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Transport of Cesium from Reprocessing Concentrate Solutions through Flat-Sheet-Supported Liquid Membranes: Influence of the Extractant

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ABSTRACT

The influence of the extractant on cesium transport from nuclear fuel reprocessing concentrate solutions to demineralized water through flat-sheet-supported liquid membranes (FSSLMs) was studied using four crown ethers of different lipophility: dibenzo-21-crown-7 (DB21C7), benzo-21-crown-7 (B21C7), *t*-butylbenzo-21-crown-7 (tBuB21C7), and *n*-decylbenzo-21-crown-7 (nDecB21C7). The distribution coefficients of cesium show that the four crown ethers have similar extraction capacities which follow the sequence nDecB21C7 > tBuB21C7 > B21C7 > DB21C7. DB21C7 and B21C7 cannot be used in cesium transport experiments due to the low organic solubility of the former and the great tendency of the latter to form a third phase between the organic phase and the aqueous extraction phase. Of the other two crown ethers, nDecB21C7 leads to faster cesium transport than tBuB21C7 due to the stronger lipophilic character of the former; this allows a higher crown-ether concentration in the membrane to be obtained when nDecB21C7 is used.

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INTRODUCTION

Reprocessing nuclear fuels produces medium-activity liquid wastes which are treated by evaporation. The distillate is discharged into the environment and all active and inactive salts are concentrated, leading to a considerable volume of waste. This concentrate must be stored in geological formations after embedding due to the activity of long-life radionuclides such as actinides, cesium, and strontium. It is desirable to remove these radionuclides from the concentrate in order to significantly decrease the volume of waste to be stored in geological formations. The decontamination of this concentrate can be obtained by coupled transport through supported liquid membranes (SLMs) (1–3).

An SLM is composed of an organic liquid adsorbed onto a microporous support separating two aqueous solutions: the first contains the permeating ions (feed solution) and the second one is free of these ions (stripping solution). The transport of ions through the membrane occurs when a chemical gradient is established between the two aqueous solutions. The use of selective neutral extractants (such as crown ethers, CMPO) as membrane carriers leads to coupled transport across the membrane. When an evaporation concentrate (comprised mainly of nitric acid and sodium nitrate in high concentrations) is used as the feed solution, nitrate ions will be cotransported with the given cation. If a large concentration difference in nitrate ions exists between the two aqueous solutions (e.g., when demineralized water is used as a stripping solution), it is possible to transfer the cations against their concentration gradient. The main advantage of SLMs over traditional separation technologies is the low inventory of the organic phase used; hence, only small amounts of extractant are necessary, and very selective and expensive extractants can be used. Other advantages are lower capital and operating cost, low energy consumption, mass transfer in one step, possibility of achieving high separation factors, and possibility of concentrating the recovered species during separation (4).

We explained in our last paper (5) that crown ethers form very size-selective complexes with alkali and alkali-earth cations and are efficient extraction agents for the decontamination of the concentrate. A comparison between the cavity radius of the crown ethers and the ionic radius of the cations shows that cesium ions will be complexed best with the 21-crown-7 (21C7); this crown ether is too soluble in water. The addition of aliphatic or aromatic substituents (like cyclohexano or benzo groups) increases the hydrophobicity of the macrocycle that makes it usable in solvent extraction processes. Cyclohexano substituents cause a minimal reduction of the complexing ability of the crown ether whereas benzo or other electron-withdrawing substituent groups reduce the macrocycle complexing power (6, 7). However, benzo-substituted crown-ethers produce a stronger bond with alkali metal than with alkaline-earth metals; the more rigid benzo-substituted crown-ethers may maintain a cavity in solution, thus allowing the alkali ions to benefit from the fact that no rearrangement of the coordinating oxygen is necessary. On the contrary, the doubly charged alkaline-earth ions require a configuration that is more easily attained by the flexible cyclohexano crown-ethers than by the rigid benzo crown-ethers (8). Furthermore, Ramadan et al. (9) indicated that DC18C6 is not an efficient extractant for cesium and found distribution coefficients lower than 10^{-4}.

In view of these considerations, the most efficient crown ethers for cesium extraction from the evaporation concentrate are the benzo derivatives of 21-crown-7. This is consistent with the work of Blasius (10) who studied cesium extraction from synthetic medium-activity effluents from reprocessing operations of nuclear fuels using several crown ethers. He found that the 18C6 and 24C8 crown-ether families showed less selectivity from cesium than the 21C7 crown-ether family. In the latter, DB21C7 was found to be a better extractant than DC21C7.

The benzo derivatives of crown ethers show lower solubility in the organic solvent than cyclohexano derivatives (11). This solubility can be increased by adding aliphatic groups to the aromatic groups of the crown ether which become more lipophilic. Thus, di-*tert*-butylbenzo-21-crown-7 (DtBuB21C7) was used for cesium extraction with organic diluents of low polarity such as toluene, which cannot solubilize the DB21C7 (12). Also, these more lipophilic crown ethers show higher cation transport through SLMs (due to a higher carrier concentration in the membrane) and form more stable SLMs (due to less carrier washing by the aqueous solutions). Thus, the use of benzo derivatives of crown ethers with long aliphatic chains, such as the *n*-decyl or *t*-decyl groups, on the aromatic rings shows a strong increase of the extractant's distribution coefficient (13).

For the present work we studied the influence of extractant lipophility on cesium transport through SLMs from a given concentrate of evaporation to demineralized water by using several crown ethers of the benzo derivatives of the 21C7 family. We used dibenzo-21-crown-7 (DB21C7) and three specially synthetized monobenzo derivatives of 21C7: benzo-21-crown-7 (B21C7), *tert*-butylbenzo-21-crown-7 (tBuB21C7), and *n*-decylbenzo-21-crown-7 (nDecB21C7). It is better to synthesize monobenzo derivatives than dibenzo derivatives in view of the higher yields obtained with the former.

First, we determined the solvent solubility of these four crown ethers as well as the distribution coefficients of cesium and the extractant between the organic phase and the aqueous phases; and finally, we carried out cesium transport through flat-sheet-supported liquid membranes (FSSLMs). This type of SLM was used because it is easier to handle.

EXPERIMENTAL

Reagents

The synthetic concentrate was prepared according to the average composition of the evaporation concentrate from medium-activity liquid wastes from reprocessing operations of nuclear fuels (see Table 1). Before use, the solution was filtered to remove insolubles. All the reagents used in this solution, as well as in the other aqueous solutions used as the feed solution, were analytical-grade products from Prolabo. All these aqueous solutions were doped with ^{137}Cs. The ^{137}Cs source used was a solution of CsCl in HCl (1 mol·L^{-1}) from Amersham.

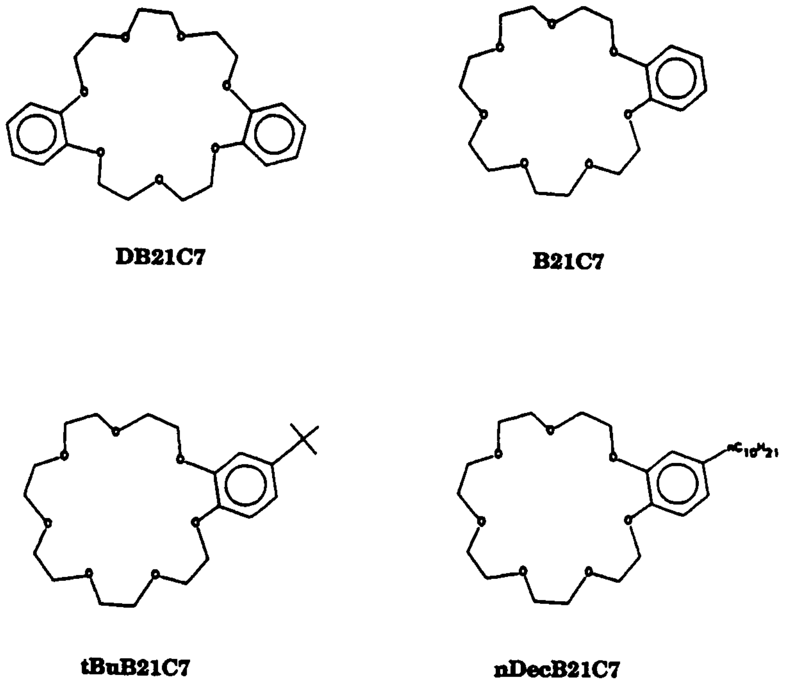
The aqueous stripping solution was demineralized water purified with MilliQ from Millipore.

DB21C7 was obtained from Parish with a purity of 98%. B21C7, tBuB21C7, and nDecB21C7 were synthesized by Chimie+ (Marseille) with a purity of 95% in all cases (Fig. 1).

The *n*-hexylbenzene used as a diluent was obtained from Aldrich, and the isotridecanol, used as a phase modifier, is an Exxon product.

TABLE 1
Composition of the Synthetic Concentrate

Compound	Concentration (g/L)
HNO_3	63
$NaNO_3$	290
$Mg(NO_3)_2 \cdot 6H_2O$	158
$Ca(NO_3)_2 \cdot 4H_2O$	3.5
$Fe(NO_3)_3 \cdot 9H_2O$	1.1
NaCl	0.7
NaF	0.07
$Na_2O \cdot SiO_2 \cdot 5H_2O$	0.2
$Na_3PO_4 \cdot 12H_2O$	3.4
$Na_2SO_4 \cdot 10H_2O$	3.0
NH_4NO_3	8.0
Tri(*n*-butyl)phosphate	0.15
$UO_2(NO_3)_2 \cdot 6H_2O$	4.7



DB21C7 **B21C7**

tBuB21C7 **nDecB21C7**

FIG. 1 Chemical structure of dibenzo-21-crown-7 (DB21C7) (a); benzo-21-crown-7 (B21C7) (b); t-butylbenzo-21-crown-7 (tBuB21C7) (c); and *n*-decylbenzo-21-crown-7 (nDecB21C7) (d).

Membranes

The SLMs were prepared by soaking the polymeric support (Celgard 2500) for almost 24 hours in each of the organic solutions used and then leaving it to drip for a few seconds before being placed in the transport cell.

Celgard 2500 (Celanese Separation Products, Charlotte, North Carolina, USA) is a 2.5×10^{-3} cm thick microporous polypropylene film with a nominal porosity of 45%, an effective pore size of 0.04 μm, a pore tortuosity of 2, and a critical surface tension of 35 $\mathrm{mN \cdot m^{-1}}$.

Determination of Cesium Distribution Coefficients

The distribution coefficients of cesium, D_{Cs}, defined as the equilibrium ratio of the cesium species between the organic and the aqueous phases, were determined at room temperature (25°C) by mixing the same volume of each phase at 100 rpm in a polypropylene test tube and then measuring the cesium count in each phase by gamma spectrometry analyses of ^{137}Cs.

The determination of D_{Cs} in extraction experiments was performed by mixing 10 mL of the organic phase and 10 mL of the aqueous feed solution. To determine D_{Cs} in stripping, 8 mL of the last organic phase were mixed with 8 mL of the aqueous stripping solution.

Duplicate runs of each experiment were routinely performed.

Determination of Crown-Ether Distribution Coefficients

The distribution coefficients of the four crown ethers between the organic and the aqueous phases (D_E) were determined using the method proposed by Horwitz (18) in order to measure the crown-ether concentration in the aqueous phase after partitioning. Eight milliliters of the organic phase which contained various crown-ether concentrations ($[E]_0$) was mixed with 40 mL of the aqueous phase for 30 minutes at room temperature (25°C). Thus, the crown ether was partitioned between both phases with a distribution coefficient D_1. After centrifugation, 35 mL of the aqueous phase was mixed, in the same conditions as above, with 7 mL of diluent. Thus, the crown ether was partitioned between both phases with a distribution coefficient D_2. After centrifugation, 6 mL of the organic phase were mixed with 6 mL of the synthetic concentrate which contained ^{137}Cs; the cesium distribution coefficient (D_{Cs}) was determined following the method indicated above. Since we knew the relation between D_{Cs} and the initial crown-ether concentration in the organic phase, the crown-ether concentration in the second organic phase ($[E]_2$) was determined. Taking into account the two equilibriums and considering that $D_1 = D_2 = D_E$, we obtain the following expression:

$$[E]_2 = \frac{D_E R}{(D_E + R)^2}[E]_0 \tag{1}$$

where R is the volume ratio of the phases (V_{aq}/V_{org}). In our case $R = 5$; thus, D_E can be determined.

Cesium Transport through FSSLM

The transport experiments were carried out in a permeation cell comprised of two papallelepiped compartments made of Persplex thermostated at 25 ± 0.1°C. The diagram of this cell is shown in Fig. 2. The membrane area was 29.37 cm^2 (rectangular), and the volumes of feed and stripping solutions were identical (125 mL).

At the beginning of each experiment, the SLM was inserted between the two compartments and the cell was tightly clamped with two Teflon

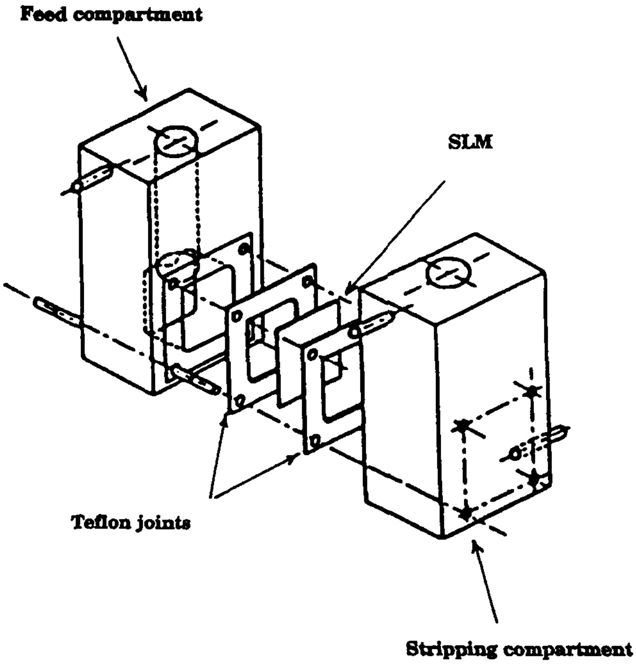


FIG. 2 Exploded diagram of the transport cell used.

joints with screws. Time $t = 0$ was taken as the moment when the stirring of aqueous phases was started (with magnet stirrers). The stirring speed was 500 rpm in the feed solution, which allows the minimum thickness of the aqueous feed boundary layer to be obtained. The permeation of cesium across the membrane was measured by periodic sampling of both aqueous solutions. The cesium counts in each phase were determined by gamma spectrometry analyses. The pH of the stripping solution was also monitored regularly.

Cesium permeability through the SLM was determined by plotting ln C/C_0 versus time for the first 6 or 7 hours. In the transport model proposed by P. R. Danesi (4), permeability is defined by

$$\ln C/C_0 = -(\epsilon S/V)P_{Cs}t \tag{2}$$

where C = feed concentration of cesium at time t, C_0 = feed concentration of cesium at time 0, S = membrane area (cm^2), V = volume of feed solution (cm^3), ϵ = support porosity, t = permeation time (h), and P_{Cs} = cesium permeability (cm h^{-1}).

Cesium Measurement by Gamma Spectrometry

Cesium counts where determined by gamma spectrometry analyses of ^{137}Cs by using a detection chain from Intertechnique, equipped with Ge detectors. The counting time was always long enough to obtain a relative error of less than 5% for the activity measurements.

RESULTS AND DISCUSSION

Organic Solubility of the Extractants

First, the organic solubility of the four crown ethers was measured and, simultaneously, the possible formation of a third phase between the organic phase and the synthetic concentrate was observed when both solutions were mixed. *n*-Hexylbenzene was used as the diluent, with isotridecanol (1.5 mol·L^{-1}) as the phase modifier. The results are shown in Table 2. We can see that DB21C7 solubility is slightly lower than that of the monobenzo derivatives of 21C7. This lower solubility could be due to the existence of two benzo groups on the ring which cause stronger dipolar interactions between the molecules. This is consistent with organic solubility experiments carried out in our laboratory with the dibenzo-24-crown-8 (DB24C8), in which we found only very polar or very protic diluents can dissolve this crown ether at high concentrations (14).

When B21C7 was used with a concentration higher than 0.05 mol·L^{-1}, a third phase between the organic and the aqueous phases appeared. On the other hand, tBuB21C7 and nDecB21C7 can be used with *n*-hexylbenzene (1.5 mol·L^{-1} isotridecanol) at high concentrations (>0.5 mol·L^{-1}).

Distribution Coefficients of Cesium

The distribution coefficients of cesium (D_{Cs}) at various crown-ether concentrations in the organic phase were determined using the four crown ethers of interest; *n*-hexylbenzene (1.5 mol·L^{-1} isotridecanol) was used

TABLE 2
Organic Solubility of Crown Ethers in *n*-Hexylbenzene (0.5 mol/L isotridecanol)

Crown ether	Solubility (mol/L)
DB21C7	0.025 to 0.05
B21C7	>0.5
tBuB21C7	>0.5
nDecB21C7	>0.5

as the diluent and the synthetic concentrate as the extraction aqueous phase. The results obtained are shown in Fig. 3. We observe that in all cases the slope of the straight line of log D_{Cs} versus log$[E]_0$ has a value near 1. This agrees with the 1:1 stoichiometry for the cesium nitrate/crown ether complexes.

For the four crown ethers, the values of the cesium distribution coefficient were very similar in the concentration range studied. In all cases, for a given concentration of crown ether, we can observe the following sequence of distribution coefficients: nDecB21C7 > tBuB21C7 > B21C7 > DB21C7. The D_{Cs} values obtained were 10 times lower than those of D_{Sr} obtained with DC18C6 and DtBuC18C6 as extractants (5).

On the other hand, no plateau of the D_{Cs} values at high crown-ether

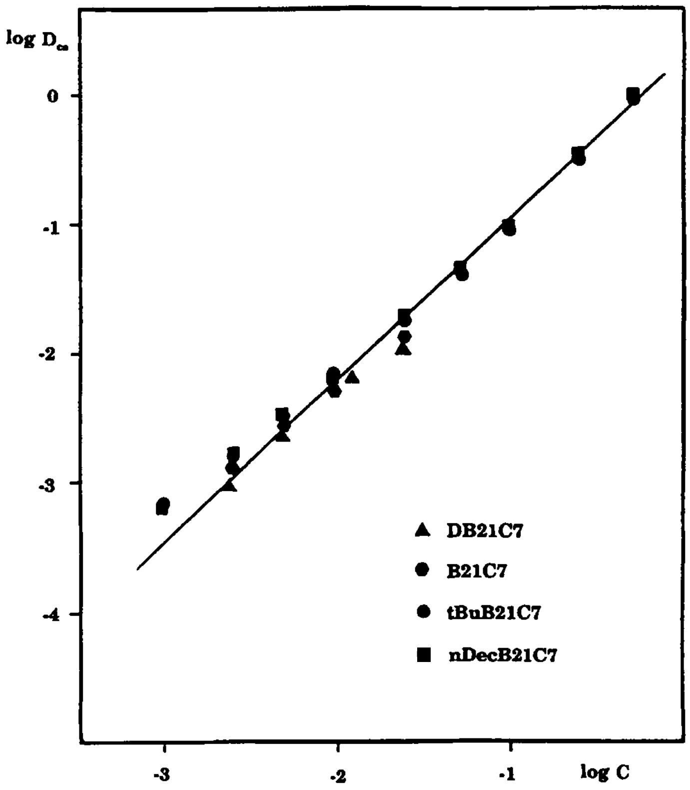


FIG. 3 Distribution coefficients of cesium versus initial crown-ether concentration in the organic phase for DB21C7, B21C7, tBuB21C7, and nDecB21C7. Diluent: *n*-hexylbenzene (1.5 mol·L^{-1} isotridecanol). Aqueous solution: synthetic concentrate.

concentrations due to associations between extractant molecules was observed. Similar results were obtained with strontium extraction experiments carried out with DC18C6 and DtBuC18C6 (5).

We also carried out stripping experiments using tBuB21C7 and nDecB21C7 as extractants at an organic concentration of 0.5 mol·L^{-1}. The D_{Cs} values obtained were 0.64 for tBuB21C7 and 0.49 for nDecB21C7. These values are relatively high and similar to the extraction values.

Distribution Coefficients of the Crown Ethers

The distribution coefficients of the four crown ethers studied were determined by a method which uses the radiometric determination of ^{137}Cs and allows the use of acid aqueous solutions. Thus, three different aqueous phases were used: demineralized water, an aqueous solution composed of HNO_3 (1 mol·L^{-1}), and another one composed of $LiNO_3$ (5.8 mol·L^{-1}) and HNO_3 (1 mol·L^{-1}) (the latter solution has an ionic strength similar to the synthetic concentrate but sodium was replaced by lithium as this cation is practically not complexed by these crown ethers, unlike sodium).

All the results obtained are shown in Table 3. We can observe the following sequence for the distribution coefficients of the crown ethers for any aqueous phase: nDecB21C7 > tBuB21C7 > DB21C7 > B21C7 [when the aqueous phase was composed of HNO_3 (1 mol·L^{-1}) and $LiNO_3$ (5.8 mol·L^{-1}), the distribution coefficient of DB21C7 was similar to that of the B21C7]. Thus, there is strong dependence between the lipophilic character of the crown ether and its distribution coefficient.

We also noted that the influence of the aqueous solution composition was relatively weak in comparison with the effect of crown-ether lipophility.

Cesium Transport through FSSLMs

Cesium transport through an FSSLM was carried out using tBuB21C7 and nDecB21C7 as carriers at a concentration of 0.5 mol·L^{-1}. The feed

TABLE 3

Distribution Coefficients of Crown Ethers between the Aqueous Phase and Organic Phase Composed of *n*-Hexylbenzene (1.5 mol/L isotridecanol)

Aqueous phase	DB21C7	B21C7	tBuB21C7	nDecB21C7
Demineralized water	41	11	65	160
Nitric acid (1 mol/L)	47	23	56	230
Lithium nitrate (5.8 mol/L) + nitric acid (1 mol/L)	56	58	130	150

solution was the synthetic concentrate, the stripping solution was demineralized water, and *n*-hexylbenzene (1.5 mol·L^{-1} isotridecanol) was used as the diluent. In both cases we determined the cesium permeability of the membrane as shown in Fig. 4. Under our own conditions, cesium permeability (P_{Cs}) can be expressed by the following equation (4):

$$P_{Cs} = \frac{D_{Cs}}{D_{Cs}\Delta_a + \Delta_0} \tag{3}$$

The transport resistance due to diffusion by the aqueous feed boundary (Δ_a) has a value of 0.66 h·cm^{-1}, obtained in previous experiments (15). The transport resistance due to diffusion by the membrane (Δ_0) can be expressed as follows:

$$\Delta_0 = \tau d_0/D_0 \tag{4}$$

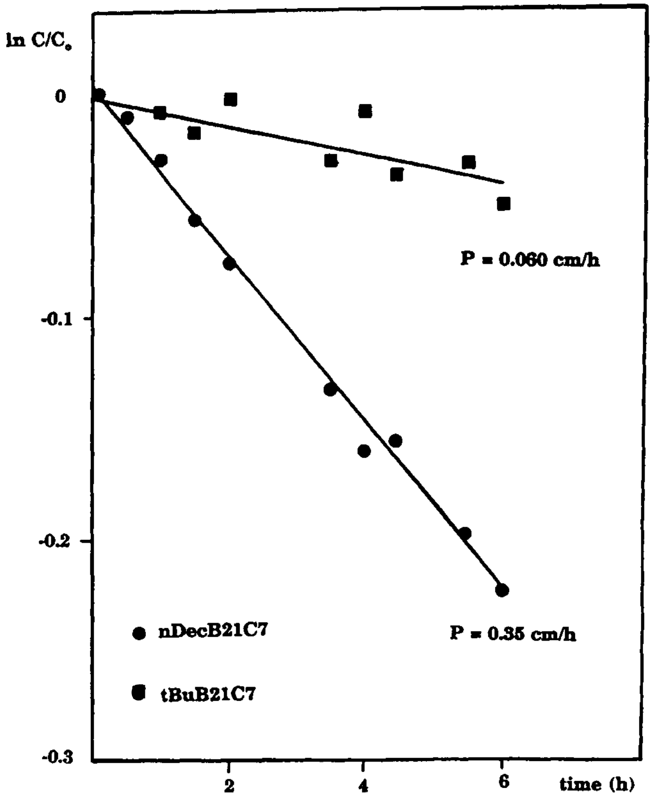


FIG. 4 Cesium permeability of the SLM. Crown-ether concentration: 0.5 mol·L^{-1}. Organic diluent: *n*-hexylbenzene (1.5 mol·L^{-1} isotridecanol). Aqueous feed solution: synthetic concentrate. Aqueous stripping solution: demineralized water.

where τ = tortuosity of the support pores
d_0 = thickness of the membrane
D_0 = membrane diffusion coefficient of the cesium-containing species.

The value of the support parameters, τ and d_0, are known (2 and 25×10^{-4} cm, respectively). The D_0 value can be evaluated by the Wilkie and Chang equation (16):

$$D_0 = \frac{7.4 \times 10^{-8} X^{0.5} T M^{0.5} 3600}{\mu V^{0.6}} \tag{5}$$

where X = association parameter of the diluent (1)
T = absolute temperature (298 K)
M = molecular weight of the diluent (167.45 g·mol^{-1})
μ = dynamic viscosity of the diluent (1.95 cP)

The only parameter of Δ_0 which depends on the extractant nature is the molar volume of the extractant (V). In our case, V values of 468.7 cm^3·mol^{-1} for tBuB21C7 and 605.4 cm^3·mol^{-1} for nDecB21C7 were evaluated using the group contribution method of Le Bas (17). Thus, we found D_0 values of 0.57 and 0.66 cm^2·s^{-1} for tBuB21C7 and nDecB21C7, respectively. The values of Δ_0 can be calculated from Eq. (4) and were found to be 8.7×10^{-3} and 7.5×10^{-3} h·cm^{-1} for tBuB21C7 and nDecB21C7, respectively.

In order to evaluate cesium permeability, we needed to know the distribution coefficient of cesium between the membrane and the aqueous feed solution; therefore, it was necessary to know the real organic concentration of the crown ether in the SLM which could be evaluated by using the relationship between D_{Cs} and the initial organic concentration of the extractant previously found. The determination of the crown-ether concentration in the membrane was achieved by using the volume ratio between the membrane and the aqueous solutions (R = 1920); the membrane volume was measured by weight, and the distribution coefficients of the crown ethers between n-hexylbenzene (1.5 mol·L^{-1} isotridecanol) and the two aqueous solutions previously determined (for each crown ether we used the average value of all the values obtained). The D_{Cs} evaluated were 0.021 and 0.037 for tBuB21C7 and nDecB21C7, respectively.

Finally, as D_{Cs}, Δ_a, and Δ_0 values were known, and rearranging Eq. (3), the following expression can be obtained:

$$\frac{1}{P} = \Delta_a + \frac{\Delta_0}{D_{Cs}} \tag{6}$$

From Eq. (6) the contribution to the transport resistance due to diffusion

TABLE 4
Evaluated and Experimental Cesium Permeability of the Membrane with tBuB21C7 and nDecB21C7 as Extractants

Crown ether	Evaluated Cs permeability (cm/h)	Experimental Cs permeability (cm/h)
BuB21C7	0.036	0.060
nDecB21C7	0.054	0.35

by the aqueous feed boundary layer can be evaluated. As $\Delta_a = 0.66$ h·cm^{-1} and Δ_0/D_{Cs} are 0.42 and 0.20 h·cm^{-1} for tBuB21C7 and nDecB21C7, respectively, it can be seen that the contribution to the overall resistance to mass transfer is not negligible.

The cesium permeability of the membrane was evaluated from Eq. (3). The results obtained and the experimental values are shown in Table 4. Although nDecB21C7 leads to a slightly lower complex diffusion coefficient than tBuB21C7, the former extractant allows faster transport of cesium due to the higher values of D_E that lead to a greater crown-ether concentration in the membrane. Furthermore, the slightly stronger extractant power of nDecB21C7 as compared to that of tBuB21C7 must contribute to this faster cesium transport.

The differences between the evaluated and experimental values could be due to the uncertainty in the evaluation of the coefficients in the membrane and the errors in the determination of the D_E and R values.

The cesium permeabilities of the membrane obtained were lower than those of strontium (5) due to a D_{Cs} that is much lower than the D_{Sr}. We can see that cesium transport at the end of the experiment (193 hours) was 30 and 40% for tBuB21C7 and nDecB21C7 as membrane carriers, respectively. These values are much lower than those for strontium transport found at a similar cation permeability (5, 15). This different behavior could be due to the D_{Cs} values for the stripping processes, which are very high and much closer to the extraction values than those of D_{Sr}. Thus, cesium transport stops before that of strontium.

CONCLUSIONS

The four crown ethers studied have more or less the same cesium extractant power: DB21C7 cannot be used due to its low solubility in *n*-hexylbenzene (1.5 mol·L^{-1} isotridecanol). We can assume the same for the majority of the diluents because a relatively high concentration of the extractant has to be obtained in the membrane in order to obtain good cesium transport. On the other hand, monobenzo derivatives of 21C7 can easily be solubilized in organic diluents. Among these compounds, B21C7 cannot be used due to a strong tendency to form a third phase between the organic phase and the synthetic concentrate.

Faster cesium transport through the SLM can be obtained with nDecB21C7 as the extractant than with tBuB21C7. This is mainly due to the greater lipophilic character of nDecB21C7.

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