THE ELECTROOSMOTIC EFFECTS ARISING FROM THE INTERACTION OF THE SELECTIVELY ANION AND SELECTIVELY CATION PERMEABLE PARTS OF MOSAIC MEMBRANES

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ABSTRACT It has been previously shown, theoretically and in model system experiments, that mosaic membranes composed of anion-selective (electropositive) and cation-selective (electronegative) parts interposed between electrolytic solutions of different concentrations give rise to local electrical circuits. In this work with model systems it is shown that these currents produce electroosmosis. In systems with permselective electronegative membranes and KCl solutions, the electroosmotic water transport was 16 moles/faraday. With the permselective electronegative membrane replaced by more porous electronegative membranes, the electroosmotic effects were about twice as high. With Li salts, the water transport was considerably larger. A system with a permselective electropositive membrane of 50 cm^2 effective area and an electronegative membrane of 120 cm^2 gave internally generated currents up to 20 ma. In extrapolating from the results with macromodels to effects with true mosaics, *i.e.* microsystems, it is stressed that current depends on the linear distance over which membranes interact. In true mosaic membranes, the current pathways will be of the same order as the dimensions of individual membrane microelements; the sum of all local microcurrents will be correspondingly larger than the current in the macromodel, and the electroosmotic effects will be proportionately greater. Electroosmotic effects with true charge-mosaic membranes may be of the same order or larger than the liquid transport by normal and anomalous osmosis which might occur across the individual parts of the charge-mosaic.

INTRODUCTION

Many of the involved electrochemical *in vivo* functions of living membranes are attributed to the intricacies of their physichochemical microstructure. Complex membranes, aside from still more involved cases, may be either "layered" structures composed of several layers of different properties or "mosaic" membranes consisting of parts of different properties in juxtaposition. With membranes of porous character, one of the simplest cases is a membrane which is a mosaic composed both of ideally anion-selective and ideally cation-selective parts.

In earlier papers it has been shown that spontaneous electric currents arise when a mosaic membrane composed of permselective, exclusively cation-permeable and exclusively anion-permeable parts is placed between electrolytic solutions of different concentration (1-3). According to the theory of such systems, cations move from the concentrated to the dilute solution across the cation-selective parts of the membrane, and an equivalent number of anions move through its anion-selective parts. This movement of ions constitutes a flow of current.

We have recently reported on the electroosmotic behavior of permselective membranes and similar membranes of higher porosities when an externally generated current is passed through them (4). A series of polystyrene–sulfonic acid–collodion membranes of graded porosities was tested, the least porous being of the permselective type with a water content of 9 per cent by volume, the most porous having a water content of 75 per cent. In the densest membranes the electroosmosis was 3.2 moles of water/faraday with KCl and 6.4 moles of water/faraday with LiCl. As the porosity of the membranes is increased, electroosmosis per faraday increases; with the most porous membranes, of the porosity of dialyzing membranes, the values with KCl were 50 moles of water/faraday and with LiCl, 90 moles of water/faraday.[1] The ratio of the quantities of water transported per faraday in the presence of K^+ and Li^+ was fairly independent of the water content of the membrane, an observation which could be anticipated on theoretical grounds, as was pointed out previously, because of the lower electrolytic mobility of the Li^+ ion (4). (See also below under Experimental.)

In view of these results it becomes of interest to investigate the electroosmotic effect which could be expected to arise in mosaic membrane systems in which internally generated currents flow. The mosaic systems which will be considered here include some with permselective membranes only, as well as some with membranes of higher porosities (and correspondingly lower degrees of ionic selectivity).

THEORETICAL CONSIDERATIONS

For a physicochemical investigation of mosaic membranes from the theoretical and experimental point of view, it is necessary to study macromodel systems. These models must embody all the essential elements of true mosaic membranes composed of microscopic parts, and lend themselves to clear theoretical discussion and to quantitative experimental investigation.

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[1] According to a recent review, the most probable hydration numbers for K^+ and Li^+ are about 1 and 3, respectively (5). Thus it can be concluded that even in the densest membranes used in the above experiments, the greater part of the observed transport of water is due to electroosmosis. With the more porous membranes, virtually the whole transport of water is due to electroosmosis in the classical sense, the contribution of the water of hydration making at most a minor contribution to the total effect.

The basic theory of mosaic membranes is most easily developed by reference to a sequence of line drawings. Fig. 1*a* illustrates schematically a single membrane with adjacent negatively charged exclusively cation-permeable and positively charged exclusively anion-permeable parts, the plus and the minus signs in the figure referring to the charge of the membrane. Fig. 1*b* shows the spatial and electrical separation of the cation-selective and anion-selective parts of the membrane, the dashed arrows indicating the directions of the two membrane potentials arising at the two membranes. Fig. 1*c* shows the spatially separated cation-selective and anion-selective parts joined electrically by the dilute salt solution in an all-electrolytic circuit, the solid arrows indicating the direction of the flow of the (positive) current (1-3).

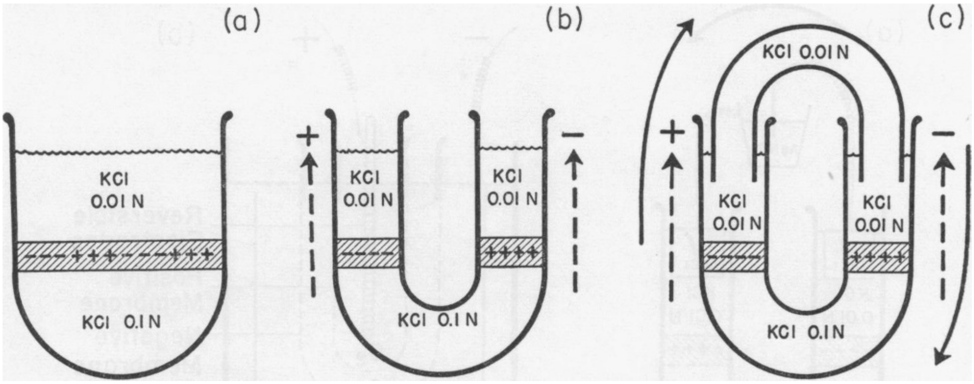

FIGURE 1 Pictorial development of the theory of electrolyte permeability of mosaic membranes which are composed of ideally anion-selective and ideally cation-selective parts. (*a*) Mosaic membrane with adjacent cation-selective (electronegative) and anion-selective (electropositive) parts. (*b*) Spatial and electrical separation of the cation-selective and anion-selective parts of the membranes. (*c*) The spatially separated cation-selective and anion-selective parts joined electrically by a bridge of solution in an all-electrolytic circuit.

In principle, the simplest way to determine the current in the system of Fig. 1*c* is to measure, in a system of suitable geometry, the potential difference across a part of the system which has an accurately measurable and invariant resistance. Models of this type have been constructed with permselective membranes of a very high degree of ionic selectivity. The quantity of electricity which flowed in a given time was found to agree quantitatively with the quantity of electrolyte transferred from the more concentrated to the more dilute solution (3).

Systems of the type shown in Fig. 1*c* are not suitable for the planned study of spontaneous electroosmosis across mosaic membranes. Another electrochemically equally satisfactory approach to the quantitative study of mosaic systems consists of cutting the system of Fig. 1*c* at some suitable point and attaching to the two open ends of the interrupted circuit two symmetrical electrodes that can reversibly take current from and return it to the system. The two electrodes in turn are connected to each other by some conventional current-measuring instrument, as shown in Fig. 2*a*. A schematic drawing of an experimental model is given in Fig. 2*b*. In Fig. 2*b* the two membranes are arranged concentrically so that three compartments are formed. The fairly narrow middle compartment contains the dilute salt solution,[2] and the innermost and outermost compartments contain the concentrated solution, electrical contact being made through 2 Ag|AgCl electrodes. With these systems much greater currents can be obtained because the total resistance of the circuit is greatly reduced by the use of a pair of properly shaped and located auxiliary electrodes. With this experimental arrangement, too, it was shown that the amount of current which flowed during a given time was exactly equivalent to the amount of KCl which was transported from the concentrated to the dilute solution (2). It is obvious that the equivalence of these two quantities is tied to the condition that both membranes are of practically perfect ionic selectivity.

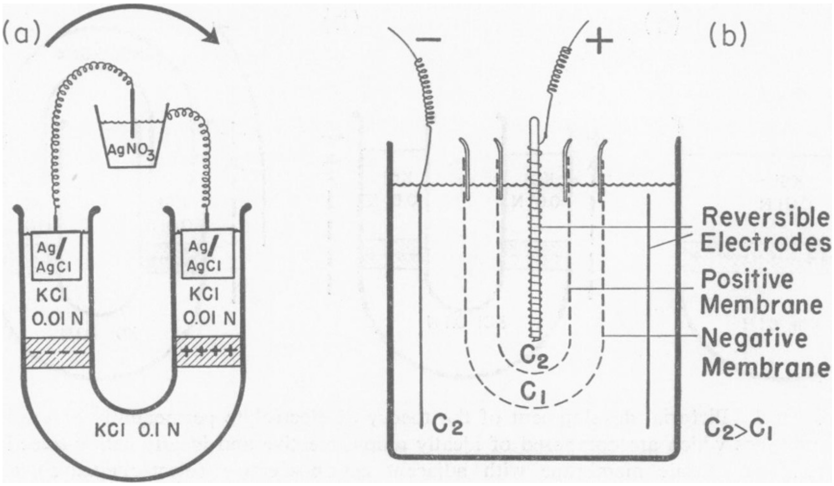

FIGURE 2 Mosaic membrane models with auxiliary electrodes through which the current flows. (*a*) A schematic model. (*b*) The experimental model.

Such a model system with suitable modification was used in the present work to study the electroosmotic effects arising in mosaic membrane model systems. The relationship of the effects observed with such models to the effects which might arise across true mosaic membranes will be discussed later.

Electric currents will also flow in mosaic membrane systems in which one or both membranes are "leaky." In such cases the equivalence of quantity of current

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[2] Placing the dilute solution in the middle compartment makes it possible to determine moderate increases in concentration with the required accuracy.

and quantity of electrolyte moving into the dilute solution cannot exist. Here, electrolyte is transferred into the dilute solution not only by the discussed electrical mosaic membrane mechanism, *i.e.* the flow of a current, but also by the leakage of electrolyte, the simultaneous diffusion of both anions and cations across a membrane or membranes of less than perfect ionic selectivity.

In the present study we are not concerned with the relationship of the quantity of current that flows in a mosaic system and the quantity of electrolyte transferred. We are dealing here with the relationship of the quantity of current that flows in such systems and the quantity of *solvent* that is transported electroosmotically by this current from concentrated to the dilute solution. In other words, we will study the ratio of moles of water per faraday in its dependence on the properties of the membranes, both highly selective and leaky membranes being used.

EXPERIMENTAL

The experimental plan to be pursued must be introduced with some considerations of a general nature.

The *geometry of the system,* that is, the size and shape of its component parts, is inconsequential from the point of view of the theory, provided the essential electrical features of the model are not altered. Likewise, the electrolyte used, the porosity of the membranes, the absolute concentrations of the two electrolyte solutions, and their concentration ratio may be freely selected as required by the experiment. For our purpose it was necessary to adjust these various factors so that maximal electroosmotic effects could be obtained.

One of the most important steps in the direction of decreasing the *electrical resistance of the experimental model* systems consists in the selection of a suitable geometric arrangement. It is helpful to increase the cross-sectional dimensions of all component parts of the system and to make all dimensions in the direction of the flow of the current as small as possible. This is particularly true of the dilute solution.

One may also increase the absolute concentration of the solutions in order to decrease the resistance of the system. The full exploitation of this possibility is limited by the fact that the "leak" of the membranes increases as the absolute concentration of the two solutions is raised (2-4, 6-8).

Another possibility of increasing the current lies in the choice of a high concentration ratio, which would result in an increase in the E.M.F. of the system. The usefulness of this possibility also is limited. Too high a concentration of the concentrated solution increases the leak, and too low a concentration of the dilute solution unduly increases the resistance of the system.

The *resistance of the membranes* can be lowered by the choice of membranes of higher porosity (6-8). However, the usefulness of this approach also is restricted. If membranes of too low a resistance are used, the leakage of non-critical ions is increased too much and the membrane potential is lowered significantly. In addition, the osmotic movement of water increases as the porosity of the membranes is increased and anomalous osmosis may arise. On the other hand, increasing membrane porosity results in a greater electroosmosis per faraday. The selection of the membranes is only one of the several facets of the experimental task of maximizing this quantity. In this context one should recall that the resistances of the membranes, particularly the resistance of the electroosmotically more effective, more highly porous membrane (see below), are only a small fraction of the resistance of the whole circuit in the model system. Consequently the E.M.F. which acts upon the latter membrane is only a small fraction of the total E.M.F. arising in the system; it would be practically impossible to determine it with any useful degree of accuracy. This, however, is not necessary in the context of the present investigation which is concerned with the number of moles of water transported electroosmotically per faraday of current flowing in the model system.[3]

In choosing *the electrolytes to be used,* the following considerations concerning the electrical resistance and the electroosmotic behavior of structures of ion exchange character are pertinent. Though in their simplest form these considerations are valid in a quantitative manner only with structures of not too high porosities in contact with fairly dilute solutions, they are a most useful and at least a semiquantitative guide also with respect to membranes of fairly high porosity and solutions of several tenths' molarity, as those used in our experiments.

The electrical resistances of ion exchange bodies in general, including ion exchange (ionic) membranes, are higher when equilibrated with ions of low electrolytic mobility, such as Li^+, than with ions of higher mobility, such as K^+. In fairly open exchangers, such as the ion exchange resins first studied by Heymann and O'Donnell, the ratio of the resistances in the K^+ and Li^+ states, 1:1.86, is nearly the same as that of the reciprocals of the ionic mobilities of these two ions in free solution, 1:1.95 (9). Similar results were reported for collodion matrix membranes (10). The degree of dissociation of the movable counterions in the membranes is virtually the same with the various species of the univalent counterions. The Li^+ ions, because of their larger hydrated size, have a larger frictional interaction with the surrounding water, whether in free solution or in the pores of a membrane; the electrical resistance of the membrane in the Li^+ state is correspondingly higher.

The larger frictional interaction of the electromigrating Li^+ ions with the water means a larger transference of momentum from the former to the latter, while the hydraulic resistance of the membanes, as far as it is known, is not affected by the nature of the univalent counterions. In membranes, this higher frictional resistance of the Li^+ becomes manifest as a correspondingly greater quantity of water transported electroosmotically *per ion* electromigrating across the membrane (4).[4] For a given permissible percentage degradation of the system, Li^+ will yield a higher total volume of water transported than K^+. Thus, we can conclude that while any common strong uni-univalent electrolyte can be expected to be suitable for our planned experiments, a given membrane will yield higher ratios of moles of water moved per faraday if we use ions of

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[3] The reader will note that the frame of reference in the current investigation is different (moles of water per faraday) from that ordinarily used in most systematic electrokinetic studies in which the applied E.M.F. is the independent variable. While this is the obvious approach in such electrokinetic studies *per se,* it is of interest to recall that the recent very extensive technological literature on electroosmosis describes this effect in terms of moles of water per faraday, for essentially the same reasons which make this approach advisable in the present study (for some references compare reference 4).

[4] With a given potential difference applied to a given membrane in the Li state, it would take about twice as long to transfer one equivalent of Li^+ ions across it as it would take with the membrane in the K state and K^+ ions. The electroosmotic flow of water per unit of time with a given potential difference across a given ionic membrane would be sensibly the same, independent of the nature of the univalent counterions.

low electrolytic mobility such as Li+. For the purpose of comparison with our previous results, we have used the chloride and iodide salts of the alkali metals, Li, Na, and K (4).

The nature of the *electrodes* to be used, in essence, is immaterial provided they are reversible, not polarized too much by the current intensities likely to be encountered, and of low resistance. The two latter factors are influenced favorably by large cross-sectional dimensions. Large silver-silver halide electrodes in conjunction with halide solutions are the obvious choice (2). The problem of concentration polarization at the four membrane/solution interfaces is not too serious, since the number of coulombs flowing in the system is measured directly. Stirring is helpful in reducing any polarization which may arise on closed circuit.

Our *apparatus*, similar to that used by Neihof and Sollner (2), is shown diagrammatically in Fig. 3. It consisted essentially of two test tube–shaped membranes of different diameters, one placed inside the other so that three compartments were formed. The concentrated electrolyte solution was placed in the middle compartment formed by the two membranes, dilute solution was put inside the inner membrane and in the beaker into which the outer membrane was immersed. The outside solution was stirred by a magnetic stirrer. The two silver-silver halide electrodes were made from silver screen in test tube shape so that they could be placed within 1 to 2 mm of the membrane surfaces. The electrodes were coated electrolytically with a thin layer of AgCl or AgI depending on which halide was used in the experiment.[5] Being in contact with identical solutions, they did not contribute to the E.M.F. arising in the system. The circuit was completed by the addition of a low resistance milliammeter (5Ω) and a switch in order that the system could be studied on open and closed circuit. The experiments were carried out in an air-conditioned room (21°C) so that volume changes due to fluctuations in temperature were insignificant.

In all of our experiments the outer membranes were electronegative membranes of the collodion matrix type which were impregnated with polystyrene sulfonic acid (PSSA) (7). They were cast on test tubes 40 mm in diameter; their effective area was about 120 cm^2. Their porosity was varied by swelling the dense permselective membranes in ethanol of various concentrations (a membrane swollen; *e.g.*, in 95 per cent ethanol being designated as Alc-95) (4, 11).[6] The inner membranes were permselective protamine collodion membranes of low resistance, 10 to 15 ohms-cm^2 (8). They were cast on 25 × 100 mm tubes; their effective area was about 50 cm^2. Their porosity was not varied because no adequate method is available for the preparation of electropositive collodion matrix membranes of graded porosities. Thus the potential across the inner membrane was always higher, and the electroosmotic effects across it were smaller than with the more porous PSSA membranes.

In our models in which large concentration differences of electrolytes exist across

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[5] In order to minimize the resistance of the electrodes, the quantity of halide deposited on them was kept low, not much in excess of the quantity of electricity to be passed in one experiment. Since the resistivity of AgI is considerably lower than that of AgCl, the resistance of silver electrodes when covered with AgI is lower than that of the same electrode covered with an equivalent quantity of AgCl, a significant advantage in our experiments.

[6] The membranes used varied in porosity from fairly dense permselective membranes (unswelled, Alc-0) to membranes with porosities corresponding to conventional dialyzing membranes. The smallest molecules which do not diffuse to any significant extent across the membranes Alc-0, Alc-90, and Alc-95 are urea (MW 60), salmine (MW 3000-6000), and serum albumin (MW 70,000), respectively (4).

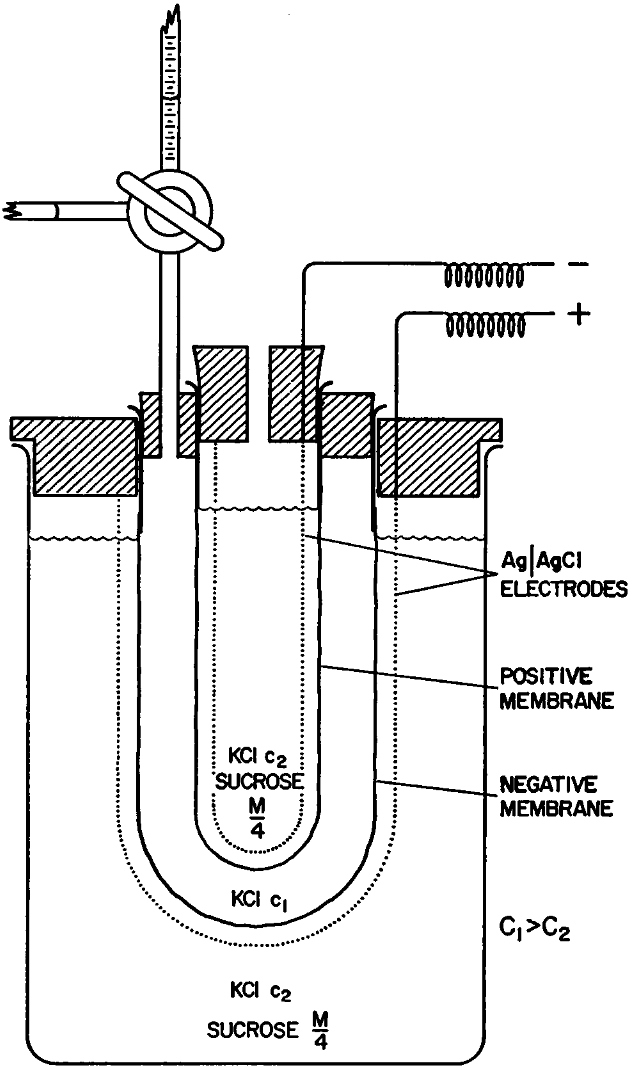

FIGURE 3 Apparatus used for the measurement of the electroosmotic effects arising from the interaction of a selectively anion- and a selectively cation-permeable membrane.

the membranes, both normal and anomalous osmosis can cause considerable fluid movement from the dilute to the concentrated solution, especially if the one membrane is of high porosity. These effects across one membrane will be independent of the analogous effects across the other membrane; superimposed on them will be the possible electroosmosis brought about by the interaction of the two membranes. In preliminary experiments it was found that the normal and anomalous osmosis were so great that it would be very difficult to detect the relatively small electroosmotic effects that might arise from the interaction of the two membranes. Therefore, to decrease drastically the single-membrane effects, sucrose was added to the dilute electrolyte solution in sufficient concentration to counterbalance approximately such volume changes. No attempt was made to obtain an exact balance; it was only necessary to adjust these volume effects to the point where they would be of the same order of magnitude, in the same or opposite direction, as the anticipated electroosmosis.

To carry out an experiment the three compartments were filled with the solutions to be tested and the level in the osmometer was adjusted to about the middle of the graduated capillary so that volume changes in either direction could be measured. With the switch open, the rate of movement of the meniscus was determined over a period of several minutes; then the switch was closed and the rate of movement was again determined. This procedure was repeated several times during each experiment.

RESULTS

Table I gives the detailed data of a typical experiment showing how the final results were obtained.

The reproducibility of our results was tested in a series of experiments, each performed like the one described in Table I. In seven consecutive experiments

TABLE I

THE VOLUME EFFECTS ON OPEN AND CLOSED CIRCUIT IN THE SYSTEM
Ag |AgCl| 0.002 M KCl, 0.25 M SUCROSE |PSSA MEMBRANE*| 0.2 M KCl|
PROTAMINE-COLLODION MEMBRANE‡ |0.002 M KCl,
0.25 M SUCROSE |AgCl| Ag

Time interval on open circuit	Volume change	Time interval on closed circuit	Current	Volume change
min.	*µl*§	*min.*	*ma*	*µl*§
5	−18			
		5	10.0	−32
5	−19			
		5	9.5	−29
5	−16			
		5	9.0	−28
5	−17			
20	−70	15	9.5 (mean)	−89

$\frac{-70}{20} = -3.5$ µl/min. $\frac{-89}{15} = -5.9$ µl/min.

Electroosmosis = −5.9 + 3.5 = − 2.4 µl/min.
= 0.133 mmole of water/min.-9.5 ma
= 23 moles of water/faraday

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*Membrane swelled in 90 per cent ethanol.

‡Permselective membrane, resistance = 10 ohms-cm^2 in 0.1 M KCl.

§The minus sign indicates movement of liquid from the concentrated solution to the dilute solutions.

carried out under nominally identical conditions, the results were 21, 20, 26, 26, 24, 28, and 19 moles of water/faraday. Thus, for these particular conditions, the volume transported was 23.4 ± 1.2 moles/faraday.[7]

The results of experiments in systems with PSSA membranes of different porosities, with the same permselective protamine membrane, and with several electrolytes are summarized in Table II. The figures listed in the last column, moles of water/faraday, are the averages of two or more experiments in all but one instance. It will be seen that with KCl, the electroosmosis in moles of water/faraday increases with increasing porosity of the PSSA membranes in the same way as in the previously reported experiments with an externally applied E.M.F. (4). Similar results were obtained with the sodium and lithium halides.

It was also observed, but is not shown in the table, that the current decreases as the porosity of the PSSA membranes is increased due to the large reduction in potential originating there. As a consequence, the electroosmosis in microliters per

TABLE II

ELECTROOSMOTIC EFFECTS IN THE MOSAIC SYSTEM Ag |AgX| 0.002 M ELECTROLYTE, 0.25 M SUCROSE |PSSA MEMBRANE| 0.2 M ELECTROLYTE| PROTAMINE-COLLODION MEMBRANE* |0.002 M ELECTROLYTE, 0.25 M SUCROSE |AgX| Ag

PSSA Membrane				
Designation of membrane	Water content	Electrolyte	Number of experiments	Electroosmosis
	vol. per cent			*moles of water/faraday*
Alc-0	15	KCl	2	16
Alc-90	30	KCl	7	23
Alc-93	45	KCl	6	34
Alc-95	55	KCl	5	42
Alc-90	30	NaI	3	29
Alc-93	45	NaI	3	43
Alc-95	55	NaI	2	47
Alc-90	30	LiCl	2	32
Alc-90	30	LiI	5	33
Alc-95	55	LiI	1	60

*Permselective membrane, resistance = 10 ohms-cm^2 in 0.1 M KCl.

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[7] To show the influence of sucrose in the dilute solution on the accuracy of the measurement of the electroosmosis, a few data were obtained in experiments without sucrose with a series of PSSA membranes of different porosities. With 0.2 M KCl/0.002 M KCl, the ratio of electroosmosis to the sum of normal and anomalous osmosis was about 1/7 in a system with an unswelled membrane, about 1/15 in a system with a membrane swelled in 90 per cent ethanol, and about 1/100 or less in the case of a membrane swelled in 97 per cent ethanol.

minute reached a maximum in the systems with PSSA membranes of an intermediate degree of porosity, such as the Alc-90 membrane. Further, it was noted that in the iodide systems the current was always higher than in the corresponding chloride systems; for example, with an Alc-95 membrane and NaI the current was 4 to 5 ma, whereas with the same membranes and KCl, the current was never greater than 2 ma. This difference is due to the lower resistance of the Ag|AgI electrodes as compared to the Ag|AgCl electrodes.[5]

According to previous results the rate of electroosmosis with KCl solutions across permselective PSSA collodion membranes (Alc-0) is about the same as that across permselective protamine membranes (4). Applying this fact to the first experiment in Table II, we see that the contribution of each membrane in this mosaic system to the total electroosmosis is about 8 moles of water/faraday. When the porosity of the electronegative membrane is increased in our mosaic system, the increase in electroosmosis in moles of water/faraday occurs entirely across this membrane. In the experiments with KCl and the Alc-95 membrane, the total electroosmosis was 42 moles/faraday. If we subtract the contribution of the protamine collodion membrane, 8 moles/faraday, the contribution of the electronegative membrane is 34 moles/faraday; this agrees with the figure of 30 moles/faraday obtained for electroosmosis with an external E.M.F. across such a membrane (4).

The effect of the hydrated ionic size of the cation, as it was predictable, is also very similar to that which was observed in electroosmosis with an externally applied E.M.F. For example, in the present work with the Alc-95 PSSA membrane, the electroosmosis with potassium, sodium, and lithium was 34, 39, and 52 moles of water/faraday, respectively. In our previous experiments with an externally applied E.M.F. and a somewhat more porous membrane (Alc-97 PSSA), the corresponding values were 50, 68, and 88, respectively (4).

To determine approximately the effective resistance of the mosaic system on closed circuit, a decade resistance box was added to the circuit. In a system with Ag|AgI electrodes and an Alc-90 PSSA membrane, the current without added resistance was 13 ma. As the resistance was increased by known increments, the corresponding drop in current was recorded. The resistance of the original system without added resistance was then calculated to be 14Ω. In a further experiment the resistance box and the milliammeter (resistance 5Ω) were removed from the circuit; the resistance of the circuit was now 9Ω; the corresponding current then was $13 \times 14/9 = 20$ ma. The observed increase in the electroosmosis in microliters per minute under these conditions was in agreement with this increased current, 3.4 μl/min., as compared to 2.5 μl/min. with the milliammeter in the circuit.

DISCUSSION

The experimental data which have been presented here are quite straightforward and do not require extensive comment. The relationship of the quantity of solvent transported by electroosmosis to the quantity of current flowing in the mosaic membrane system is in agreement with that which was observed in systems with an externally applied E.M.F. For our further discussion it is necessary to emphasize the obvious fact that any modification which was made in our system to decrease its resistance always led to an increase in current and a corresponding increase in the rate of electroosmosis.

In extrapolating from the results obtained with the macromodel systems to the effects to be expected with true mosaics, *i.e.* in microsystems, one basic feature of the macrosystems must be stressed, namely, the linear distances in the direction of current flow over which the two membranes interact. While the potentials that arise across the two membranes (with given solutions) are independent of the size of the membranes, the intensity of the current is determined by the resistance of the circuit or circuits, which resistance is largely determined by the linear distances of the system in the direction of the flow of the current. In a true mosaic membrane (see Fig. 1*a*) the pathways of the current through the solutions will be of the same order of magnitude as the dimensions of the individual microelements of the membrane. In macrosystems the linear dimensions of the models are of the order of magnitude of a few centimeters (Fig. 1*c*) and even with the use of auxiliary electrodes (Fig. 3) the length of the pathway of the current through the solutions is of the order of at least several millimeters. In true mosaic membranes the electronegative and electropositive parts of the membrane are separated from each other by small, microscopic or even submicroscopic distances and act upon each other directly without interposed electrodes. The sum of all the local microcurrents and the current density will be correspondingly larger than in the macromodel. In a true mosaic membrane system the local currents which are generated could easily be greater by two orders of magnitude, or more, than those measured in our model system. Thus, it becomes evident that the rate of any current-produced effects in the macro-system will be lower than those arising with a true mosaic in inverse proportion to the distances involved in the two systems. The electroosmotic effect due to the charge mosaic of such a membrane might be of the same order of magnitude or even larger than both the transport of liquid across the membrane by normal osmosis and by the anomalous osmosis which might arise across the various separate parts of a charge-mosaic membrane.

The discussion of electroosmotic effects in true and model mosaic membranes has been confined to systems in which the two solutions were of different concentration but of the same electrolyte. It is obvious that analogous electroosmotic effects can also arise in systems in which a charge-mosaic membrane separates solutions of different (or mixed) electrolytes either of the same or different concentrations.[8] It is readily understood that, except in rare cases, potentials will arise

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[8] The normal and anomalous osmotic effects arising in systems with solutions of different com-

across the two different membranes which will be different, in many instances even of opposite sign. Thus, electric currents are bound to flow; they will be accompanied by electroosmotic effects, the magnitude of which will depend on the porosities of the membranes and the ions involved. A few test experiments proved the existence of such an effect. For instance, with 0.1 M LiCl in the one solution and 0.1 M KI in the other and with permselective membranes, a significant current, 1 ma, was obtained transporting 18 moles of water/faraday.[9]

The question of whether or not the electroosmotic transport of water across charge-mosaic membranes is actually involved in water transport in living systems is outside the scope of this paper; here it only has been demonstrated that significant electroosmotic transport by a mechanism involving charge-mosaic membranes is possible.

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position and (non-mosaic) membranes of a single sign of charge have recently been described (12) (and shown to occur even if both solutions are in the osmolality range of 0.2 to 0.4).

[9] These particular electrolytes were used, to take advantage of the relatively high bi-ionic potentials, Cl^- *versus* I^- across the electropositive membrane and Li^+ *versus* K^+ across the electronegative membrane (13).