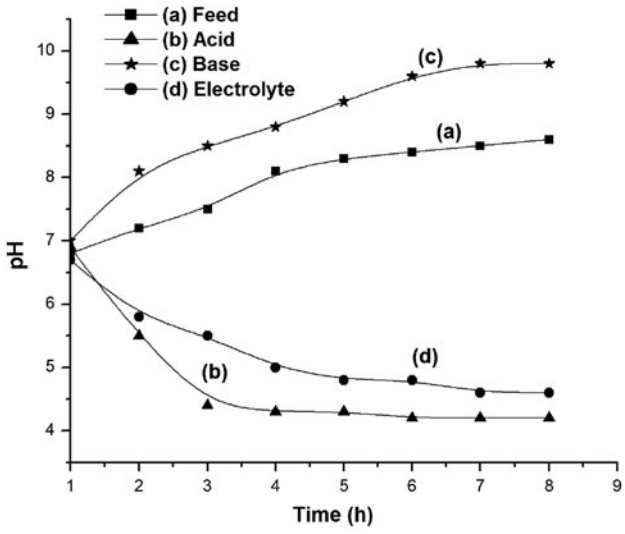


Fig. 4. Change in pH value observed in all the compartments with time during BPMED process.

3.2. BPMED stack performance

The efficiency of BPMED is strongly affected by the transport properties of ions and solute through IEMs. As an initial step, the pH of the solution in all the five compartments was observed at regular intervals of time (15 min) and a graph was plotted as shown in Fig. 4. It was observed that the pattern of change in pH with time was similar in both the cases. From the graph it was further clear that the acid and electrolyte compartments (ECs) remained acidic in nature, whereas the base and feed compartments (FCs) remained basic. The reason for the observed change in pH from neutrality in the FC was mainly attributed to the migration of ions from the BC to the FC, which thus suggested that CEM was more susceptible to leakage of ions than AEM. In the case of EC, since only negligible leakage of OH^- ions was possible through CEL due to the presence of an intermediate layer, the observation of greater ion leakage suggested that the H^+ ion concentration was greater than the OH^- ion in EC. These results, however, do not imply that the H^+ ions did not migrate through AEM to the other compartments, but comparatively the leakage of these ions was lesser.

The change in pH was confirmed by the conductivity measurements in AC, BC, and FC as shown in Fig. 5. Since, the ionic mobility of protons is higher than the hydroxyl ions, correspondingly the conductivity value was found to be higher in the case of AC when compared with the BC. Since 0.01 N of acid and base was added initially to the distilled

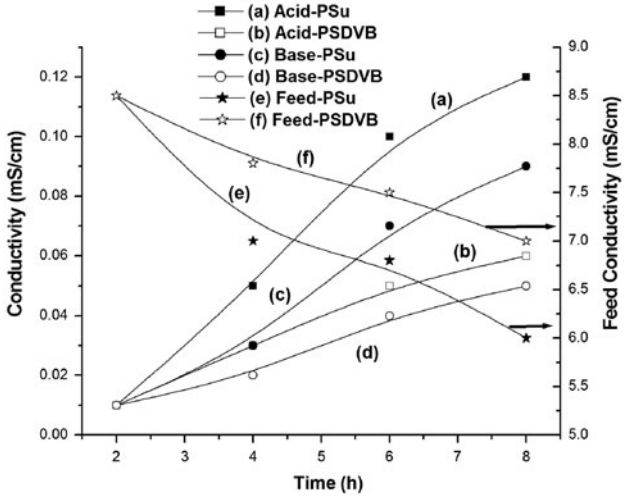


Fig. 5. Conductivity measurements observed in acid, base, and FCs for PSu and PSDVB-based membrane systems in BPMED process.

water in the respective compartments, the solutions initially had some conductivity values other than the conductivity values of distilled water. In the case of AC and BC for both membrane types (PSDVB and PSu), the conductivity was found to increase with time which suggested the presence of acid and base in the respective compartments. It was further observed that the amount of acid increase was greater than the amount of base increase at corresponding time intervals. The reason for this has been already discussed

above. On comparing the two membrane types, PSu-based membrane showed greater acid and base conductivity than that obtained using PSDVB-based membrane system. In the case of FC, the conductivity was lower for PSu-based membrane than PSDVB-based membrane, indicating that removal of ions from the central compartment was greater in the former than in the latter. This clearly confirmed that both the synthesized and commercial membranes displayed a capacity to migrate certain number of ions from the central compartment to opposite electrodes under an electric field, as was evident from their current efficiency values.

Fig. 6 shows the acid/base concentration and the transport number of ions for both the membrane systems. Due to the large electric field appearing at the membrane interface, an excess of OH^- and H^+ ions were produced due to the field-enhanced chemical reaction. These water ions permeate the corresponding exchange layers of the BPM and enter into the adjacent solutions resulting in the formation of acid and base. From the pH and conductivity studies, it was evident that acids and bases of certain concentrations were produced in the ACs and BCs, respectively, of the present EDBPM stack using two types of systems (PSu and PSDVB). From the graph it was further observed that in both the cases (i.e.) acid and alkali yields from the PSu-based system showed a higher concentration than the PSDVB-based commercial membrane. Among the two concentrations, the increase in alkalinity was found to be greater than the increase in acidity in both the cases. This observation was found to be in direct contrast to the higher levels of acid production observed from Figs. 3 and 5 corresponding to the TDS and conductivity studies. The lower acid yield observed here can be explained as occurring due to the small leakage of OH^- ions through CEL of the BPM into the AC. However, this leakage was minimized by the intermediate layer and electrodes placed beside the BPM. In the case of BC, although the leakage of OH^- ions through CEM occurred, leading to a low basic ion concentration, the migrated OH^- ions would also consequently further decrease the acid concentration. Thus, the alkalinity concentration was higher than the acid concentration. The behavior regarding the transport number of ions for both membrane systems revealed that PSu-based systems displayed better transport of sodium and chloride ions through CEM and AEM, respectively, due to its higher acid/base yield than the PSDVB-based system. Since the current efficiency depends upon the ion transport and ionic mobility, it was expected that the PSu-based system would show higher efficiency than the commercial one.

Fig. 7 reveals the water dissociation flux for protons and hydroxyl ions for both membrane systems. On comparing them, the PSu-based membrane system was found to display higher flux than commercial PSDVB-based systems. At the same, in both the systems, flux due to BC was found to be better than acid flux with time. This observation was attributed to the higher base yield concentration obtained than the acid yield during the performance.

Fig. 8 shows the current efficiency and energy consumption for the designed systems. These parameters are generally used to determine the feasibility of any electrochemical process for higher process efficiency.

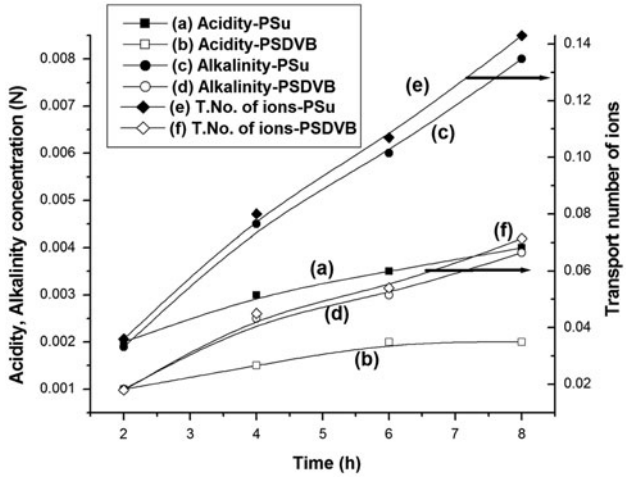


Fig. 6. Effect of acid/alkali yield concentration on transport number of ions for two BPMED systems with time.

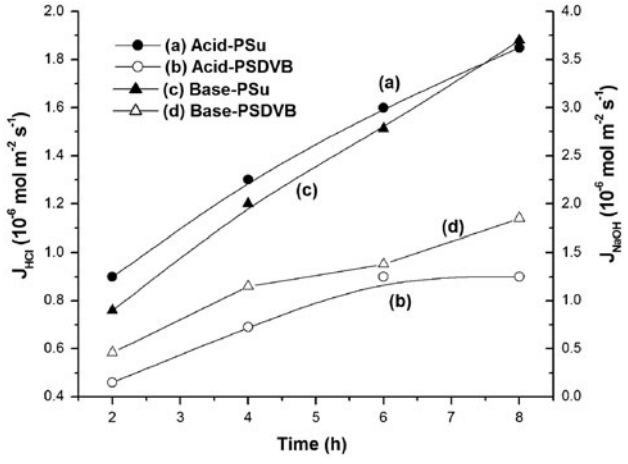


Fig. 7. Variation of water dissociation fluxes in both ACs and BCs with respect to time parameter.

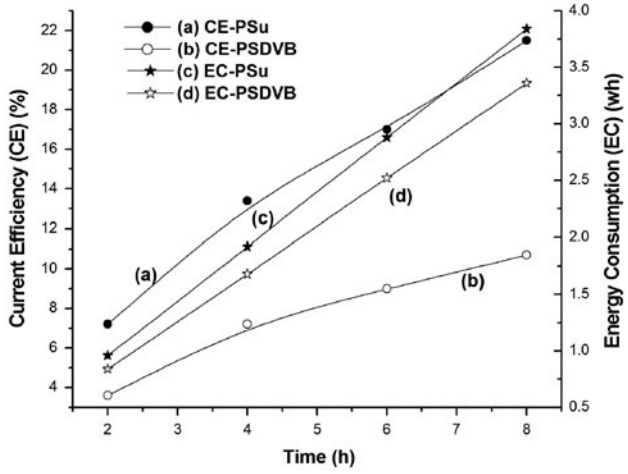


Fig. 8. Variation of current efficiency and energy consumption for PSu- and PSDVB-based systems with different time intervals observed during 8 h BPMED performance.

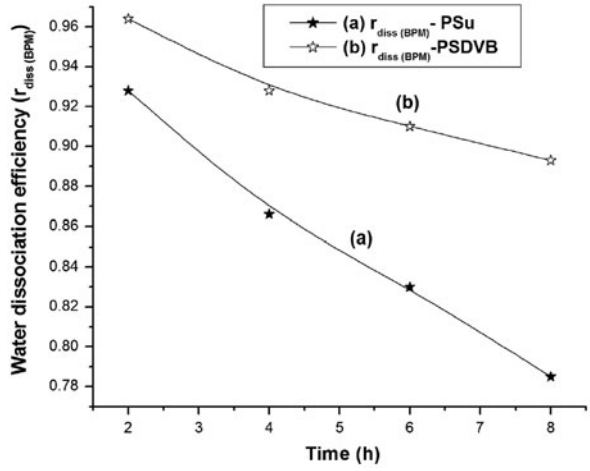


Fig. 9. The relation between WDE of the PSu- and PSDVB-based BPM membranes and operation time during 8 h performance.

The graph shows that the current efficiency of the PSu-based system was much higher than the commercial PSDVB-based systems. On the other hand, analysis of energy consumption showed that the PSu-based system showed a little higher value than the commercial PSDVB-based system. The increase in the energy consumption can be mainly attributed to the fact that a large part of the total electrical energy is consumed to overcome the electrical resistance. Though this may be perceived as a drawback, it needs to be noted that it is possible to overcome the higher energy consumption through higher current efficiency, higher transport number, higher acid/base yield, and increased removal of ions from the FC.

The water splitting capacity/efficiency is one of the most important parameters which determine the process efficiency. The WDE is mainly associated with the BPM and in turn the whole BPMED stack. The pH variations in the ACs and BCs (Fig. 3) confirmed that water dissociation occurred at BPM interface. The result of the WDE test for both the membrane systems is shown in Fig. 9. From the graph it was observed that the PSDVB-based system displayed a little higher efficiency than the PSu-based systems. The lower WDE for the PSu-based system was attributed to its higher ion transport number which is evident from Fig. 5. From the results, it is also inferred that with increase in time, the WDE decreased for both the systems (PSu and PSDVB). This was mainly because, at the initial stages of the experiment, dissociation of water molecules was greatly enhanced due to the second Wien effect. However, once the higher concentration of H^+/OH^- had been reached for that particular feed concentration, the rate of H^+/OH^- production by BPMED process remained constant for some time after which it started to decrease with further increase in process time. This suggested that the mass transfer of Na^+ and Cl^- ions through the IEMs diminished due to the decrease in NaCl concentration in the feed solution. Together with the increase in the concentrations of Na^+ and Cl^- ions in the BCs and ACs, the molecular back diffusion through IEMs caused by the high concentration gradient may hinder the transport of Na^+ and Cl^- ions. Since the WDE mainly depends on the transport number of these ions it was observed to

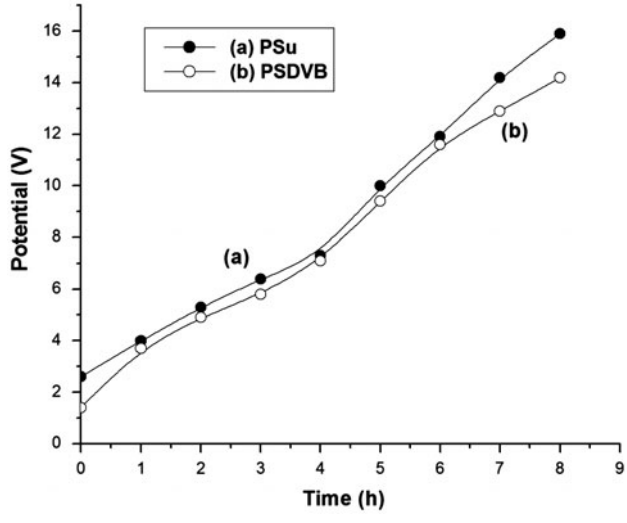


Fig. 10. BPMED membrane stack potential for synthesized PSu and commercial PSDVB-based membrane systems during 8 h time interval.

decrease with increase in time process. Fig. 10 represents the potential variation with time from which it was obvious that the potential utilized by the PSu-based system was a little higher than that of the PSDVB-based system. This indirectly results in higher energy consumption by the former than by the latter system during the later period of performance.

4. Conclusion

In this study, the feasibility of using PSu-based BPMED for desalination of seawater was reported. The BPMED process was able to remove ions from the salt solution with a higher efficiency of about 21% in the case of PSu-based system as against 10% in the case of commercial PSDVB-based system. In addition, during this procedure, no secondary waste was produced and the HCl (0.004 N for PSu system and 0.002 N for PSDVB system) and NaOH (0.008 N for PSu system and 0.004 N for PSDVB system) produced during the performance could be reused for pH control during the synthesis. In conclusion, the BPMED process provided an efficient and environment-friendly purification method by achieving a "one-step" removal of impurity ions.

Acknowledgments

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