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Effect of Plasticizer on Carrier-Mediated Transport of Zinc Ion through Cellulose Triacetate Membranes

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NOTE

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INTRODUCTION

In practical applications of liquid membranes containing a mobile carrier, three main types of membranes have been studied: liquid surfactant, supported liquid, and solvent polymeric membranes (*1–6*). In these membranes the solvent polymeric membrane which consists of a polymer film containing a plasticizer generally exhibits a low ionic permeability compared with the other types of membranes, although it has a high stability and durability. This is because the transference of the carrier in the membrane is depressed by its high viscosity and high electrical resistance. Accordingly, the ionic permeability may be improved by selecting an appropriate plasticizer.

In the present work the fluxes of zinc ion across cellulose triacetate membranes containing bathophenanthroline as a carrier of zinc ion have been determined using various kinds of plasticizers and their mixtures, and the effect of plasticizers on the flux has been demonstrated.

EXPERIMENTAL

In 200 mL dichloromethane, 2.5 g cellulose triacetate (CTA) obtained from Eastman Kodak Co. were dissolved. Five milliliters of the CTA solution, 2 mL of a dichloromethane solution containing 10 m*M* bathophenanthroline (4,7-diphenyl-1,10-phenanthroline), and 0.25 to 5 mL of a dichloromethane solution containing 10 vol% plasticizer were mixed in a glass culture dish (flat bottom, 6 cm diameter). The culture dish was then covered with a glass plate and left overnight at room temperature. In the meantime the mixed solution was slowly air-dried. After the culture dish was filled with water, the resulting membrane was peeled from its bottom. The thicknesses of the membranes were 0.02 to 0.13 mm. The bathophenanthroline was obtained from Dojindo Lab. The plasticizers used are given in Table 1. The plasticizer ONPPE was obtained from Eastman Kodak Co.; ONPOE from Dojindo Lab.; PNPHE, DBPT, MPEG, EPEG, BPBG, EB, EEB, and DEGOE from Tokyo Kasei Kogyo Co.; TBEP from Aldrich Chemical Co.; TEHP from Wako Pure Chemical Ind.; and TMTP from Kanto Chemical Co.

The apparatus used for measuring the zinc-ion flux was similar to that previously used (*6*): the permeation cell consisted of two cylindrical glass compartments (half cell volume, 40 mL; effective membrane area, 7.07 cm^2).

The CTA membrane was fixed between two compartments of the cell. To each compartment was then added 40 mL of an aqueous solution: one (Compartment I) contained initially 0.5 m*M* zinc sulfate and 0.1 *M* sodium chloride, and the other (Compartment II) contained 0.5 m*M* zinc sulfate and 0.05 *M* sodium sulfate. All permeation experiments were made at 25°C. The zinc-ion concentrations in the two compartments were determined as a function of time by the zincon method (*7*), after 0.2 mL of the aqueous solutions had been pipetted off. Measurements of the membrane potential were carried out by the method described elsewhere (*8*).

RESULTS AND DISCUSSION

Plots of zinc-ion concentrations in the two compartments and membrane potential against time for the CTA membranes containing ONPOE, TBEP, and their 1:1 mixture are shown in Fig. 1. The release of zinc ion into the Compartment II increased by combining ONPOE and TBEP. A similar behavior was observed for the other combinations of plasticizers. Table 1 shows the fluxes of zinc ion for the CTA membranes

TABLE 1

Fluxes of Zinc Ion across the Cellulose Triacetate Membranes Containing Bathophenanthroline and Plasticizers

No.	Plasticizer[a]	Zinc-ion flux[b] × 10^7 (mol/cm^2 · h)
1	*o*-Nitrophenyl phenyl ether (ONPPE)	1.03
2	*o*-Nitrophenyl *n*-octyl ether (ONPOE)	0.76
3	*p*-Nitrophenyl *n*-heptyl ether (PNPHE)	0.74
4	Tris(2-*n*-butoxyethyl) phosphate (TBEP)	0.50
5	Tris(ethylhexyl) phosphate (TEHP)	0
6	Tri-*m*-tolyl phosphate (TMTP)	0.27
7	Di-*n*-butyl phthalate (DBPT)	0.33
8	Methyl phthalyl ethyl glycolate (MPEG)	0
9	Ethyl phthalyl ethyl glycolate (EPEG)	0
10	*n*-Butyl phthalyl *n*-butyl glycolate (BPBG)	0.29
11	Ethyl benzoate (EB)	0.36
12	2-Ethoxyethyl benzoate (EEB)	0.19
13	Diethylene glycol monooleyl ether (DEGOE)	0.08
14	ONPOE-TBEP	1.46
15	ONPOE-TEHP	1.17
16	ONPOE-TMTP	0.66
17	ONPOE-EB	1.38
18	ONPOE-EEB	1.06
19	ONPOE-DEGOE	1.21
20	PNPHE-TBEP	1.17
21	PNPHE-TEHP	0.94
22	PNPHE-TMTP	0.43
23	PNPHE-EB	1.37
24	PNPHE-EEB	0.98
25	PNPHE-DEGOE	0.98
26	ONPPE-TBEP	1.58
27	TEHP-TBEP	0.52
28	TMTP-TBEP	0.83
29	DBPT-TBEP	1.00
30	MPEG-TBEP	0.50
31	EPEG-TBEP	0.68
32	BPBG-TBEP	1.02

[a] Plasticizers 14 to 32 are 1:1 mixtures.

[b] Mean values in the range of 1 to 5 h after the start of the zinc-ion transport.

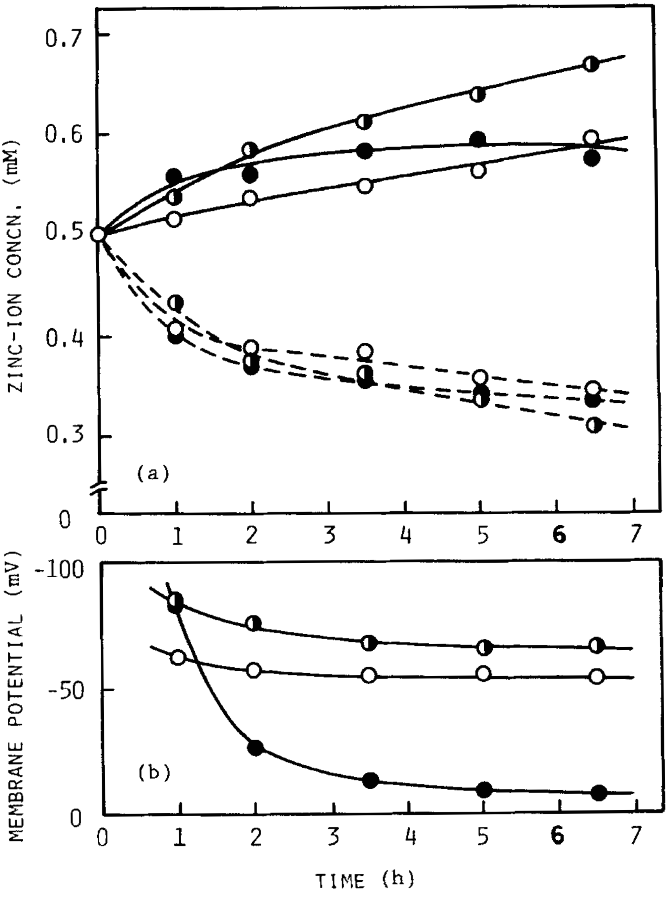

FIG. 1. Plots of zinc-ion concentrations in the two compartments (a) and membrane potential (b) against time for the CTA membranes containing ONPOE (○), TBEP (●), and their 1:1 mixture (◑). The dashed and solid lines (a) represent the concentration curves in Compartments I and II, respectively. The polarity of the membrane potential (b) was negative in Compartment II with respect to Compartment I. The plasticizer content was 2.4 mL/g · CTA.

containing various kinds of plasticizers and their 1:1 mixtures. The mixtures of nitrophenol derivatives and TBEP or EB were most effective in transporting zinc ion. The nitrophenol derivatives have high dielectric constants (5). In this experiment they indicated comparably high fluxes of zinc ion. On the other hand, when TBEP or EB was used independently in the present membrane system, the membrane potential decreased rapidly with the elapse of time, and at the same time, the release of zinc ion into Compartment II decreased remarkably (Fig. 1). The addition of these plasticizers may lower the lipophilicity of the membrane: a reasonable lipophilicity increases the zinc-ion flux.

Plots of zinc-ion flux against the volume fraction of TBEP in ONPOE–TBEP mixture are shown in Fig. 2. A maximal flux was observed at a volume fraction of TBEP of 0.33.

Figure 3 shows the effect of plasticizer content on the zinc-ion flux. The value of J × L (J: zinc-ion flux, L: membrane thickness) in Fig. 3 is approximately proportional to the diffusion constant of carrier in the membrane. Hence, the diffusion rate of the carrier rises linearly with the plasticizer content.

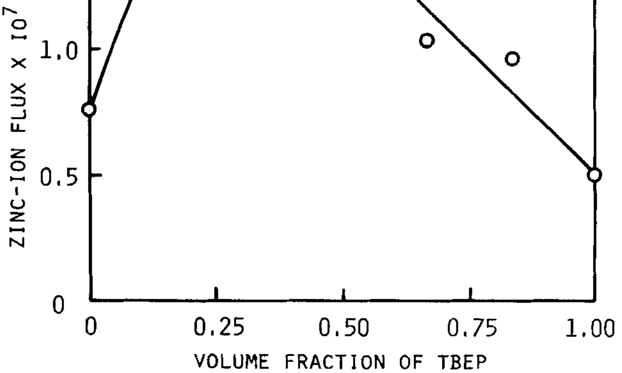

FIG. 2. Plots of zinc-ion flux against volume fraction of TBEP in ONPOE–TBEP mixture. The plasticizer content was 2.4 mL/g · CTA.

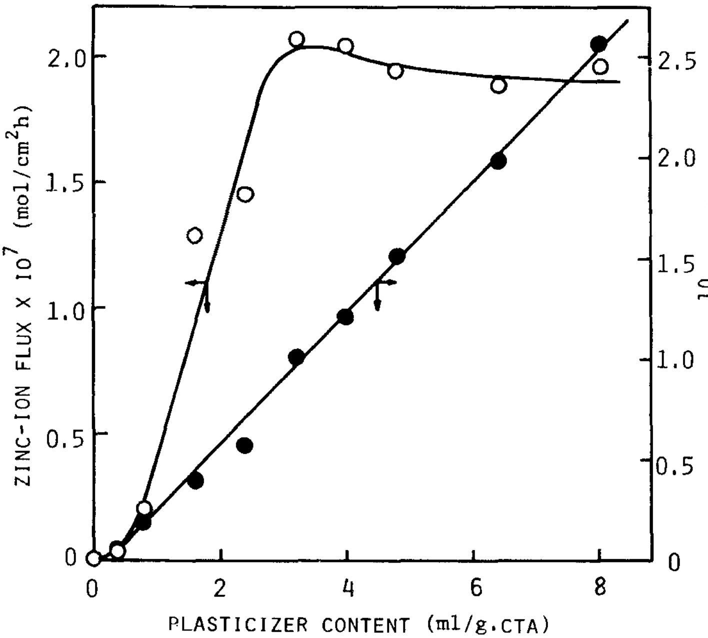

FIG. 3. Plots of zinc-ion flux (○) and value of J × L (●, J = zinc-ion flux, L = membrane thickness) against plasticizer content for the CTA membrane containing a ONPOE-TBEP 1:1 mixture.

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