

Liquid membrane phenomena: Characterization of liquid membranes generated by lecithin[†]

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The transport behaviour of liquid membranes generated by lecithin at the sintered glass membrane-water interface has been studied. Data on hydraulic permeability, electroosmotic velocity, streaming potential and streaming current have been used to calculate the transport coefficients (R_{ik}) accounting for liquid membranes, using the theory developed by Kedem and Katchalsky for composite series membranes. The trends observed in degree of coupling corresponding to liquid membranes are indicative of the applicability of liquid membranes as model systems for biological membranes. The efficiencies of electro-kinetic energy conversion for both electroosmosis and streaming potential have been evaluated and the results thus obtained have been shown to be consistent with the steady state thermodynamic theory.

Introduction

The spontaneous orientation of surfactant molecules at the interface leads to a reduction in interfacial energy and is of fundamental importance in the formation of molecular aggregates and thin films which persist in liquid state¹. Thus, liquid membranes form spontaneously even without impingement as a result of surfactant capacity of dissolved molecules². Surface activity is, of course, related to the hydrophilic and hydrophobic balance of these molecules¹.

Because of the surface active nature of lecithin^{3,4}, it is natural to expect that in the liquid membranes generated by lecithin the hydrophobic end of these molecules will be preferentially oriented towards the supporting membrane and hydrophilic end will be drawn outward away from the supporting membrane^{1,3}. Liquid membranes generated in this way form composite structure with the supporting membrane with which they act in series⁴⁻⁷.

In this paper we report electroosmosis of water through liquid membranes generated by lecithin at the sintered glass membrane-water interface. The transport data have been analysed using the theory of thermodynamics of irreversible processes^{8,9} with a view to evaluating transport coefficients accounting for liquid membranes alone and calculating the degree of coupling and the efficiency of energy conversion across liquid membranes. Analysis of the data indicates formation of multilayers of liquid membranes. Evidence is also provided for the suitability

of these membranes as model system for biological membranes.

Theory

The phenomenological relations for the simultaneous transport of matter and electricity in the linear range can be expressed as⁸,

$$J_v = L_{11} \Delta P + L_{12} \Delta \phi \quad \dots (1)$$

$$I = L_{21} \Delta P + L_{22} \Delta \phi \quad \dots (2)$$

where J_v and I are volume flow and flow of electricity, respectively, and ΔP and $\Delta \phi$ are their conjugate driving forces. Coefficients L_{ik} are called the phenomenological coefficients and in view of Onsager's reciprocal relation (ORR), we have

$$L_{12} = L_{21} \quad \dots (3)$$

In view of Eqs (1) and (2), the conversion efficiency, η for electroosmosis and streaming potential can be written as¹⁰

$$\eta_e = -J_v \Delta P / I \Delta \phi \quad \dots (4)$$

$$\eta_s = -I \Delta \phi / J \Delta P \quad \dots (5)$$

where subscripts e and s stand for phenomena of electroosmosis and streaming potential respectively.

In a coupled system the maximum conversion efficiency, η_{max} , is uniquely determined by degree of coupling, q , which is related to the transport coefficients in the following manner¹¹,

$$q = -\frac{R_{12} \text{ or } R_{21}}{\sqrt{R_{11} R_{22}}} \quad \dots (6)$$

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where R_{ik} coefficients are related to the L_{ik} coefficients by Eq. (7).

$$R_{ik} = |L_{ik}|/|L| \quad \dots (7)$$

$|L_{ik}|$ being the minor of the determinant corresponding to L_{ik} and $|L|$ is the determinant of the matrix of L_{ik} . Since for the symmetrical matrix L_{ik} the complements $|L'_{ik}|$ are symmetrical too, the R_{ik} 's obey the ORR.

$$R_{ik} = R_{ki} \quad \dots (8)$$

It has been shown in our earlier work⁷ that for the series membranes the degree of coupling and maximum conversion efficiency are composed additively of elementary contributions of the constituent membrane elements and each contribution is weighted by a fraction of straight coefficients. These relations can be written as⁷, for degree of coupling,

$$q^T = \sum_r q^r b_{ik}^r \quad \dots (9)$$

and for maximum conversion efficiency,

$$\sqrt{\eta_{\max}^T} = \sum_r \sqrt{\eta_{\max}^r} b_{ik}^r \quad \dots (10)$$

where

$$b_{ik}^r = \left(\frac{R_{11}^r R_{22}^r}{R_{11}^T R_{22}^T} \right)^{1/2}$$

and superscripts T and r stand for total composite series membrane and rth membrane element in the series system.

Materials and Methods

Lecithin (Sigma) was recrystallized from absolute alcohol before use in order to avoid possible formation of black films³. Water was distilled once over potassium permanganate and ethanol was purified by the method described in the literature⁶. Aqueous solutions of lecithin were prepared by the method used earlier^{5,12,13}. The critical micelle concentration, CMC, of aqueous solutions, determined as reported earlier⁶, was found to be $1.7 \pm 0.1 \times 10^{-5} M$.

The electroosmotic cell used in the present study and details of procedure followed for the measurements of hydraulic permeability, electroosmotic velocity, streaming potential and streaming current have already been described in an earlier publication⁶. Sintered glass membrane (porosity G4) of thickness $2.43 \times 10^{-2} m$ and area $7.74 \times 10^{-4} m^2$, which separated the transport cell into two compartments was used to support the liquid membranes. The whole study was made at constant temperature by placing the electroosmotic cell in a thermostat maintained at $38 \pm 0.1^\circ C$.

In the present work, liquid membranes were generated in two situations, i.e., on one side and on both sides of the supporting membrane. In situation (i), one compartment (say I) was filled with lecithin solution of desired concentration and other compartment (say II) was filled with water. In situation (ii), both compartments were filled with lecithin solutions of same concentration. In all experiments the solution was monitored with a conductivity meter (Toshniwal, India) to note the aging of the solution and if any change in the conductance was recorded, the solution was replaced by fresh aqueous solution of the same concentration.

Results and Discussion

The data of hydraulic permeability, electroosmotic velocity, streaming potential and streaming current obtained in situation (i) and in situation (ii) have been plotted in Figs 1-4. In all cases straight line plots were obtained which are in conformity with the linear Eqs (1) and (2). Values of various phenomenological coefficients, viz., L_{11} , L_{12} , L_{21} and L_{22} for all concentrations of lecithin and for both situations estimated from the slopes of above mentioned plots are summarized in Table 1. However, it should be pointed out that the maximum error, calculated from the estimates of various errors, in experimentally determined values was never more than $\pm 5.0\%$. The validity of ORR is also obvious from data in Table 1.

Concentration dependence of transport coefficients

An examination of the values of phenomenological coefficients (Table 1) reveals that the L_{ik} coefficients decrease with increasing concentration of lecithin. An obvious implication of this observation is that the decrease in L_{ik} values is due to possible formation of multilayers^{6,7,13,14} at the interface by lipid-lipid interactions and hydrogen bonding in aqueous medium. The lowering of interfacial energy is consistent with the fact that adsorption also occurs spontaneously. Since interfacial energy decreases with increase in concentration, the adsorption is expected to increase and exceed monolayer capacity of the surface¹⁵. In such a situation, a model based on multilayer adsorption is indicative¹⁵. Though physical existence of these layers arranged in series cannot be distinguished in our macroscopic transport experiments, qualitatively, their contribution in increasing the resistance is not too insignificant to be ignored (Table 2).

Evaluation of transport coefficient for liquid membranes

It is plausible to consider under the present experimental conditions that the series membrane gener-

ated in situation (i) consists of two membrane elements, viz., supporting membrane and one unit of lecithin liquid membrane. While in situation (ii), the series system can be described by three constituent

membrane elements, i.e., the supporting membrane and two unit layers of lecithin liquid membrane—one unit on each side of the supporting membrane. With these considerations, we can write^{9,16} for situation (i) as,

$$R_{ik}^T = R_{ik}^g + R_{ik}^l \quad \dots (11)$$

and for situation (ii) as

$$R_{ik}^* = R_{ik}^g + 2R_{ik}^l \quad \dots (12)$$

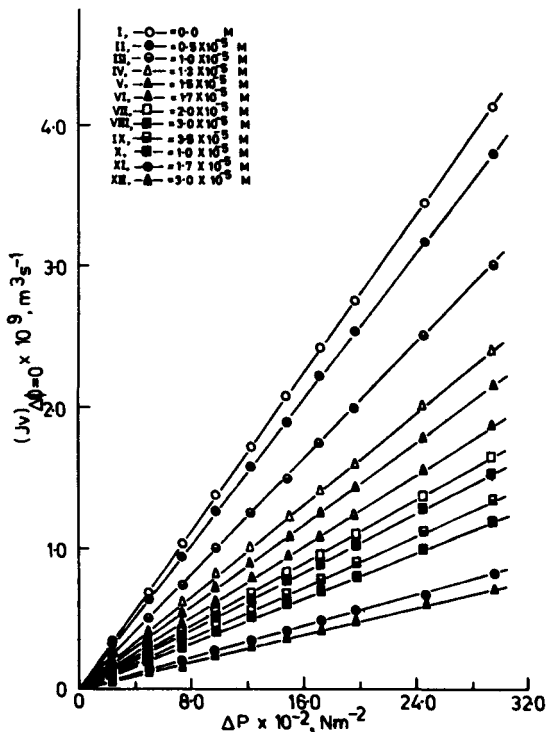


Fig. 1—Hydraulic permeability data for various concentrations of lecithin [Curves X to XII represent situation (ii)]

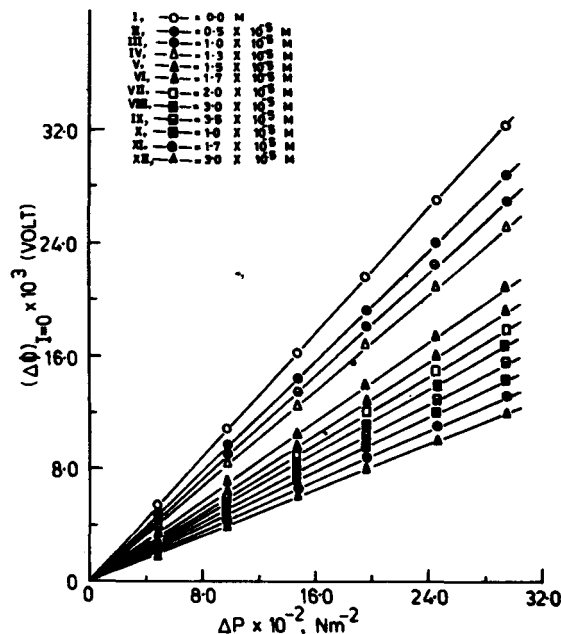


Fig. 3—Streaming potential data for various concentrations of lecithin [Curves X to XII represent situation (ii)].

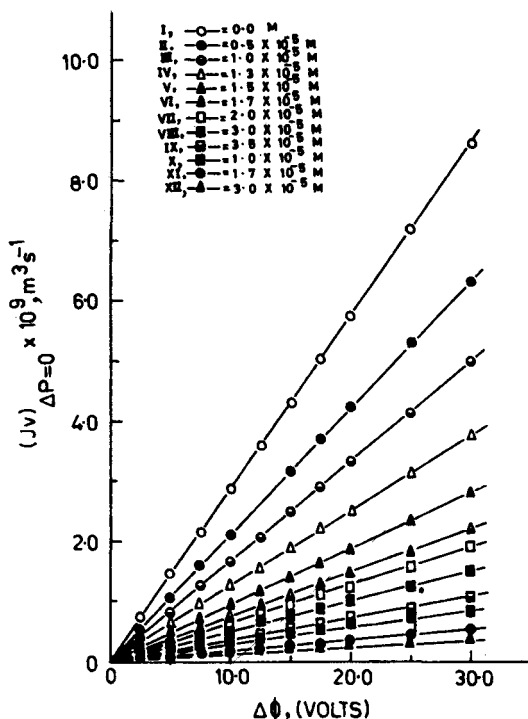


Fig. 2—Electroosmotic velocity data for various concentrations of lecithin [Curves X to XII represent situation (ii)]

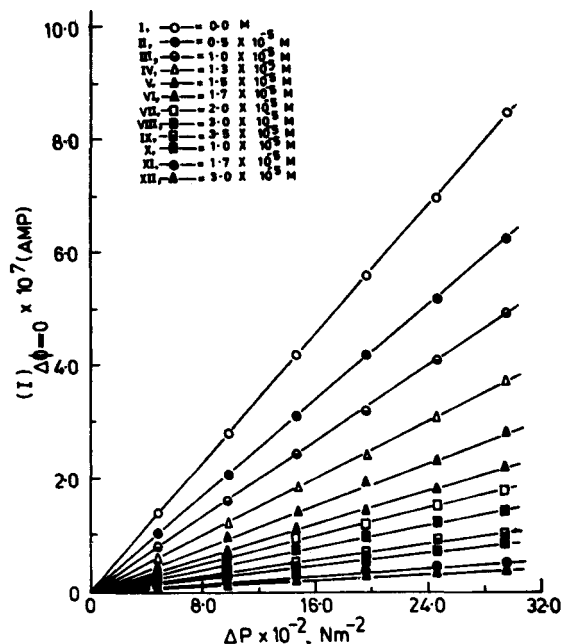


Fig. 4—Streaming current data for various concentrations of lecithin [Curves X to XII represent situation (ii)].

Table 1—Values of L_{ik} coefficients at different concentration of Lecithin

[Lecithin] ($10^{-5}M$)	$L_{11} \times 10^{12}$ ($m^2N^{-1}S^{-1}$)	$L_{12} \times 10^{10}$ (m^3AJ^{-1})	$L_{21} \times 10^{10}$ (m^3AJ^{-1})	$L_{22} \times 10^4$ (AV^{-1})
(a) Situation (i)				
0.0	1.407	2.88	2.89	0.264
0.5	1.292	2.12	2.11	0.217
1.0	1.020	1.66	1.64	0.180
1.3	0.812	1.26	1.24	0.146
1.5	0.734	0.94	0.92	0.130
1.7	0.633	0.74	0.74	0.114
2.0	0.560	0.64	0.63	0.103
3.0	0.522	0.50	0.50	0.088
3.5	0.456	0.36	0.35	0.068
(b) Situation (ii)				
1.0	0.402	0.284	0.280	0.058
1.7	0.280	0.182	0.182	0.041
3.0	0.240	0.140	0.138	0.034

In Eqs (11) and (12) superscripts g, l, T and * stand for sintered glass membrane, lecithin liquid membrane, series membrane in situation (i) and series membrane in situation (ii) respectively. Before making use of Eqs (11) and (12) for evaluating the values of transport coefficients corresponding to liquid membrane in two situations, it is necessary to transform L_{ik} coefficients given in Table 1 into R_{ik} coefficients using Eq. (7). R_{ik} coefficients thus estimated are recorded in Table 2. The values of R_{ik}^l in Table 3(a) were calculated using Eq. (11) whereas the values of R_{ik}^g given in Table 3(b) were obtained using Eq. (12). The decreasing trend of R_{ik} coefficients (Table 2) and R_{ik} coefficients (Table 3) further corroborates the formation of multilayers at the interface.

Liquid membranes generated in the present study have water permeability lower than that reported for BLM³ but closer to that for biomembranes¹⁷. Values of electrical resistance of freely formed lipid bilayers in general are much higher³ than those for biomembranes^{3,17}. Liquid membranes formed in this study have values of electrical resistance comparable with those reported for biomembranes^{3,17}. This indicates the applicability of these membranes as models for biomembranes (Table 4).

Degree of coupling

Degrees of coupling for both modes, namely, electroosmosis and streaming potential, have been calculated¹³ using Eq. (6). From Table 2 it is obvious that the degrees of coupling for both modes are equal as a consequence of ORR. Further it can be

seen from Table 2 that q decreases with increasing concentration of lecithin in the dispersion. This trend is less pronounced for q values accounting for liquid membranes alone (Table 3).

We now focus attention on testing Eq. (9) which in situation (i) can be expressed as,

$$q^T = q^g \left(\frac{R_{11}^g R_{22}^g}{R_{11}^T R_{22}^T} \right)^{1/2} + q^l \left(\frac{R_{11}^l R_{22}^l}{R_{11}^T R_{22}^T} \right)^{1/2} \quad \dots (13)$$

and in situation (ii)

$$q^* = q^g \left(\frac{R_{11}^g R_{22}^g}{R_{11}^* R_{22}^*} \right)^{1/2} + 2q^l \left(\frac{R_{11}^l R_{22}^l}{R_{11}^* R_{22}^*} \right)^{1/2} \quad \dots (14)$$

The values of the q^T can be computed at any concentration of the lecithin solution using the values of q^g , $R_{11}^g R_{22}^g R_{11}^T$ and R_{22}^T (Table 2a) and q^l , R_{11}^l and R_{22}^l (Table 3a) in Eq. (13). Functionally, R_{ik}^g are the values corresponding to zero concentration. R_{ik}^T are the values corresponding to any concentration of lecithin and R_{ik}^l are the values corresponding to the liquid membrane alone at any definite concentration of lecithin. The value of q^T thus computed at $1.7 \times 10^{-5}M$ concentration of lecithin comes out to be 2.75×10^{-2} which agrees well with values determined from Eq. (6) (Table 5). To test Eq. (14), the values of R_{11}^l and R_{22}^l were noted from Table 3(b) and R_{11}^* and R_{22}^* values were noted from Table 2(b) at $1.7 \times 10^{-5}M$ concentration. The values of q^* estimated from Eq. (14) were found in good agreement (Table 5). However, when values of R_{11}^l and R_{22}^l were used from Table 3(a), the values of q^* obtained from Eq. (14) were not found to agree with the va-

Table 2—Values of various resistance coefficients, β , η_{\max} and q for series systems of lecithin liquid membrane

[Lecithin] (10^{-5} M)	$R_{11} \times 10^{-12}$ ($m^{-5} N S$)	$-R_{12} \times 10^{-6}$ ($m^{-3} A^{-1} J$)	$-R_{21} \times 10^{-6}$ ($m^{-3} A^{-1} J$)	$R_{22} \times 10^{-4}$ (ohm)	$\beta_s \times 10^3$	$\eta_e \max \times 10^3$ from	$\eta_s \max \times 10^3$ from	$\eta_e \max \times 10^3$ from the plot	$\eta_s \max \times 10^3$ from the plot	$q_e \times 10^2$
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(a) When liquid membranes were generated in series with supporting membrane interface on one side; situation (i)

0.0	0.710	7.750	7.780	3.787	2.252	2.248	0.558	0.562	0.57	0.57	4.72	4.74
0.5	0.773	7.520	7.561	4.608	1.603	1.588	0.401	0.397	0.38	0.40	3.90	4.00
1.0	0.980	9.050	8.943	5.555	1.500	1.465	0.375	0.366	0.34	0.33	3.87	3.83
1.3	1.231	10.628	10.459	6.849	1.340	1.227	0.335	0.324	0.32	0.32	3.66	3.60
1.5	1.362	9.510	9.641	7.692	0.926	0.887	0.231	0.222	0.22	0.20	3.04	2.97
1.7	1.580	10.254	10.240	8.772	0.758	0.759	0.189	0.189	0.20	0.19	2.75	2.75
2.0	1.785	10.922	11.095	9.708	0.710	0.688	0.177	0.172	0.14	0.14	2.62	2.66
3.0	1.915	10.884	10.862	11.364	0.544	0.544	0.136	0.136	0.12	0.10	2.33	2.33
3.5	2.192	11.609	11.287	14.705	0.418	0.395	0.103	0.090	0.09	0.09	2.04	1.58

(b) When the supporting membrane interface was sandwiched between layers of lecithin liquid membrane; situation (ii)

1.0	2.488	12.180	12.094	17.241	0.346	0.336	0.086	0.084	0.080	0.084	1.859	1.640
1.7	3.571	15.854	15.854	14.370	0.288	0.288	0.072	0.072	0.068	0.070	1.698	1.698
3.0	4.166	17.156	16.912	29.411	0.240	0.233	0.060	0.058	0.048	0.068	1.549	1.527

[Lecithin] ($10^{-5}M$)	Table 3—Values of various resistance coefficients (R_{ik}^1), q^1 and η_{\max}^1 for lecithin liquid membranes							
	$R_{11}^1 \times 10^{-12}$ ($m^{-5}NS$)	$-R_{12}^1 \times 10^{-6}$ ($m^{-3}A^{-1}J$)	$-R_{21}^1 \times 10^{-6}$ ($m^{-3}A^{-1}J$)	$R_{22}^1 \times 10^{-4}$ (ohm)	$q_c^1 \times 10^2$	$q_s^1 \times 10^2$	$\eta_{e,\max}^1 \times 10^4$ from plot η_c^1 vs ΔP	$\eta_c^1 \times 10^4$ from q^1
	(a) R_{ik}^1 values evaluated considering unit layer of liquid membranes, i.e., situation (i)							
1.0	0.270	1.300	1.263	1.768	1.88	1.68	0.88	0.88
1.7	0.869	2.500	2.460	4.985	1.20	1.18	0.34	0.36
3.0	1.205	3.134	3.082	7.577	1.04	1.04	0.24	0.27
	(b) R_{ik}^1 values evaluated considering two unit layers of liquid membranes, i.e., situation (ii)							
1.0	0.889	2.215	2.157	6.727	0.91	0.88	0.20	0.21
1.7	1.430	4.052	4.037	10.302	1.06	1.05	0.26	0.28
3.0	1.728	4.703	4.566	12.812	0.99	0.97	0.20	0.24

Table 4—Comparison of permeabilities and resistances of lecithin, BLM and biomembranes			
System	Permeability	Method	Resistance
Lecithin	$10^{-5}ms^{-1}$	Pressure difference	10^2 ohm m^2
BLM	10^{-5} to $10^{-6}ms^{-1}$	Osmotic and diffusion	1 to 10^7 ohm m^2
Biomembranes	10^{-5} to $10^{-6}ms^{-1}$	Osmotic and diffusion	10 to 10^4 ohm m^2

[Lecithin] ($10^{-5}M$)	Table 5—Comparison of the values of q^T and q^* calculated from Eqs (13) and (14)				
	$q^T \times 10^2$		$q^* \times 10^2$		
	Calc. from Eq. (13)	Calc. from Eq. (6)	Calc. from Eq. 14		
			Using R_{ik} values from Table 3(a)	Using R_{ik} values from Table 3(b)	Calc. from Eq. (6)
1.0	3.87	3.87	1.58	1.86	1.86
1.7	2.75	2.75	1.36	1.70	1.70
3.0	2.33	2.33	1.29	1.54	1.55

values determined using Eq. (6). This discrepancy is indicative of multilayer formation of liquid membranes.

Electro-kinetic energy conversion

From the discussion of steady state energy conversion defined by Eqs (4) and (5), the following conclusions have been arrived at^{10,18,19}:

(1) The maximum values for conversion efficiency, η_{\max} , for a fixed value of input force always occurs when the output force equals half its steady state value.

(2) The values of η_{\max} are independent of the input force.

(3) $\eta_{e,\max} = \eta_{s,\max}$, which in fact is a consequence of ORR.

Following the method described earlier¹⁰, the values of η_e and η_s were calculated using Eqs (4) and (5) for sintered glass membrane, series membranes in situation (i), series membranes in situation (ii) and

also for lecithin liquid membranes alone. Representative relationships between η_e and ΔP and η_s and $\Delta\phi$ corresponding to two fixed values of input forces are shown in Figs 5 and 6. These figures clearly demonstrate the validity of all the three conclusions listed above. The values of $\eta_{e,\max}$ and $\eta_{s,\max}$ for all concentrations and for all systems obtained from the maxima of the curves of the type shown in Figs 5 and 6 are given in Tables 2 and 3.

The values of η_{\max} for all systems were also computed from the values of L_{ik} (Table 1) using the relationship²⁰,

$$\eta_{\max} = \frac{(1 + \beta_{io})^{1/2} - 1}{(1 + \beta_{io})^{1/2} + 1} \quad \dots (15)$$

where

$$\beta_{io} = \left(\frac{L_{ii}L_{oo}}{L_{io}^2} - 1 \right)^{-1} \quad \dots (16)$$

in which subscripts o and i represent the output and input powers. The values of η_{max} thus estimated match the values obtained from experimental curves of the type shown in Figs 5 and 6. The quantity β in Eq. (15) is termed as figure of merit²⁰ and it follows as a consequence of ORR that

$$\beta_{io}(\text{or } \beta_e) = \beta_{oi}(\text{or } \beta_s) \quad \dots(17)$$

The validity of Eq. (17) is obvious from Table 2. The values of η_{max} calculated using Eq. (15) are in fair agreement with the values predicted from the maxima of the curves plotted between η_e and ΔP and η_s and $\Delta\phi$ (Table 2). This indicates that the results discussed here are consistent with the theoretical deductions¹⁰ and experimental findings⁷.

The Eq. (10) for series system in situation (i) can be written as,

$$\sqrt{\eta_{max}^T} = \sqrt{\eta_{max}^g} \left(\frac{R_{11}^g R_{22}^g}{R_{11}^T R_{22}^T} \right)^{1/2} + \sqrt{\eta_{max}^i} \left(\frac{R_{11}^i R_{22}^i}{R_{11}^T R_{22}^T} \right)^{1/2} \quad \dots(18)$$

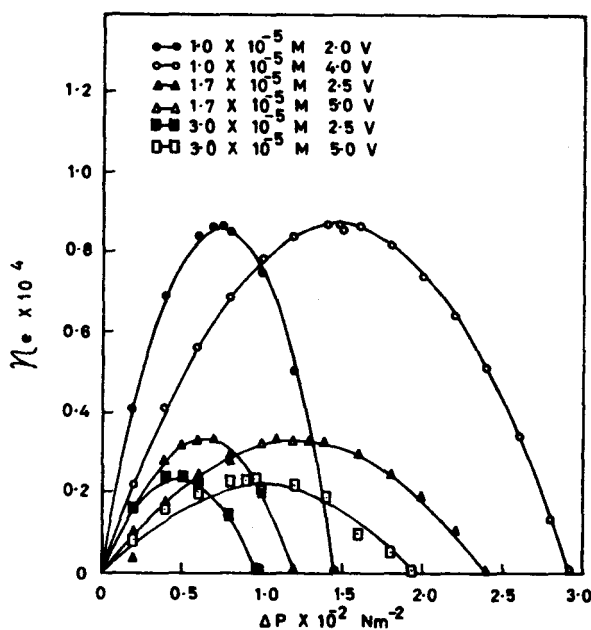


Fig. 5—Dependence of η_e on output force ΔP accounting for liquid membrane alone.

In situation (ii), the Eq. (10) can be recast as

$$\sqrt{\eta_{max}^*} = \sqrt{\eta_{max}^g} \left(\frac{R_{11}^g R_{22}^g}{R_{11}^* R_{22}^*} \right)^{1/2} + 2 \times \sqrt{\eta_{max}^i} \left(\frac{R_{11}^i R_{22}^i}{R_{11}^* R_{22}^*} \right)^{1/2} \quad \dots(19)$$

The Eqs (18) and (19) were tested by a method similar to that used in testing Eqs (13) and (14). The Eq. (18) was found valid in toto whereas Eq. (19) was found valid only when values of R_{ik}^i and R_{ik}^i were taken from Table 3(b) and Table 2(a) respectively. The comparison of the η_{max}^T and η_{max}^* values has been shown in Table (6).

The discrepancies obtained in q^* (Table 5) and η_{max}^* (Table 6) values can be rationalized in terms of polarity of flow which leads to a correlation between macroscopic observables and membrane struc-

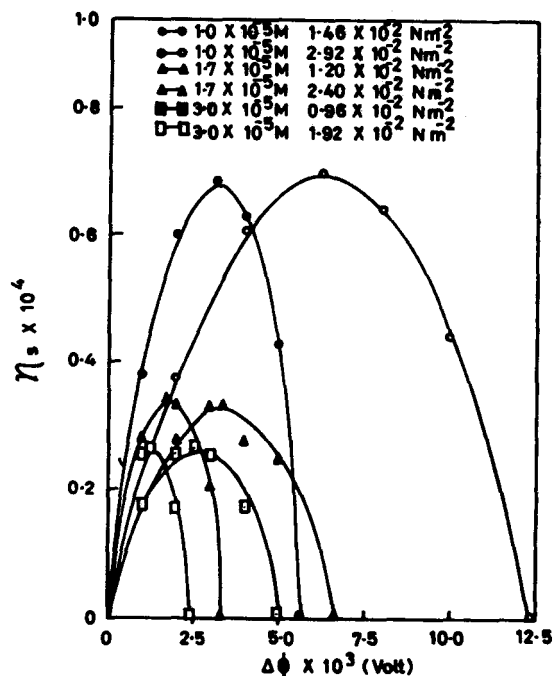


Fig. 6—Dependence of η_s on output force $\Delta\phi$ accounting for liquid membrane alone.

Table 6—Comparison of the values of η_{max}^T and η_{max}^* calculated from Eqs (18) and (19)

[Lecithin] (10 ⁻⁵ M)	$\eta_{max}^T \times 10^3$		$\eta_{max}^* \times 10^3$		
	Calc. from Eq. (18)	Predicted from the plot of η_e^T vs ΔP	Calc. from Eq. (19)		Predicted from the plot of η_e^* vs ΔP
			Using R_{ik} from Table 3(a)	Using R_{ik} from Table 3(b)	
1.0	0.37	0.38	0.064	0.086	0.086
1.7	0.18	0.18	0.046	0.072	0.070
3.0	0.13	0.14	0.040	0.060	0.060

ture¹⁶. The polarity of flow is expected to arise from the transition layers which may be considered as additional membrane elements. This situation is likely to occur in the present system because of specific orientation²¹ of the surfactant molecules at the interface and interaction between the surfactant molecules themselves by lipid-lipid interaction through hydrogen bonds²².

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