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Transport and conductivity properties of an advanced cation exchange membrane in concentrated sodium hydroxide

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An electrochemical concentrator for application to the chlorine-caustic industry is currently under development. In it 30 to 35 wt % NaOH enters the anolyte and catholyte chambers and exits at 20 and 50 wt %, respectively. Consequently, in support of the electrochemical concentrator development, the conductance and transport properties of advanced cation exchange membranes in concentrated sodium hydroxide, are being investigated. The membrane voltage drop, sodium ion transport and water flux of these membranes in 20 to 35 wt % sodium hydroxide anolyte and 30 to 50 wt % sodium hydroxide catholyte at 75° C are presented. To better understand the behaviour of these membranes, electrolyte sorption measurements were conducted in the anolyte/catholyte environment appropriate for the electrochemical concentrator. The water uptake data appear to correlate well with the conductance data and the combined NaOH and water sorption data are consistent with the sodium ion transport data.

1. Introduction

An electrochemical concentrator coupled with a membrane chlor-alkali electrolysis cell system is currently under development [1, 2, 3] to reduce the energy requirements for the production of chlorine and concentrated caustic soda (50 wt % NaOH). Currently, the concentration of the sodium hydroxide produced in an advanced membrane electrolysis cell is typically 30 to 33 wt %. The sodium hydroxide is concentrated to the 50 wt % commercial product by evaporation using the by-product hydrogen and additional fuel. The electrochemical concentrator eliminates the need for the caustic soda evaporation step and reduces the energy requirement of the electrolysis process. The electrochemical concentrator is an alkaline fuel cell containing anode and cathode compartments due to the presence of a cation selective membrane (Fig. 1). By-product hydrogen from the brine electrolysis process and oxygen from the air are consumed in the electrochemical concentrator, generating DC power and concentration sodium hydroxide from 30 to 50 wt % in the cathode compartment while diluting sodium hydroxide in the anode compartment.

In support of the development of our electrochemical concentrator, state-of-the-art and advanced cation exchange membranes in concentrated sodium hydroxide (i.e., 20 to 30 wt % anolyte and 30 to 50 wt % catholyte, respectively) are being studied. The membrane properties of interest are: (1) conductivity, (2) cation selectivity (i.e., the fraction of ionic charge carried by cations), and (3) water transport. The membrane conductivity determines the voltage drop across the membrane which limits the electrical power output of the electrochemical concentrator. It also limits the operating current density of the electrochemical concentrator which, in turn, determines the capacity of the concentrator. Additionally, the Na^+ transport and OH^- rejection properties of the membrane determine the current efficiency of the electrochemical concentrator. Finally, the water transport together with the Na^+ transport properties of the membrane determine the separation efficiency of the electrochemical concentrator.

Advanced cation selective membranes typically consist of a perfluorinated carbon backbone structure for chemical stability with cation exchange groups attached. The cation exchange groups are typically carboxylic, sulphonic, or a combination of both. Perfluorinated carboxylate membranes exhibit excellent permselectivity but poor conductivity. Perfluorinated sulphonate membranes exhibit good conductivity but poor permselectivity. Consequently, advanced bimembranes consist of a thin carboxylate layer for permselectivity and a thicker sulphonate layer for physical support and minimal decreased conductivity. Data on the electrochemical properties of these membranes is limited and a detailed understanding on the nature of the properties of carboxylate and sulphonate membranes is lacking. In general, the conductance properties are correlated to the electrolyte sorption properties of these membranes. Consequently, the electrolyte sorption behaviour of candidate membranes in the concentrated caustic environment of interest for application in the electrochemical concentrator were determined. The electrolyte sorption data are used to explain the electrochemical behaviour of these membranes.

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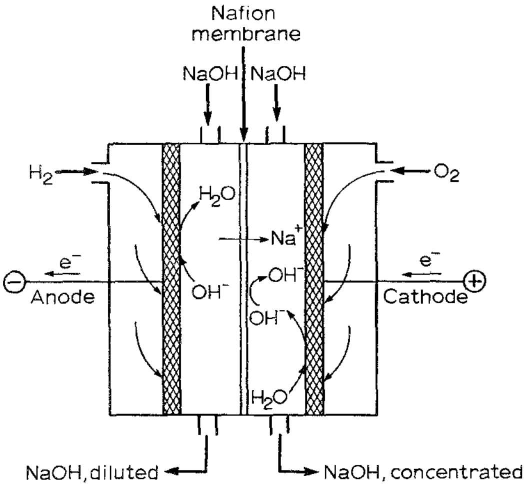

Fig. 1. Schematic illustration of the processes occurring in the electrochemical concentrator. Anode reaction: $H_2 + 2OH^- \rightarrow 2H_2O + 2e^-$; cathode reaction: $2e^- + H_2O + \frac{1}{2}O_2 \rightarrow 2OH^-$.

2. Experimental details

For initial evaluation in concentrated caustic solutions and to get a baseline for the measurement technique and procedure, a commercially available cation exchange membrane labeled NAFION* 901 from E.I. DuPont deNemours and Company was obtained. NAFION 901 is a state-of-the-art perfluorinated cation exchange membrane developed for use in chlor-alkali cells. It is a reinforced bimembrane composed of carboxylate and sulphonate NAFION copolymer. Since the environment of the electrochemical concentrator is considerably different than that encountered in chlor-alkali cells, experimental membranes being developed by DuPont were also evaluated. They include: (1) *membrane designated G1.* This is a thin version of NAFION 117, a commercially available perfluorinated sulphonate membrane used in the H_2O and HCl electrolysis cells; (2) *membrane designated A1.* This is a reinforced bimembrane consisting of carboxylate and sulphonate perfluorinated copolymer; and (3) *membrane designated E1.* This is an unreinforced bimembrane consisting of carboxylate and sulphonate perfluorinated copolymer. The membrane conductivities were measured in a two compartment cell modeled after a design reported in the literature [4]. The cell was constructed entirely of TEFLON† fluorocarbon polymer and the membrane was postioned between the two electrolyte compartments with provisions for electrolyte stirring. The exposed area of the membrane was $9.75\,\mathrm{cm}^2$. Cell heating and temperature control were accomplished by TEFLON encapsulated immersion heaters and RTD temperature controllers. Water cooled condensors were positioned above the gas vents to minimize water loss by evaporation. A constant current was applied across the membrane via platinum screen electrodes using a PAR 371 Potentiostat/Galvanostat. The potential drop was measured as a function of current density across the membrane using two Luggin capillary probes on each side of the membrane. These probes provided solution contact with the dynamic hydrogen reference electrodes (DHE) [5].

In order to determine the electrolyte resistance contribution to the measured voltage drop, the cell constant (cm^{-1}) of our conductivity cell was measured without a membrane and using a $250\,\mathrm{g\,dm}^{-3}$ sodium chloride solution of known specific conductivity [6]. From the voltage-current data, the electrolyte resistance was determined (all current-voltage data were linear with > 0.997 correlation). The resistance data were combined with reported specific conductivities and exposed area ($9.75\,\mathrm{cm}^2$) to calculate the cell constant, $0.09475 \pm 0.0014\,\mathrm{cm}^{-1}$. The cell constant and current-voltage data were used to calculate the specific conductivities for sodium hydroxide solutions (20 to 50 wt %) at a temperature of 75° C. The current-voltage characteristics for various membranes and various anolyte and catholyte concentrations were then measured. From the cell constant, electrolyte conductivities, and symmetry of our cell, the voltage drop, E_e, due to electrolyte resistance for equal and different anolyte-catholyte conditions were determined. The specific conductivity of the membrane, κ ($\Omega^{-1}\,\mathrm{cm}^{-1}$), was then calculated from:

$$\kappa = \frac{id}{(E_{m+e} - E_e)} \tag{1}$$

where i is the current density ($\mathrm{A\,cm}^{-2}$), d is the membrane thickness (cm), E_{m+e} is the voltage drop of the membrane plus electrolyte (V), and E_e is the voltage drop due to the electrolyte (V).

If the membrane exhibits pure ohmic behaviour, then the specific conductivity of the membrane is a constant and is independent of the current density. However, if the membrane exhibits non-ohmic behaviour, then the membrane voltage drop becomes a non-linear function of the current density. In this case the specific conductivity of the membrane, as calculated from Equation 1, is dependent on the current density and caution should be exercised in using the appropriate current density to compute the κ value from the membrane voltage drop.

The sodium ion transport and water transport properties for the various membranes were measured in a similar cell withou reference electrode compartments or Luggin capillary probes. The anode and cathode compartments separated by the test membrane were filled with caustic solutions of known weight and concentrations. A constant current was applied across the membrane for a recorded amount of time. Then the anolyte and catholyte solutions were drained from the cell, and their weights and concentrations determined. In order to obtain reproducible transport data, the cell was filled and then drained, and the anolyte and catholyte additions to the cell were accurately measured. Thus, the minimal electrolyte film retained by the cell during draining did not affect the measurements. From the electrolyte weight change, concen-

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* NAFION is a registered trademark of the E.I. DuPont deNemours & Company.

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tration change, and the amount of charge passed through the membrane, the sodium ion and water transport numbers were determined [4]. For example, the sodium ion transport number, $t_{\mathrm{Na^+}}$ was determined from the expression

$$t_{\mathrm{Na^+}} = \frac{(\Delta\mathrm{wt\ NaOH})F}{(\mathrm{MW\ NaOH})It} \qquad (2)$$

where Δwt NaOH is the weight change (g) of sodium hydroxide in the cathode (or anode) compartment before and after passage of current, MW NaOH is the molecular weight of NaOH ($40\,\mathrm{g\,mol^{-1}}$), I is the current (A), F is Faraday's constant ($96485\,\mathrm{C\,mol^{-1}}$), and t is the time of current passage (s). Note that some researchers have given nominal units of $\mathrm{mol\,F^{-1}}$ to the transport number, where F is interpreted as one mole electrons. However, this combination of units is dimensionless [7]. Ideally, for a 100 percent cation selective membrane $t_{\mathrm{Na^+}}$ should be unity. In practice, the 'leakage' of hydroxyl species from the cathode to anode compartment contributes to the ionic current so that

$$t_{\mathrm{Na^+}} + t_{\mathrm{OH^-}} = 1 \qquad (3)$$

Consequently, the sodium transport number is a measure of the current efficiency of the membrane and in a chlor-alkali environment is typically in excess of 0.9 for advanced membranes [6].

The water flux, $T_{\mathrm{H_2O}}$, is defined as the ratio of the water transport number to the sodium ion transport number, thus:

$$T_{\mathrm{H_2O}} = \left[\frac{(\Delta\mathrm{wt\ H_2O})F}{(\mathrm{MW\ H_2O})It} + 1\right] \Big/ t_{\mathrm{Na^+}}$$

where Δwt H_2O is the weight change (g) of water in the cathode compartment before and after the passage of current, MW H_2O is the molecular weight of H_2O, and the rest of the symbols are as defined previously. The second term in parentheses corrects for the water consumption at the platinum cathode (one mol of H_2O consumed per mol of charge). The water transport number may also be determined by analysing Δwt H_2O in the anode compartment. In this case, correction for the water produced by the anode reaction is necessary (one-half mole H_2O is produced per mol of charge).

Conductance and transport measurements were made in environments where the anolyte NaOH concentration ranged from 20 to 40 wt % and the catholyte NaOH concentration ranged from 30 to 50 wt %. The electrolyte temperature was controlled at 75 ± 1° C. The exposed membrane area was $9.75\,\mathrm{cm^2}$ and all the current densities reported in this work are based on this area.

Electrolyte sorption studies were conducted in 25 to 50 wt % NaOH at room temperature using an approach described previously [8]. The membrane samples were expanded in boiling 2% caustic soda, and then allowed to equilibrate to constant weight gain in the appropriate electrolyte. Since the membrane experiences large concentration gradients in the electrochemical concentrator, electrolyte sorption experiments were also conducted in a specially designed sorption cell with electrolytes of different concentrations on either side of the membrane. As encountered in the electrochemical concentrator, the carboxylate layer was always facing the higher concentration catholyte. For the higher concentrations, equilibration required approximately two weeks. The samples were blotted dry to remove free electrolyte and weighed. They were then dried at 110° C to remove the water and weighed. This process was repeated to ensure a constant weight, indicating complete water removal. Finally, the membranes were repeatedly soaked in water to remove the NaOH, oven dried and weighed. This process was repeated to ensure a constant weight, indicating complete removal of the sodium hydroxide. From these weights, the water uptake and the sodium hydroxide uptake of these membranes were determined as a function of their electrolyte environment.

3. Results and discussion

In order to define the anolyte/catholyte conditions appropriate for our electrochemical concentrator, both co-flow and counter-flow modes of operation of the membrane alkaline fuel cell concentrator were considered. In the co-flow scheme, the 30 wt % caustic soda enters at one end of the concentrator cell, and the 20 and 50 wt % caustic solutions exit at the other end. In the case of the counter flow scheme, the caustic streams enter and exit from opposite ends of the concentrator. In order to determine the effect of the co-flow versus the counter-flow on membrane performance, a membrane test matrix covering 30 to 50 wt % of NaOH catholyte and 20 to 40 wt % NaOH anolyte was designed to measure the electrochemical properties of the membrane.

3.1. *Sorption studies*

The water sorption behaviour of cation selective membranes is reportedly inversely related to the NaOH concentration. The empirical expressions for sulphonate and carboxylate membranes are [9, 10],

$$W_{\mathrm{e}} = \left(\frac{0.00995}{1 + 0.075M}\right)\exp(3980/EW) \qquad (5)$$

and

$$W_{\mathrm{e}} = \left(\frac{0.001855}{1 + 0.1065M}\right)\exp(5104/EW)$$

respectively. W_{e} is the weight percent water uptake, M is the electrolyte molarity, and EW is the equivalent weight of the polymer. Consequently, plots of the reciprocal water uptake against electrolyte concentration should be linear. In Fig. 2 we present such a plot for the four cation selective membranes studied here along with the behaviour predicted by Equations 5 and 6. The water sorption behaviour of NAFION 901 and membrane E1 (both membranes contain carboxylate and sulphonate functionalities) is close

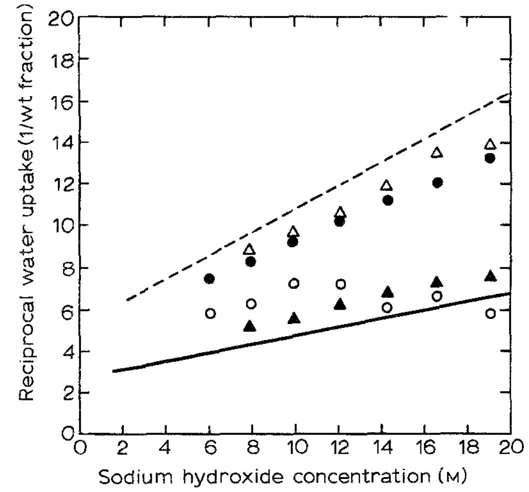

Fig. 2. Water sorption behaviour of various cation selective membranes in concentrated sodium hydroxide environment. (———) Sulphonate equation, (– – –) carboxylate equation, (△) NAFION 901, (▲) membrane A1, (○) membrane G1, (●) membrane E1.

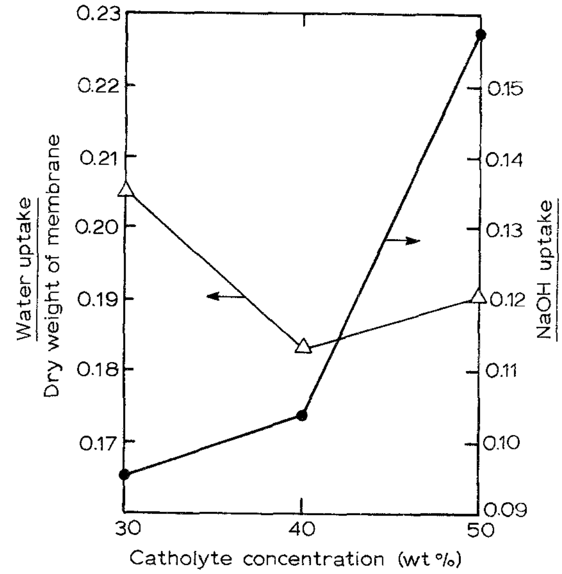

Fig. 4. Water and NaOH sorption data for membrane A1 as a function of catholyte concentration. Anolyte concentration = 20 wt %.

to that of the carboxylate membrane described by Equation 6. The observation that NAFION 901 and other bimembranes exhibit carboxylate water sorption behaviour was previously reported elsewhere [8]. The sorption behaviour of membrane G1 (the very thin sulphonate membrane) is close to that of the sulphonate membrane described by Equation 5. In contrast to other bimembranes, membrane A1 exhibits sulphonate water sorption behaviour. This is particularly surprising since this membrane exhibits excellent sodium ion selectivity in concentrated sodium hydroxide (a property usually attributed to carboxylate membranes).

In view of the unique sorption characteristics of the bilayer membrane A1, sorption experiments were conducted with the anolyte/catholyte sides of this membrane exposed to different concentrations of NaOH. Two different cases were investigated. In case I, the catholyte concentration was fixed at 50 wt % and the anolyte concentrations were varied from 20 to 40 wt %. In case II, the anolyte concentration was fixed at 20 wt % and the catholyte concentration was varied from 30 to 50 wt %. Figure 3 shows the sorption data for case I. The water uptake and the NaOH uptake decreased with increasing anolyte concentration or with decreasing concentration gradient across the membrane. Figure 4 shows the sorption data obtained for case II. The water uptake decreased initially with increasing catholyte concentrations to 40 wt % and then increased as the catholyte concentration increased to 50 wt %. However, the NaOH uptake data was found to increase with increasing catholyte concentration or with increasing concentration gradient. In summary, the water uptake is sensitive not only to the catholyte/anolyte concentration gradient but also to the anolyte concentrations. The NaOH uptake increases with increasing concentration gradients.

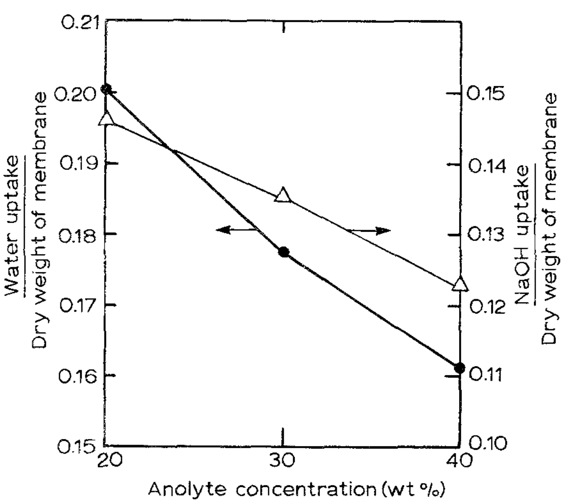

Fig. 3. Water and NaOH sorption data for membrane A1 as function of anolyte concentration. Catholyte concentration = 50 wt %.

3.2. *Conductivity measurements*

In Table 1 the specific conductivity is presented for the membranes NAFION 901, A1, E1 and G1 at 0.1 A cm^{-2} in 30 wt % NaOH and at 75° C. For NAFION 901, the specific conductivity is $4 \times 10^{-3}\,\Omega^{-1}\,\text{cm}^{-1}$, which agrees well with data reported in the literature [8] for this membrane in 30 wt % NaOH at 75° C. Membrane G1 exhibited the highest specific conductivity. This result is also in agreement with previous observations that sulphonate cation exchange membranes exhibit better conductivities than carboxylate membranes [11]. The conductivity of membrane E1 was close to NAFION 901 whereas membrane A1 was about two

Table 1. Specific conductivity in 30 wt % NaOH at 75° C at 100 mA cm^{-2}

Membrane	*Specific conductivity* (Ω^{-1} cm^{-1}) × 10^3
901	4.0
A1	8.2
E1	5.0
C1	14.0

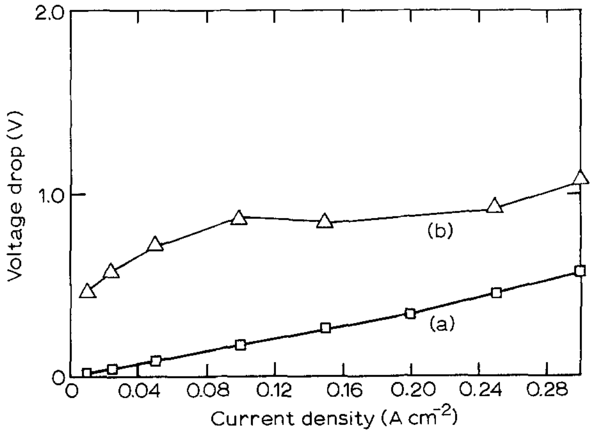

Fig. 5. Membrane A1 voltage drop as a function of current density at 75° C, and (a) 20 wt % anolyte/30 wt % catholyte, (b) 30 wt % anolyte/50 wt % catholyte concentration.

times more conductive than NAFION 901. The conductivity of these membranes increases in the order: NAFION 901 → E1 → A1 → G1. These data are rationalized by the water sorption data in Fig. 2. The water uptake increases in the same order NAFION 901 → E1 → A1 → G1. These results agree withthe previous observations; the higher the water sorption characteristic of the membrane, the higher the conductivity of the membrane.

Since membrane A1 exhibited the most promising combination of conductivity and selectivity [3], its performance under the broader range of electrolyte conditions appropriate to both counter-flow and co-flow operation of the electrochemical concentrator was measured. Figure 5 shows the voltage drop data for membrane A1 as a function of the applied current density for two different NaOH concentrations and gradients. The data in Fig. 5a were obtained with 20 wt % anolyte and 30 wt % catholyte whereas those in Fig. 5b were obtained with 30 wt % anolyte and 50 wt % catholyte. Two distinct features of these curves are noteworthy. At low NaOH concentrations (Fig. 5a), the membrane exhibits ohmic behaviour as indicated from the linear relationship between the membrane voltage drop and the membrane current density. However, at higher concentrations of NaOH and with higher gradients (Fig. 5b), the membrane exhibited non-ohmic behaviour as indicated from the non-linear relationship between the membrane voltage drop data and the current density. The membrane shifts from low ohmic conductivity at low current densities ($< 0.05\,\mathrm{A\,cm^{-2}}$) to high ohmic conductivity at higher current densities ($> 0.1\,\mathrm{A\,cm^{-2}}$). This behaviour was observed in all cases where the catholyte concentration was greater than 35 wt % NaOH. This phenomenon is particularly beneficial for application in the high catholyte concentration of the electrochemical concentrator, since it may enable the concentrator to operate at higher current densities.

Figure 6 shows the voltage drop data for membrane A1 at $100\,\mathrm{mA\,cm^{-2}}$ over a range of anolyte concentrations from 20 to 40 wt % and catholyte concentrations from 30 to 50 wt % NaOH. The data are plotted as a function of anolyte concentration for various fixed catholyte concentrations. At any fixed catholyte concentration, the membrane voltage drop decreases with decreasing anolyte concentration (or with increasing catholyte/anolyte concentration gradient). This effect is more pronounced at higher catholyte concentrations. For example, consider the data with 50 wt % catholyte. The membrane voltage drop is about 1.8 V with 35 wt % anolyte whereas with 20 wt % anolyte the membrane voltage is only about 0.2 V. In other words the conductivity of the membrane decreased by almost an order of magnitude when the anolyte concentration was reduced by about one half. This behaviour favours co-flow operation in the electrochemical concentrator.

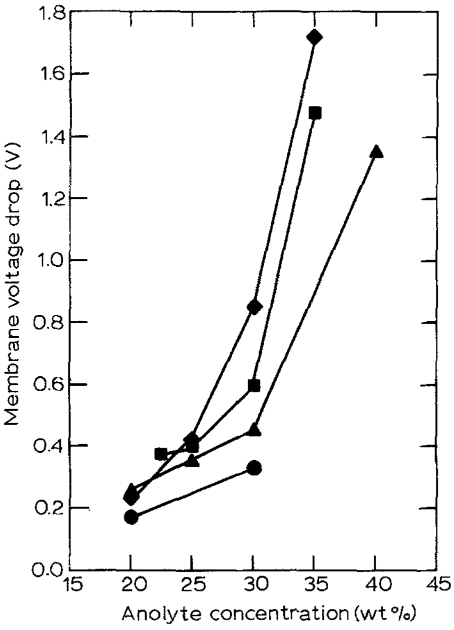

Fig. 6. Voltage drop for membrane A1 at $0.1\,\mathrm{A\,cm^{-2}}$ as a function of anolyte concentration for various catholyte concentrations: (●) 30, (▲) 40, (■) 45 and (◆) 50 wt %.

The effect of the anolyte concentration on the conductivity of the membrane is correlated with the water sorption characteristics. At fixed catholyte concentration, the sorption of water increases with decreasing anolyte concentration (see Fig. 3). At a fixed anolyte concentration, the membrane voltage drop increases with increasing catholyte concentration (or with increasing catholyte/anolyte concentration gradient). Similarly, at an anolyte concentration of 20 wt % NaOH, the water sorption decreases as the catholyte concentration increases from 30 wt % to 40 wt % and then increases as the catholyte concentration is further increased to 50 wt %. Correspondingly, the membrane voltage drop initially increases and then decreases slightly as the catholyte concentration is increased from 30 to 50 wt %.

3.3. *Transport measurements*

Figure 7 shows the sodium ion transport data as a function of catholyte concentration ranging from 30 to 50 wt % NaOH and for different anolyte concentrations ranging from 20 to 35 wt %. In general, the

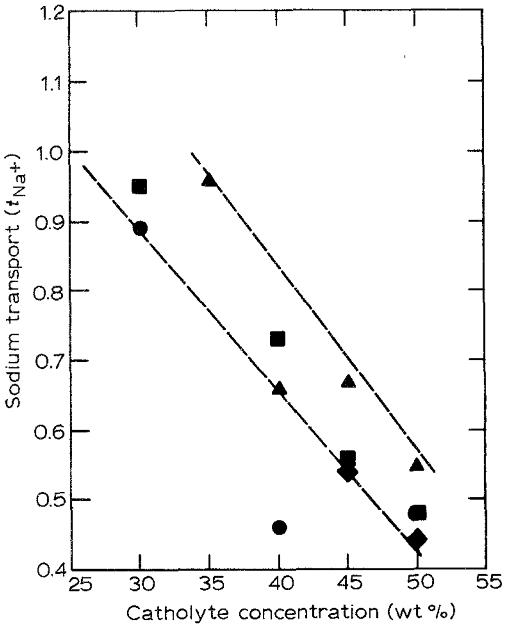

Fig. 7. Sodium transport for membrane A1 as a function of catholyte concentration for various anolyte concentrations: (●) 20%, (▲) 25%, (■) 30% and (◆) 35%.

sodium ion transport number is observed to decrease with increasing catholyte concentration for any given anolyte concentration. Figure 4, shows that the NaOH sorption increases with increasing catholyte concentration for a given anolyte concentration. Earlier workers [4] report that increased NaOH sorption correlates to increasing NaOH concentration. Additionally, Mauritz and coworkers [12] suggest that enhanced hydroxide ion transport (or reduced sodium ion transport) occurs in environments of decreasing water content due to increased polarization of the O–H bond. The water sorption data in Fig. 4 are consistent with the above explanation and support the reduction of sodium ion transport with increasing catholyte concentration at a given anolyte concentration.

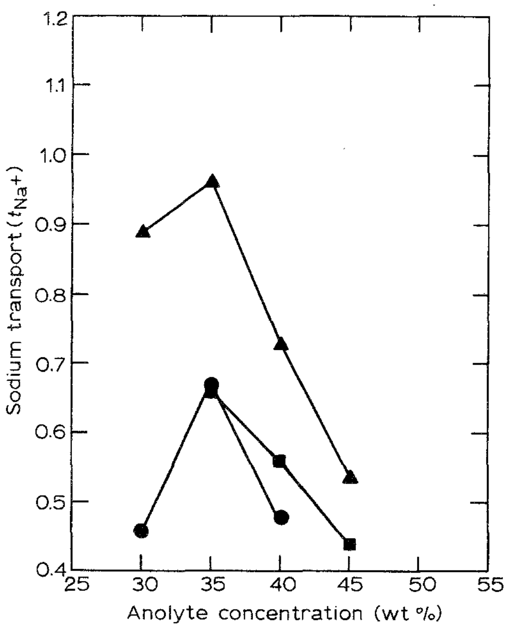

Fig. 8. Sodium transport for membrane A1 as a function of anolyte concentration for various concentration gradients (Δ) across the membrane: (▲) 10, (■) 15 and (●) 20.

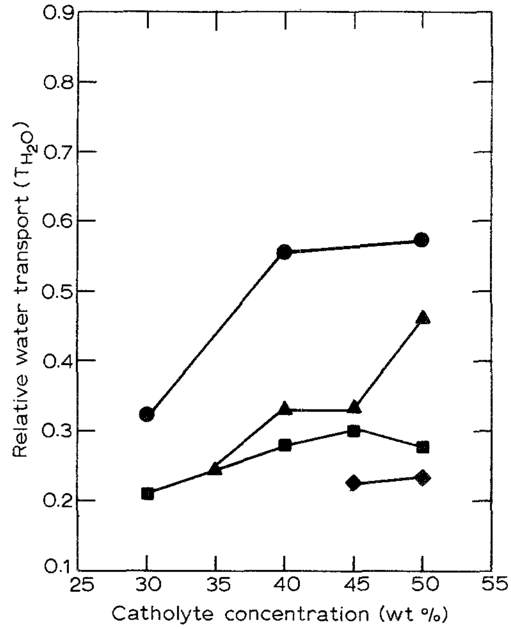

Fig. 9. Relative water transport for membrane A1 against catholyte concentration for various anolyte concentrations: (●) 20%, (▲) 25%, (■) 30% and (◆) 35%.

Figure 8 shows the sodium ion transport data as a function of anolyte concentration for different concentration gradients across the membrane. For a given concentration gradient, there appears to be a maximum in the transport number for sodium ion at an anolyte concentration of 25 wt %. It is also evident that the concentration gradient alone does not control the sodium transport properties. However, for any given anolyte concentration, the sodium transport number increased with decreasing concentration gradient.

A measure of the water flux is given by the relative water transport number, T_{H_2O} (recall that T_{H_2O} is the ratio of t_{H_2O} to t_{Na^+}). In the present experiments, the water flux is determined by electro-osmosis due to the current flux and osmosis due to the concentration gradient. In Fig. 9 T_{H_2O} as a function of catholyte concentrations for different anolyte concentrations is plotted. As the concentration gradient across the membrane increases, the water flux increases. For the 50% catholyte : 30% anolyte case (counter-flow), T_{H_2O} is approximately 2.5. For the 50% catholyte : 30% anolyte case (co-flow), T_{H_2O} is approximately 5.5.

4. Summary and conclusions

The performance of a state-of-the-art chlor-alkali cation selective membrane as well as several developmental membranes in concentrated sodium hydroxide environment has been reported. An advanced developmental membrane designated A1, exhibited the best compromise in terms of conductivity and selectivity. The concentrations of the electrolyte in contact with the anode and the cathode side of the membrane appear to control the conductivity and the transport properties of the membrane.

For any given catholyte concentration, the conductivity of the membrane increased with decreasing anolyte concentrations, the effect being more pronounced at higher catholyte concentrations. Likewise, at fixed catholyte concentration, the water uptake of the membrane increased with decreasing anolyte concentrations. Thus the data in this work suggest, in agreement with previous observations, that the conductivity of the membrane increases with increasing water uptake. For any given anolyte concentration the sodium ion transport number decreased with increasing catholyte concentration. Correspondingly, the NaOH sorption increased and the water sorption decreased with increasing catholyte concentrations, thus, rationalizing the transport behaviour of the membrane. No correlation was observed between the concentration gradient across the membrane and the transport data or the conductivity data.

These membrane performance data are critical to the development of the electrochemical concentrator or any similar membrane separation process. To elucidate this importance, we consider the distinction between a chlor-alkali cell and the electrochemical concentrator. If the chlor-alkali cell, the electrolyte chambers are well mixed due to the vigorous gas evolution at the anode (Cl_2) and cathode (H_2). Consequently, the electrolyte chambers are modeled as continuously stirred tank reactors (CSTR) and the membrane is exposed to a constant anolyte/catholyte environment [13].

However, in the electrochemical concentrator, the anolyte and catholyte streams are not well mixed and behave as plug flow reactors [14]. Consequently, the membrane is exposed to an anolyte/catholyte environment which changes along the length of the membrane. Since the conductivity, water flux and sodium transport are functions of the anolyte/catholyte environment, these membrane characteristics change along the length of the membrane.

These membrane performance data will be used in an electrochemical concentrator model to optimize operating variables with respect to power output and concentrating ability.

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