

This article was downloaded by:

On: 25 January 2011

Access details: *Access Details: Free Access*

Publisher *Taylor & Francis*

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Separation Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713708471>

Influence of Membrane Solvent on Strontium Transport from Reprocessing Concentrate Solutions through Flat-Sheet-Supported Liquid Membranes

J. F. Dozol^a; J. Casas^a; A. M. Sastre^b

^a Commissariat a L'energie Atomique Centre D'etudes Nucleaires De Cadarache, Durance, France ^b

Department of Chemical Engineering, Universitat Politècnica De Catalunya E.T.S.E.I.B., Barcelona, Spain

To cite this Article Dozol, J. F. , Casas, J. and Sastre, A. M.(1993) 'Influence of Membrane Solvent on Strontium Transport from Reprocessing Concentrate Solutions through Flat-Sheet-Supported Liquid Membranes', *Separation Science and Technology*, 28: 11, 2007 — 2022

To link to this Article: DOI: [10.1080/01496399308016730](https://doi.org/10.1080/01496399308016730)

URL: <http://dx.doi.org/10.1080/01496399308016730>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Influence of Membrane Solvent on Strontium Transport from Reprocessing Concentrate Solutions through Flat-Sheet-Supported Liquid Membranes

J. F. DOZOL and J. CASAS

COMMISSARIAT A L'ENERGIE ATOMIQUE
CENTRE D'ETUDES NUCLEAIRES DE CADARACHE
13108 SAINT PAUL LEZ DURANCE, FRANCE

A. M. SASTRE

DEPARTMENT OF CHEMICAL ENGINEERING
UNIVERSITAT POLITÈCNICA DE CATALUNYA
E.T.S.E.I.B., DIAGONAL 647, E-08028 BARCELONA, SPAIN

ABSTRACT

The influence of membrane solvents on strontium transport from nuclear fuel reprocessing concentrate solutions to demineralized water through a flat-sheet-supported liquid membrane has been studied using dicyclohexano-18-crown-6 as the extractant and Celgard 2500 as the solid support. Even though the highest values of the distribution coefficients of strontium were obtained with nitrated compounds as membrane solvents, strontium permeabilities were determined only when a membrane solvent was used for which stable SLMs were obtained. Among the latter, the use of 4-nonylphenol as a phase modifier is not satisfactory for long-term strontium transport experiments due to its reactivity with the nitric acid of the aqueous feed solution. We achieved a good correlation between strontium permeability and two parameters of the membrane diffusion coefficient (molecular weight and viscosity of the membrane solvent) for aromatic solvents modified with isotridecanol or 1-decanol. The best results were obtained with *n*-hexylbenzene (0.7 mol·L⁻¹ isotridecanol) which should lead to a high strontium decontamination by hollow-fiber-supported liquid membranes. The transport of nitric acid and nonradioactive cations through the membrane was not greatly influenced by the membrane solvent used.

INTRODUCTION

Nuclear fuel reprocessing operations produce medium activity liquid wastes which are treated by evaporation. The distillate is discharged into

2007

the environment and all active and inactive salts are concentrated, leading to a large volume of wastes. This concentrate must be stored in geological formations after embedding, due to the activity of long-life radionuclides (actinides, cesium, strontium). It is desirable to remove these radionuclides from the concentrate in order to decrease sharply the volume of wastes to be stored in geological formations. The decontamination of this concentration can be obtained by coupled-transport through supported liquid membranes (SLM) (1–3).

An SLM is comprised of an organic liquid adsorbed onto a microporous support separating two aqueous solutions: the first contains the permeating ions (feed solution), the second is free of these ions (stripping solution). 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 a concentrate from evaporation (comprised mainly of nitric acid and sodium nitrate in high concentrations) is used as a 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), the cations can be transferred 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 needed, and very selective and expensive extractants can be used. Other advantages are lower capital and operating costs, low energy consumption, mass transfer in one step, the possibility of achieving high separation factors, and the possibility of concentrating the recovered species during separation (4).

The membrane solvent plays an important role in coupled transport through SLMs. On the one hand, the low volume of organic phase in the pores of the support forces the solvent to have certain properties which allow high membrane lifetimes (stable SLMs) to be obtained (5). On the other hand, the membrane solvent must solvate the nitrate anions well in order to obtain high distribution coefficients of the cations between the organic phase and the aqueous feed solution (6). However, it must not transfer the nitric acid through the membrane by proton interaction because the build-up of nitrate ions in the stripping solution decreases the driving force and the permeation of cations slows down and may even stop. A solvent of low basicity should be used to minimize nitric acid transfer through the membrane (2).

In the present work we have studied the influence of the membrane solvent on strontium transport through SLMs from a given concentrate

from an evaporator to demineralized water, using dicyclohexano-18-crown-6 (DC18C6) as the membrane carrier (representative crown-ether for strontium extraction) and Celgard 2500 as the solid support. Following a bibliography survey, several organic solvents with low mutual solubility with water and a high boiling point (in order to obtain the most stable SLMs possible) were chosen. These solvents have to solubilize the extractant in high concentrations ($0.5 \text{ mol}\cdot\text{L}^{-1}$ in DC18C6) in order to obtain effective strontium transport through the SLM (7). In some cases it was necessary to add a phase modifier to the organic solution to avoid the formation of a third phase in contact with the aqueous feed solution. These phase modifiers are polar compounds [in order to increase the complex solubility in the solvent which thus prevents the appearance of the third phase (8)] with a strong lipophilic character (to achieve a stable SLM).

We determined the distribution coefficients of strontium as a function of the solvent or the solvent/phase-modifier mixture used. Then we studied the influence of the membrane solvent on strontium transport through flat-sheet-supported liquid membranes (FSSLMs). This type of SLM was used for its easier handling.

EXPERIMENTAL

Reagents

The organic solvents and phase modifiers used are listed in Table 1. All the compounds, except isotridecanol, were obtained from Aldrich, with a purity of between 97 and 99%. Isotridecanol was an Exxon product.

The aqueous feed solution used was prepared according to the average composition of the evaporation concentrate of medium-activity liquid wastes from nuclear fuel reprocessing operations (see Table 2). All the reagents used in the solution were analytical grade products from Prolabo. The solution was filtered before use so as to remove insolubles. The solution was usually spiked with ^{85}Sr . The source of the ^{85}Sr used was an aqueous solution of SrCl_2 from Amersham.

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

Dicyclohexano-18-crown-6 (DC18C6) (see Fig. 1) was a mixture of cis-syn-cis and cis-anti-cis isomers, with a purity of 99%, and was an Aldrich product.

The organic solutions were prepared by dissolution of DC18C6 ($0.5 \text{ mol}\cdot\text{L}^{-1}$) in solvents or solvent/phase-modifier mixtures.

TABLE 1
Organic Solvents and Phase Modifiers Used as
Membrane Solvents

Organic solvent	Phase modifier
<i>n</i> -Dc	Cycloheptanone 2-Undecanone
<i>p</i> -Diisopropylbenzene	
<i>n</i> -Butylbenzene	4-Nonylphenol
<i>n</i> -Hexylbenzene	1-Decanol
<i>n</i> -Heptylbenzene	Isotridecanol
<i>n</i> -Octylbenzene	2-Ethyl-1-hexanol
<i>n</i> -Decylbenzene	Alcohol benzylic
<i>n</i> -Dodecylbenzene	1-Octanol
<i>n</i> -Tridecylbenzene	2-Octanol
2-Ethyl-1-hexanol	2-Nitro- <i>p</i> -cymene
Alcohol phenetyl	4-Nitro- <i>m</i> -xylene
1-Decanol	
Isotridecanol	Nonanoic acid
2-Nitropropane	Trichloroethylphosphate
1-Nitrohexane	
Nitrocyclohexane	
4-Nitro- <i>m</i> -xylene	
2-Nitro- <i>p</i> -cymene	
1,6-Dibromohexane	
1,2,4-Trichlorobenzene	

TABLE 2
Composition of the Synthetic Concentrate

Compound	Concentration (g/L)
HNO ₃	63
NaNO ₃	290
Mg(NO ₃) ₂ ·6H ₂ O	158
Ca(NO ₃) ₂ ·4H ₂ O	3.5
Fe(NO ₃) ₃ ·9H ₂ O	1.1
NaCl	0.7
NaF	0.07
Na ₂ O·SiO ₂ ·5H ₂ O	0.2
Na ₃ PO ₄ ·12H ₂ O	3.4
Na ₂ SO ₄ ·10H ₂ O	3.0
NH ₄ NO ₃	8.0
Tri(<i>n</i> -butyl)phosphate	0.15
UO ₂ (NO ₃) ₂ ·6H ₂ O	4.7

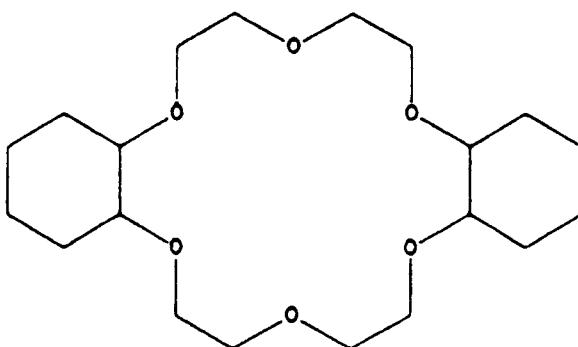


FIG. 1 Chemical structure of dicyclohexano-18-crown-6 (DC18C6).

Membranes

The SLMs were prepared by soaking the polymeric support (Celgard 2500) for almost 24 hours in each organic solution used, then letting it drip for a few seconds before placing it 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 $\text{mN}\cdot\text{m}^{-2}$.

Determination of Strontium Distribution Coefficients

The distribution coefficients of strontium, D_{Sr} , defined as the equilibrium ratio of the strontium 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 strontium count in each phase by gamma spectrometry analyses of ^{85}Sr .

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

Duplicate runs of each experiment were routinely performed.

Strontium Transport through FSSL

The transport experiments were carried out in a permeation cell comprising two parallelepiped compartments made of Perspex, thermostated

at $25 \pm 0.1^\circ\text{C}$. The design of this cell is shown in Fig. 2. The membrane area was 29.37 cm^2 (rectangular), and the volumes of the 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 joints with screws. Time $t = 0$ was taken as the moment when stirring of the aqueous phases was started (with magnet stirrers). The permeation of strontium across the membrane was measured by periodic sampling of both aqueous solutions. The strontium counts in each phase were determined by gamma spectrometry analyses. The pH of the stripping solution was also monitored regularly.

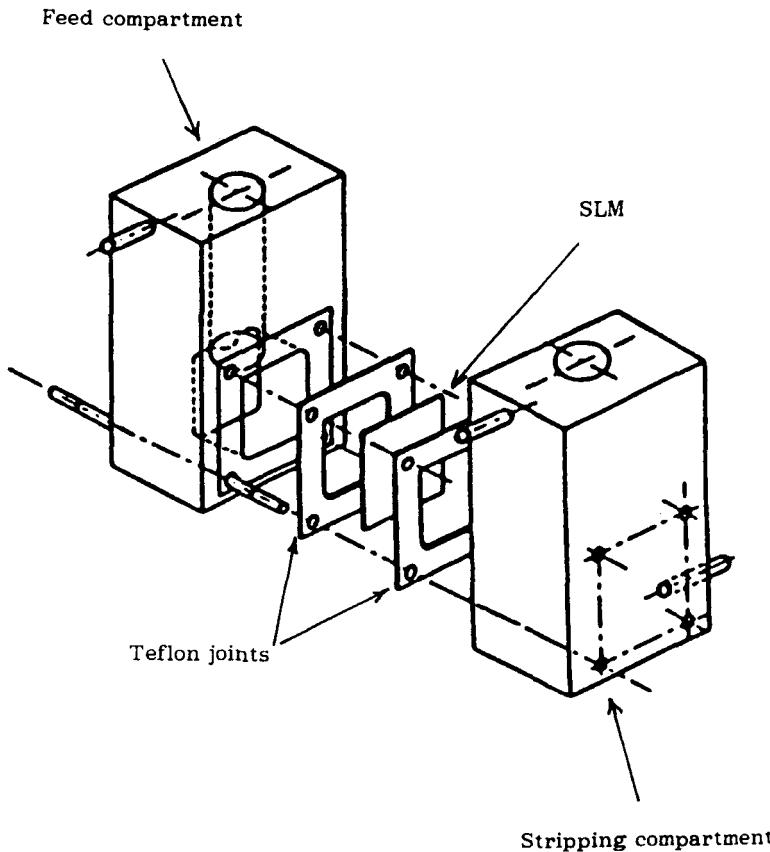


FIG. 2 Exploded diagram of the transport cell used.

Strontium 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 Danesi (4), permeability is defined by

$$\ln C/C_0 = -(\epsilon S/V)P_{\text{Sr}}t \quad (1)$$

where C = feed concentration of strontium at time t , C_0 = feed concentration of strontium at time 0, S = membrane surface (cm^2), V = volume of feed solution (cm^3), ϵ = support porosity, t = permeation time (hour), and P_{Sr} = strontium permeability ($\text{cm} \cdot \text{h}^{-1}$).

Strontium Measurements by Gamma Spectrometry

Strontium counts were determined by gamma spectrometry analysis of ^{85}Sr by using a detection chain from Intertechnique, equipped with Ge detectors.

The counting time was always sufficiently high so as to obtain under 5% relative error in the activity measurements.

Cations Transport through FSSLM

Several nonradioactive cations initially present in the aqueous feed solution (sodium, calcium, magnesium, and iron) were measured in the aqueous stripping solution at the end of the experiment by atomic absorption using a flame atomic absorption spectrophotometer (Philips PU 9200X).

RESULTS AND DISCUSSION

Solubility of DC18C6 in the Solvents

The solubility of DC18C6 in all the solvents chosen was always found to be greater than $0.5 \text{ mol} \cdot \text{L}^{-1}$ except for *n*-dodecane. This solvent was therefore not used in the experiments that followed.

Phase Modifiers

We mixed the organic solution of DC18C6 ($0.5 \text{ mol} \cdot \text{L}^{-1}$) in the different solvents with the aqueous feed solution, and we found a third phase between the two phases when the membrane solvent was an aromatic compound because of the poor solubilizing properties of this type of solvent.

A number of phase modifiers were added to the aromatic solvents. Only when long-chain alcohols were used was it possible to obtain high SLM lifetimes. This is the case for 4-nonylphenol, 1-decanol, and isotridecanol. Table 3 shows the solvent/phase-modifier mixtures ($0.5 \text{ mol} \cdot \text{L}^{-1}$ DC18C6) chosen.

TABLE 3
Organic Solvent/Phase Modifier Mixtures Used as
Membrane Solvents

Organic solvent	Phase modifier
<i>p</i> -Diisopropylbenzene	4-Nonylphenol (0.6 M)
<i>p</i> -Diisopropylbenzene	Isotridecanol (0.9 M)
<i>n</i> -Butylbenzene	4-Nonylphenol (0.4 M)
<i>n</i> -Hexylbenzene	4-Nonylphenol (0.5 M)
<i>n</i> -Hexylbenzene	1-Decanol (0.7 M)
<i>n</i> -Hexylbenzene	Isotridecanol (0.7 M)
<i>n</i> -Heptylbenzene	Isotridecanol (0.8 M)
<i>n</i> -Octylbenzene	Isotridecanol (0.9 M)
<i>n</i> -Decylbenzene	Isotridecanol (1.0 M)
<i>n</i> -Dodecylbenzene	Isotridecanol (1.1 M)
<i>n</i> -Tridecylbenzene	Isotridecanol (1.3 M)

Strontium Distribution Coefficients

Strontium distribution coefficients (D_{Sr}) were determined in extraction and stripping operations using the various organic phases studied. The results obtained are shown in Table 4.

We can see that D_{Sr} for the extraction operation was usually greater than 10 for all the organic phases tested. The highest values were found in the case of the nitro compounds due to their high polarity, and therefore the high solvation of nitrate ions, with observation of the following sequence: 2-nitropropane $>$ nitrocyclohexane $>$ 1-nitrohexane $>$ 4-nitro-*m*-xylene $>$ 2-nitro-*p*-cymene.

The values of D_{Sr} with 4-nonylphenol as a phase modifier were found to be greater than those with 1-decanol or isotridecanol due to the better solvation of the nitrate anions due to the greater protic properties of the phenols.

In stripping operations, the values of D_{Sr} were usually lower than 1. Therefore, good stripping conditions should be found with demineralized water as in the stripping phase of the strontium transport experiments.

Strontium Transport through FSSLM

We performed experiments on strontium transport through FSSLM with the various organic phases studied. Only isotridecanol or aromatic solvents with aliphatic groups of six or more carbon atoms, modified with long-chain alcohols, lead to a stable FSSLM for periods longer than 200 hours (9). In these cases (and with 1-decanol as a membrane solvent, even though the membrane was only stable for 48 hours) we determined

TABLE 4
Strontium Distribution Coefficients between the Aqueous Feed Solution and the Organic Phase (extraction) and between the Aqueous Stripping Solution and the Organic Phase (stripping)^a

Solvent	Extraction	Stripping
<i>p</i> -Diisopropylbenzene (0.6 M 4-nonylphenol)	17	0.15
<i>p</i> -Diisopropylbenzene (0.9 M isotridecanol)	11	0.032
<i>n</i> -Butylbenzene (0.4 M 4-nonylphenol)	19	0.14
<i>n</i> -Hexylbenzene (0.5 M 4-nonylphenol)	17	0.16
<i>n</i> -Hexylbenzene (0.7 M 1-decanol)	13	0.039
<i>n</i> -Hexylbenzene (0.7 M isotridecanol)	12	0.041
<i>n</i> -Heptylbenzene (0.8 M isotridecanol)	12	0.035
<i>n</i> -Octylbenzene (0.9 M isotridecanol)	13	0.046
<i>n</i> -Decylbenzene (1.0 M isotridecanol)	13	0.030
<i>n</i> -Dodecylbenzene (1.1 M isotridecanol)	13	0.030
<i>n</i> -Tridecylbenzene (1.3 M isotridecanol)	12	0.034
2-Ethyl-1-hexanol	15	0.57
Phenetyl alcohol	39	20
1-Decanol	15	0.56
Isotridecanol	10	0.31
2-Nitropropane	34	1.9
1-Nitrohexane	31	0.92
Nitrocyclohexane	32	0.96
4-Nitro- <i>m</i> -xylene	24	0.72
2-Nitro- <i>p</i> -cymene	13	0.15
1,6-Dibromohexane	18	0.41
1,2,4-Trichlorobenzene	16	0.26

^a Extractant: DC18C6 0.5 M. Aqueous feed solution: synthetic concentrate. Aqueous stripping solution: demineralized water.

strontium permeability, nitric acid transport, and nonradioactive cation transport through the FSSL.

The stirring speed of the aqueous solutions in all the experiments was 500 rpm because preliminary experiments carried out with *n*-hexylbenzene (0.5 mol·L⁻¹ 4-nonylphenol) showed that strontium permeability became independent of the aqueous solution stirring speed when the latter was greater than 500 rpm, as shown in Fig. 3. This independence indicates that a constant and minimum value of the thickness of the aqueous feed boundary layer, *d_a*, was reached (4).

The strontium permeabilities obtained are shown in Table 5. We can see that the highest strontium permeabilities were obtained with 4-nonylphenol as a phase modifier. However, in strontium transport experiments carried out with hollow fiber SLMs (7), changes in the physical characteristics of the organic phase (mainly viscosity) were observed after a long

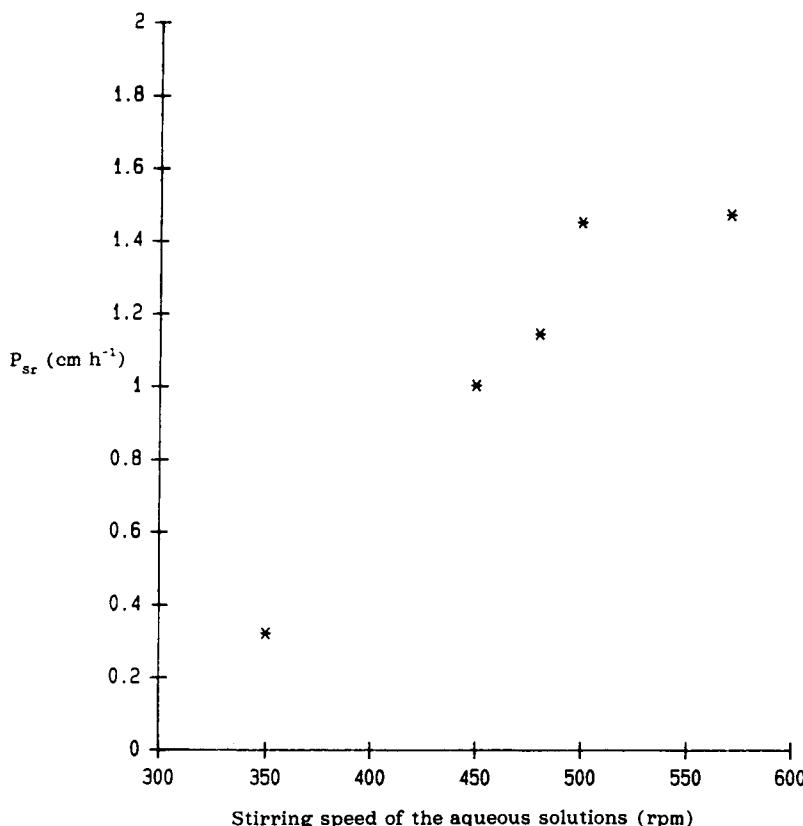


FIG. 3 Influence of the stirring speed of the aqueous phases on the strontium permeability of the SLM. Extractant: DC18C6 ($0.5 \text{ mol} \cdot \text{L}^{-1}$). Membrane solvent: *n*-Hexylbenzene ($0.5 \text{ mol} \cdot \text{L}^{-1}$ 4-nonylphenol). Aqueous feed solution: Synthetic concentrate. Aqueous stripping solution: Demineralized water.

working time, which made it necessary to stop the experiment. It is possible that the long contact time between the 4-nonylphenol and the high nitric acid content aqueous feed solution caused nitration of the aromatic ring of 4-nonylphenol.

Among the other organic phases tested, *n*-hexylbenzene ($0.7 \text{ mol} \cdot \text{L}^{-1}$ isotridecanol) led to the highest strontium permeability ($0.78 \text{ cm} \cdot \text{h}^{-1}$). That produced 80% strontium transport for the first 24 hours.

In some cases a relationship between strontium permeability and a number of membrane solvent characteristics was found. When the strontium distribution coefficient at the membrane/stripping-solution interface is

TABLE 5
Influence of the Membrane Solvent on the Strontium Permeability through the SLM^a

Membrane solvent	Strontium permeability through the SLM (cm/h)
<i>p</i> -Diisopropylbenzene (0.6 M 4-nonylphenol)	1.45
<i>p</i> -Diisopropylbenzene (0.9 M isotridecanol)	0.65
<i>n</i> -Hexylbenzene (0.5 M 4-nonylphenol)	1.46
<i>n</i> -Hexylbenzene (0.7 M 1-decanol)	0.71
<i>n</i> -Hexylbenzene (0.7 M isotridecanol)	0.78
<i>n</i> -Heptylbenzene (0.8 M isotridecanol)	0.67
<i>n</i> -Octylbenzene (0.9 M isotridecanol)	0.52
<i>n</i> -Decylbenzene (1.0 M isotridecanol)	0.45
<i>n</i> -Dodecylbenzene (1.1 M isotridecanol)	0.26
<i>n</i> -Tridecylbenzene (1.3 M isotridecanol)	0.31
1-Decanol	0.37
Isotridecanol	0.22

^a Extractant: DC18C6 (0.5 M). Aqueous feed solution: synthetic concentrate. Aqueous stripping solution: demineralized water. Stirring speed of the aqueous solutions: 500 rpm.

much lower than at the membrane/feed-solution interface, strontium permeability may be expressed by the following equation (4):

$$P_{\text{Sr}} = \frac{D_{\text{Sr}}}{D_{\text{Sr}} \Delta_a + \Delta_0} \quad (2)$$

with

$$\Delta_a = d_a/D_a \quad (3)$$

and

$$\Delta_0 = \tau d_0/D_0 \quad (4)$$

where P_{Sr} = strontium permeability ($\text{cm} \cdot \text{h}^{-1}$)

D_{Sr} = strontium distribution coefficient between the membrane and the aqueous feed solution

Δ_a = transport resistance due to diffusion by the aqueous feed boundary layer

Δ_0 = transport resistance due to diffusion by the membrane

d_a = thickness of the aqueous feed boundary layer

D_a = aqueous diffusion coefficient of the strontium-containing species

d_0 = thickness of the membrane

τ = tortuosity of the support pores

D_0 = membrane diffusion coefficient of the strontium-containing species

On the other hand, the membrane diffusion coefficient may be expressed by the Wilkie and Chang equation (10):

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

where X = association parameter of the solvent

T = absolute temperature

M = molecular weight of the solvent

μ = dynamic viscosity of the solvent

V = molar volume of the extractant

Equations (2) and (5) can be combined to obtain a new expression for strontium permeability:

$$P_{\text{Sr}} = \frac{D_{\text{Sr}}}{D_{\text{Sr}} \Delta_a + K \mu / M^{0.5}} \quad (6)$$

In Eq. (6), K includes all the terms of Δ_0 which do not depend on the nature of the solvent:

$$K = \frac{\tau d_0 V^{0.6}}{7.4 \times 10^{-8} X^{0.5} T 3600} \quad (7)$$

Equation (6) may also be expressed by

$$\frac{1}{P_{\text{Sr}}} = \Delta_a + \frac{K \mu}{D_{\text{Sr}} M^{0.5}} \quad (8)$$

Equations (6) and (8) show that the influence of solvent characteristics on the strontium permeability depends on the strontium distribution coefficient, the molecular weight, and the viscosity of the solvent.

In the cases where the aromatic solvents were modified with isotridecanol or 1-decanol, we can consider that D_{Sr} between the membrane and the aqueous feed solution is very similar, and Eq. (8) indicates that the plot of $1/P_{\text{Sr}}$ versus $\mu M^{0.5}$ must give a straight line with a slope equal to K/D_{Sr} and an intersection equal to Δ_a , as shown in Fig. 4. Indeed, we obtained the following equation:

$$1/P_{\text{Sr}} = 0.66 (\pm 0.07) + 5.0 (\pm 0.2) \mu M^{0.5}$$

As the aqueous diffusion coefficients of the metal ions are normally of the order of $10^{-5} \text{ cm}^2 \cdot \text{s}^{-1}$, we can obtain from the value of Δ_a , by using Eq. (3), a value of d_a equal to 0.0230 cm.

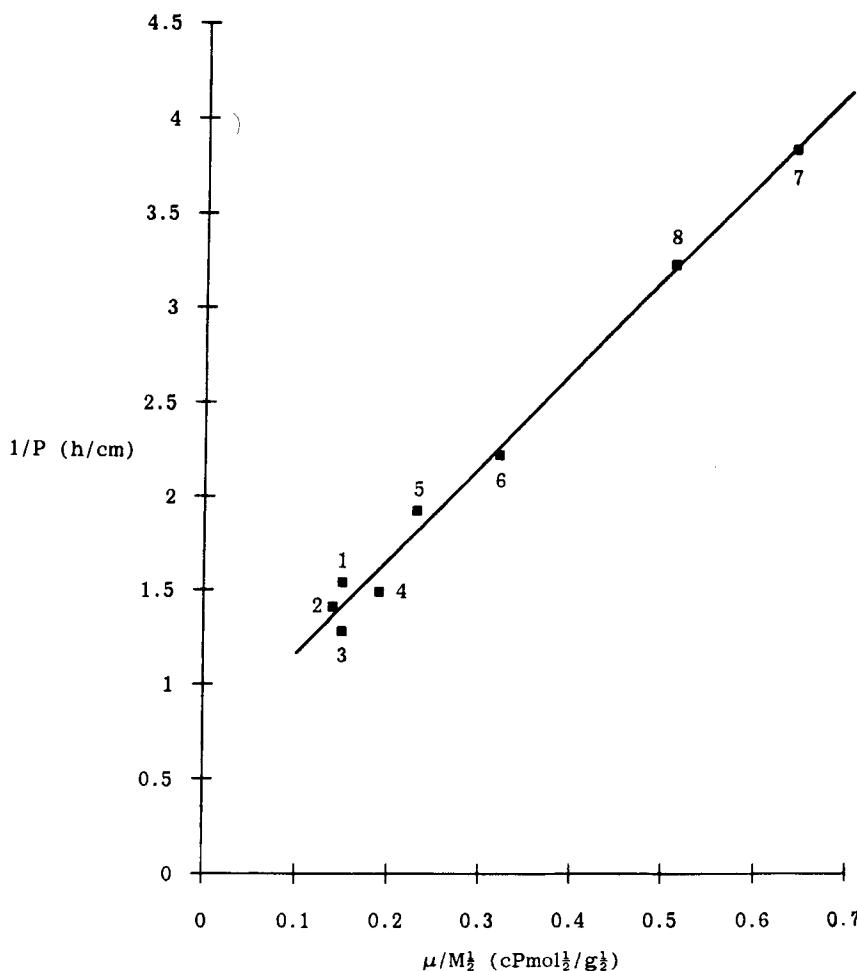


FIG. 4 Influence of the viscosity (μ) and molecular weight (M) of the membrane solvent on the strontium permeability of the SLM. 1: *p*-Diisopropylbenzene (0.9 mol·L $^{-1}$ isotridecanol); 2: *n*-hexylbenzene (0.7 mol·L $^{-1}$ 1-decanol); 3: *n*-hexylbenzene (0.7 mol·L $^{-1}$ isotridecanol); 4: *n*-heptylbenzene (0.8 mol·L $^{-1}$ isotridecanol); 5: *n*-octylbenzene (0.9 mol·L $^{-1}$ isotridecanol); 6: *n*-decylbenzene (1.0 mol·L $^{-1}$ isotridecanol); 7: *n*-dodecylbenzene (1.1 mol·L $^{-1}$ isotridecanol); 8: *n*-tridecylbenzene (1.3 mol·L $^{-1}$ isotridecanol). Extractant: DC18C6 (0.5 mol·L $^{-1}$). Aqueous feed solution: Synthetic concentrate. Aqueous stripping solution: De-mineralized water.

TABLE 6
Transport of Nitric Acid through the SLM^a

Membrane solvent	pH of the aqueous stripping solution after 24 hours
<i>p</i> -Diisopropylbenzene (0.6 M 4-nonylphenol)	0.78
<i>p</i> -Diisopropylbenzene (0.9 M isotridecanol)	0.62
<i>n</i> -Hexylbenzene (0.5 M 4-nonylphenol)	0.93
<i>n</i> -Hexylbenzene (0.7 M 1-decanol)	0.47
<i>n</i> -Hexylbenzene (0.7 M isotridecanol)	0.45
<i>n</i> -Heptylbenzene (0.8 M isotridecanol)	0.18
<i>n</i> -Octylbenzene (0.9 M isotridecanol)	0.40
<i>n</i> -Decylbenzene (1.0 M isotridecanol)	0.35
<i>n</i> -Dodecylbenzene (1.1 M isotridecanol)	0.33
<i>n</i> -Tridecylbenzene (1.3 M isotridecanol)	0.54
1-Decanol	0.38
Isotridecanol	0.44

^a Extractant: DC18C6 (0.5 M). Aqueous feed solution: synthetic concentrate. Aqueous stripping solution: demineralized water. Stirring speed of the aqueous solutions: 500 rpm.

TABLE 7
Transport of Nonradioactive Cations through the SLM^a

Membrane solvent	Time (h)	Concentration (mg/L)			
		Na	Ca	Mg	Fe
<i>p</i> -Diisopropylbenzene (0.6 M 4-nonylphenol)	30	4,100	62	<0.1	<0.1
<i>p</i> -Diisopropylbenzene (0.9 M isotridecanol)	120	2,800	30	0.38	<0.1
<i>n</i> -Hexylbenzene (0.7 M 1-decanol)	72	1,400	33	0.21	<0.1
<i>n</i> -Hexylbenzene (0.7 M isotridecanol)	120	2,800	33	0.45	<0.1
<i>n</i> -Heptylbenzene (0.8 M isotridecanol)	120	3,900	53	0.23	<0.1
<i>n</i> -Octylbenzene (0.9 M isotridecanol)	96	1,700	26	0.31	<0.1
<i>n</i> -Decylbenzene (1.0 M isotridecanol)	150	2,100	29	0.28	<0.1
<i>n</i> -Dodecylbenzene (1.1 M isotridecanol)	144	1,800	19	0.25	<0.1
<i>n</i> -Tridecylbenzene (1.3 M isotridecanol)	168	2,000	19		<0.1
1-Decanol	97	5,500	39	275	4.4
Isotridecanol	120	17,400	17	<0.1	<0.1

^a Extractant: DC18C6 (0.5 M). Aqueous feed solution: synthetic concentrate. Aqueous stripping solution: demineralized water. Stirring speed of the aqueous solutions: 500 rpm.

On the other hand, the use of isotridecanol as a membrane solvent led to the lowest values in strontium permeability because this solvent has the highest viscosity.

Finally, there were no significant differences in the transport of nitric acid (see Table 6) and the transport of sodium, calcium, magnesium, and iron (see Table 7) through the SLM with the different solvents tested. In all cases there was a considerable transport of nitric acid through the membrane (the pH of the stripping solution after 24 hours was always less than 1). Sodium was the most concentrated cation in the stripping solution, but its concentration was only a few grams per liter in all cases.

CONCLUSIONS

Even though the distribution coefficients of strontium found with nitrated compounds as a membrane solvent were the highest, this type of solvent cannot be used in SLMs due to the resulting short membrane lifetime. However, the strontium distribution coefficients were sufficiently high (>10) when organic phases that allowed stable SLMs to be obtained were used (with isotridecanol or an aromatic compound with aliphatic groups of six or more carbon atoms, modified with long-chain alcohols as membrane solvents).

For the aromatic solvents modified with isotridecanol or 1-decanol, we found a good correlation between the strontium permeability of the FSSLM and the two parameters of the membrane diffusion coefficient, which were the molecular weight of the membrane solvent and its dynamic viscosity. Among these solvents, the highest strontium permeability values were obtained with the *n*-hexylbenzene ($0.7 \text{ mol}\cdot\text{L}^{-1}$ isotridecanol) mixture.

This value was only exceeded when 4-nonylphenol was used as the phase modifier, but after a long contact time with the nitric acid this compound reacts in the aqueous feed solution. Therefore, it cannot be used in long-term experiments.

On the other hand, we obtained the least satisfactory results for strontium permeability when isotridecanol was used as the membrane solvent because of its high viscosity.

Nitric acid transport through the membrane was not greatly influenced by the solvent used. Thus, *n*-hexylbenzene ($0.7 \text{ mol}\cdot\text{L}^{-1}$ isotridecanol) is the best solvent for strontium transport through SLMs because it produced the highest permeability value. With this membrane solvent it is possible to obtain a high level of strontium transport FSSLM (80% for 24 hours despite considerable nitric acid transport) and the membrane stability is very good (more than 200 hours). In addition, the transport of nonra-

dioactive cations is not very significant (the sodium concentration is 3 g·L⁻¹ after 120 hours when the initial concentration in the aqueous feed solution is 80 g·L⁻¹). Very satisfactory strontium transport through an HFSLM can thus also be expected, leading to a high level of strontium decontamination of nuclear fuel reprocessing concentrate solutions.

ACKNOWLEDGMENTS

This work was financed by the Commission of the European Communities and the Commissariat à l'Energie Atomique (France) under contract FI 1W-0016F. J.C. is very grateful to the Commission of the European Communities for the fellowship he received.

REFERENCES

1. W. R. Dworzar and A. J. Naser, *Sep. Sci. Technol.*, 22(2&3), 677 (1987).
2. J. F. Dozol, J. Casas, and A. M. Sastre, *New Separation Chemistry Techniques for Radioactive Waste and Others Specific Applications*, Elsevier Applied Science, London, 1991, p. 173.
3. J. F. Dozol, *New Separation Chemistry Techniques for Radioactive Waste and Others Specific Applications*, Elsevier Applied Science, London, 1991, p. 95.
4. P. R. Danesi, *Sep. Sci. Technol.*, 19(11&12), 857 (1984-85).
5. Z. Lakkis-Zahreddine, Thesis, ENSC de Strasbourg, 1986.
6. E. P. Horwitz, M. L. Dietz, and D. E. Fisher, *Solv. Extr. Ion Exch.*, 8(1), 199 (1990).
7. J. F. Dozol, S. Eymard, R. Gambade, G. La Rosa, and J. Casas, CCE Research Contract FI 1W-0016F, Commissariat à l'Energie Atomique, France, 1991.
8. P. R. Danesi, *Developments in Solvent Extraction*, Ellis Horwood Limited, 1988.
9. J. F. Dozol, J. Casas, and A. M. Sastre, *J. Membr. Sci.*, To Be Published.
10. J. C. Charpentier, *Adv. Chem. Eng.*, 11, 1 (1975).

Received by editor August 10, 1992