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Effect of Phospholipids on Carrier-Mediated Transport of Lanthanide Ions through Cellulose Triacetate Membranes

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NOTE

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ABSTRACT

Fluxes of 14 kinds of lanthanide ions across cellulose triacetate membranes were determined by using mixtures of *o*-nitrophenyl *n*-octyl ether and synthetic phospholipids as plasticizers, and 4-benzoyl-3-methyl-1-phenyl-5-pyrazolone (BMPP) and hinokitiol (HIPT) as carriers. The phospholipids used were didecanoyl-, dilauroyl-, and dimyristoyl-L-α-phosphatidylcholines. The effect of these phospholipids on the flux was demonstrated. The flux using BMPP increased with an increase in ionic radius of the lanthanide and reached a maximum value at an ionic radius around that of dysprosium. Subsequently, it decreased and then increased again in the range of praseodymium to lanthanum. The variation in the flux decreased with an increase in alkyl chain length of the phospholipid. For HIPT, an appreciable increase in the flux was observed in the range of erbium to europium.

INTRODUCTION

In a previous work (1) fluxes of 14 kinds of trivalent lanthanide ions across cellulose triacetate (CTA) membranes were determined by using mixtures of *o*-nitrophenyl *n*-octyl ether (ONPOE) and quaternary ammonium salts as plasticizers, and pyrazolone derivatives as carriers. The quaternary ammonium salts used were di-*n*-alkyldimethyl- and tetra-*n*-alkylammonium bromides. For some quaternary ammonium salt-carrier systems, the flux increased with an increase in ionic radius of the lanthanide in the range of neodymium to lanthanum. This result is likely due to the contribution of the quaternary ammonium salt to the transport of the lanthanides.

For the present work, the lanthanide fluxes across the CTA membranes were determined by using phospholipids in the place of the above-mentioned quaternary ammonium salts. The phospholipids used were synthetic didecanoyl-, dilauroyl-, and dimyristoyl-L-α-phosphatidylcholines. For brevity, these will be referred to as $C_{10}PC$, $C_{12}PC$, and $C_{14}PC$, respectively. The carriers used were 4-benzoyl-3-methyl-1-phenyl-5-pyrazolone (BMPP) and hinokitiol (β-isopropyltropolone; HIPT). These carriers exhibited high fluxes of the lanthanides for the CTA membranes containing the quaternary ammonium salts (1) or polyoxyethylene *n*-alkyl ethers (2). This paper describes the effect of the phospholipids on the lanthanide flux.

EXPERIMENTAL

Five milliliters of a dichloromethane solution containing 12.5 g/L CTA, 0.2 mL of a dichloromethane solution containing 100 mM of BMPP or HIPT, and 1.0 or 2.0 mL of a dichloromethane solution containing ONPOE and phospholipid were mixed in a glass culture dish (flat bottom, 6 cm diameter). The mixed solution was air-dried for 7 hours, as described previously (3). Then the culture dish containing the resulting membrane was stored at 5°C in a refrigerator. The dichloromethane solution containing ONPOE and phospholipid was prepared as follows: 5.0 mL of a ONPOE solution containing 0.1 M phospholipid were diluted 5 (for $C_{10}PC$ and $C_{12}PC$) or 10 (for $C_{14}PC$) times with dichloromethane in a volumetric flask. The diluted solution was stored at −6°C. The thickness of the membrane obtained was about 0.06 mm. The concentrations of the plasticizer and carrier in the membrane were the same as those for the membranes used in the previous works (1, 2). The concentration of the phospholipid in the plasticizer was 0.1 M. The phospholipids, $C_{10}PC$, $C_{12}PC$, and $C_{14}PC$, were obtained from Sigma Chemical Co. (synthetic materials, product numbers P-9023, 1263, and 6392, respectively). The other reagents were the same as those used in the previous works (1, 2).

The apparatus and technique used for the permeation experiments were the same as those used in the previous works (1, 2): the half volume and effective membrane area of the permeability cell were 32 mL and 7.07 cm^2, respectively. The source phase (Compartment I) initially contained 1.0 mM lanthanide nitrate and 0.1 M sodium acetate buffer of pH 6.1, and the receiving phase (Compartment II) contained 1.0 mM lanthanide nitrate and 0.05 M sulfuric acid. The permeation experiments were performed at 25°C. The lanthanide concentrations in the two compartments after a definite time were determined by the xylenol orange method (4).

RESULTS AND DISCUSSION

Plots of lanthanum concentrations in the two compartments against time for the membranes using the phospholipids, $C_{10}PC$, $C_{12}PC$, and $C_{14}PC$, and BMPP are shown in Fig. 1. The tendencies of the lanthanide concentrations versus time for the other membrane systems were similar to those shown in Fig. 1. The lanthanide fluxes using BMPP and HIPT are shown in Figs. 2 and 3, respectively. In the present paper the lanthanide flux obtained from the permeation experiment is expressed as the mean value in the transport process for 6.5 hours. Further, it is plotted against an ionic radius (5) of the trivalent lanthanide in six-coordination for the reason given in the previous paper (4). The dashed lines in Figs. 2 and 3 represent the lanthanide flux against ionic radius curves for the membranes containing no phospholipid. The data were shown in the previous papers (1, 2).

The flux using BMPP increased with an increase in ionic radius and reached a maximum value at an ionic radius around that of dysprosium (Fig. 2). Subsequently, it decreased and then increased again in the range of praseodymium to lanthanum. However, the variation in the flux de-

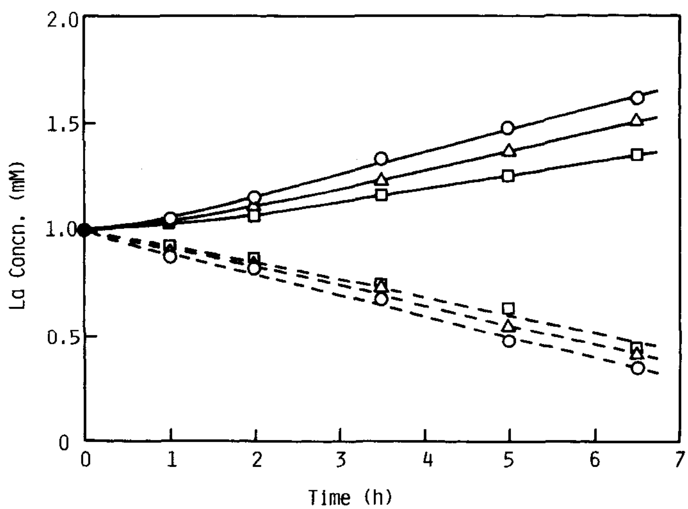

FIG. 1 Plots of lanthanum concentrations in the two compartments against time for the membranes using phospholipids ($C_{10}PC$, $C_{12}PC$, and $C_{14}PC$) and BMPP: (○) $C_{10}PC$, (△) $C_{12}PC$, (□) $C_{14}PC$. The dashed and solid lines represent the concentration against time curves in Compartments I and II, respectively.

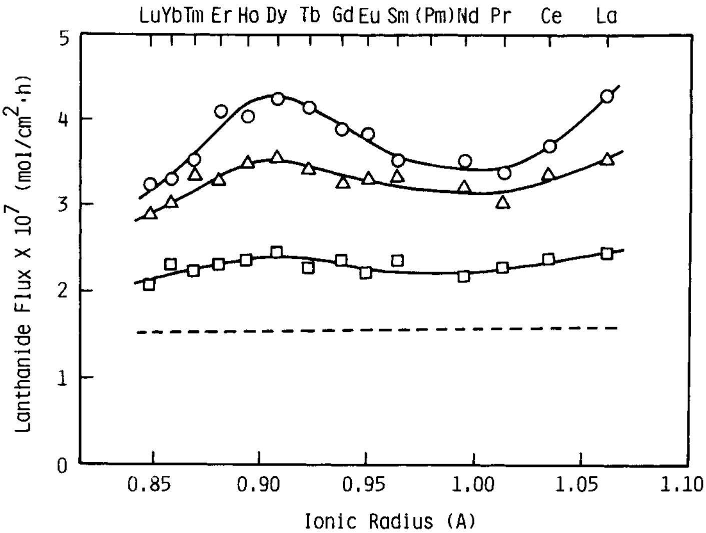

FIG. 2 Lanthanide fluxes using BMPP for the membranes containing $C_{10}PC$ (○), $C_{12}PC$ (△), and $C_{14}PC$ (□). The dashed line represents the flux against the ionic radius curve for the membranes containing no phospholipid. The data were shown in a previous paper (1).

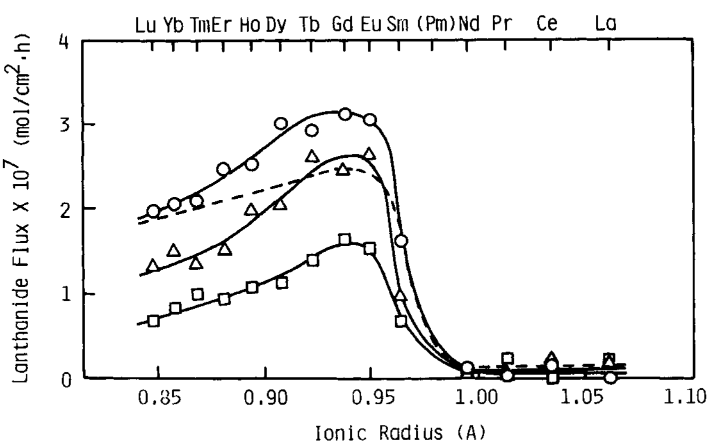

FIG. 3 Lanthanide fluxes using HIPT for the membranes containing $C_{10}PC$ (○), $C_{12}PC$ (△), and $C_{14}PC$ (□). The dashed line represents the flux against the ionic radius curve for the membranes containing no phospholipid. The data were shown in a previous paper (2).

creased with an increase in alkyl chain length of the phospholipid. As shown in a previous paper (1), the flux increased with an increase in ionic radius of the lanthanide in the range of neodymium to lanthanum for the membranes using some quaternary ammonium salts and BMPP. On the other hand, it has been known (6) that the lanthanide nitrates can be extracted by trioctylmethylammonium salt and tributyl phosphate in kerosene. The extractability of the trioctylmethylammonium salt is excellent for the lanthanides having large ionic radii, particularly lanthanum, and that of the tributyl phosphate for the lanthanides near dysprosium. These facts show that the quaternary nitrogen structure and phosphate group in the phospholipid contribute to the transport of the lanthanides. The increase in the alkyl chain length may result in the lowering of these contributions.

The fluxes using HIPT increased appreciably in the range of erbium to europium (Fig. 3), though their values were lower than those obtained for the membranes containing polyoxyethylene *n*-alkyl ethers in the previous work (2). The appreciable increase in the flux is likely due to the contribution of the phosphate group in the phospholipid to the transport of the lanthanides.

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