Transport of Pu(IV) across supported liquid membrane from nitric acid medium using cyanex-923 as the mobile receptor


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Based on the solvent extraction data, experiments were conducted with Flat-sheet Supported Liquid Membrane (FSSLM) to examine the transport behaviour of Pu(IV) to achieve its maximum recovery. This is necessary to optimize the conditions for further operation of Hollow Fiber Supported Liquid Membrane (HFSLM) and Compact Supported Liquid Membrane Flow Cell (CSLMFC). A laboratory scale hollow fiber module, having membrane surface area, 134 cm², was employed in a recycling mode using 'Enka' Accurel hollow fiber polypropylene membranes as the solid support, cyanex-923 as the mobile receptor and dodecane as the membrane solvent. Dilute plutonium nitrate solutions in 1 M HNO₃ generally constituted the source phase. Among the strippants, several reagents were tried, the most efficient strippant found was dilute uranous solution in 1.0 M HNO₃. The recovery of Pu from the Pu oxalate supernatant generated during Pu reconversion operations in the PUREX process was tried, where it was observed that ~ 94% of Pu(IV) could be transported in a single run of duration of 10 h, while employing 0.1M cyanex-923/dodecane as a mobile receptor and uranous solution in 1.0 M HNO₃ as the strippant. Parameters such as source phase concentration, source and strippant phase recirculation flow rate, concentration of carrier in the membrane and strippant, and interferences of other permeants on plutonium permeation pertaining to FSSLM, HFSLM and CSLMFC were optimized.

This followed the utilization of more application oriented configuration i.e. Hollow fiber membrane configuration for the above purpose.

With HFSLM, the $K_m$ values increased from 3.06x10⁶ to 6.27 x10⁸ s⁻¹ by increasing flow rate from 0.03 x10⁻⁶ to 0.14 x10⁻⁶ m³/s at 1 h and the percentage permeation increased from 75 to 90%. In the case of CFSLMC, the $K_m$ increased from 5.41 x10⁶ to 5.74 x10⁹ s⁻¹ while increasing flow rate from 0.27 x10⁻⁶ to 0.42 x10⁻⁶ m³/s, at 1 h and the percentage permeation increased from 65 to 85%.

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Keywords: Pu(IV) transport, FSSLM, mobile receptor, cyanex 923/dodecane, HFSLM, CSLMFC

Two types of liquid membranes are commonly in use, one being supported liquid membrane, consisting of a thin microporous film impregnated with a carrier and the other an emulsion liquid membrane. From the engineering and practical point of view, the former is of particular interest because of its stability, and simplicity of different liquid membrane configurations. With flat sheet supported liquid membranes (FSSLMs) separations can be achieved employing continuous and simultaneous processes of selective extraction and back extraction using a microporous polymer film impregnated with an organic solution containing a suitable extractant. The hollow fiber supported liquid membrane (HFSLM) performs better than FSSLM since it provides larger surface area of the membrane, and as such is viable for using on an industrial scale for separation of metal ions.

HFSLM process consists of polymeric microporous hydrophobic lumens impregnated with a suitable complexing agent that enhances the transport of a desired permeant across the membrane. Considering the modular design and the area per unit volume available, hollow fiber membrane configuration is getting very high attention. Utilizing this technique, the extraction of organic compounds, bio-products as well as metal ions, have been carried out.

In the recent past, efforts have been made to synthesize organophosphorus reagents, such as dihexyl-N,N-diethyldiakamoylmethyl phosphonate (DHDECMP or CMP) and use it above or with the mixture of tributyl phosphate (TBP) for the extraction of actinides and...
lanthanides from nitric acid medium, and in different
diluents\textsuperscript{9-13}. Another compound of this class, octyl
(phenyl)-N,N'-diisobutyl carbamoylmethylphosphine
oxide (CMPO) has received much attention for
separation of minor actinides from high level waste
solutions\textsuperscript{14-16}. The other phosphorus based compounds,
disodicyl phosphoric acid (DIDPA)\textsuperscript{17,18} and trialkyl
phosphine oxide (TRPO)\textsuperscript{19,22} have also been tried for
the above purpose.

Also, bis(2-ethylhexyl) dithiophosphate\textsuperscript{23,24}, TBp\textsuperscript{25,26},
di (2-ethylhexyl) phosphoric acid DEHPA\textsuperscript{27-29},
mono(2-ethylhexyl) phosphonic acid (H\textsubscript{2}HEHP)\textsuperscript{30,31} and
tri-n-octylphosphine oxide (TOPO)\textsuperscript{32,33}, have been
employed for the recovery of several metal ions.

During fuel reprocessing operations, uranium and
plutonium are separated from fission products and also
from each other. In the final stage, Pu(IV) is precipitated
as oxalate, which generates large volumes of oxalate
supernatant containing significant amounts of plutonium
(\(\sim 30\) mg dm\(^{-3}\)).

This paper presents the studies on the optimization
of several parameters such as source phase
concentration, strippant recirculation flow rate, carrier
cyanex 923, in membrane phase and strippant. Initially
experiments were conducted using solvent extraction
technique and then with FSSLM to examine the transport
behaviour and recovery of Pu(IV) and then HFSLM and
CSLMFC were operated under the optimum conditions.
Finally, the recovery of plutonium from oxalate
supernatant, was carried out without any feed adjustment
using cyanex 923.

**Experimental Procedure**

**Chemicals**

Cyanex 923 (mixture of four tri-n-alkyl phosphine
oxides, R\(\text{P(O)}\), R\(\text{R'}\text{P(O)}\), R\(\text{RR'}\text{P(O)}\), and R\(\text{R'R''P(O)}\)
where \(R=C\text{H}_{11}\) and \(R'=C\text{H}_{13}\) was obtained from M/s
Cytec, Canada and dodecane from Fluka and were used
as received. All other chemicals used were of analytical
reagent (AR) grade.

**Tracer solution**

Tracer used Pu (mainly \(^{239}\text{Pu}\)) was purified by the
usual anion exchange method. Oxalate supernatant with
a composition \(\text{Pu}: \sim 30\) mg dm\(^{-3}\), \(U: 5\) g dm\(^{-3}\), \(^{186}\text{Ru}: 0.0032\) mci dm\(^{-3}\), \(^{137}\text{Cs}: 0.003\) mci dm\(^{-3}\), \([\text{HNO}_{3}]: 3.0\) M and \([\text{H}_{2}\text{C}_{2}\text{O}_{4}]: 0.1\) M was used.

**Permeation experiments**

The feed and strip solutions were circulated
through the lumen and on the shell side of the fibers,
respectively, by means of calibrated peristaltic pumps.
The permeation of the radio nuclides through CSLMFC
was followed by periodically sampling the feed and/or
strip solutions. The samples were assayed by alpha
scintillation counter for the nuclides \(^{233}\text{U}, ^{239}\text{Pu}\) and
\(^{241}\text{Am}\) and by gamma scintillation counter for \(^{137}\text{Cs}\).

The membrane area was calculated as: \(A = 2\pi rNL\)
for the hollow fiber and \(A = L \cdot B\) for the compact flow
cell, where, \(r\) is the internal radius of the hollow fibers,
\(N\) is the number of fibers and \(L\) is their length. For
CSLMFC, \(L\) and \(B\) are respectively the length and
breadth of the rectangular piece.

**Supported liquid membrane cells**

Permeation experiments were carried in a single
stage two-compartment membrane cell as discussed in
detail elsewhere\textsuperscript{34}.

Pu(IV) concentration with time in the feed or
stripping solutions, were periodically assayed. All
experiments were performed at room temperature (25°C)
. The rate constant \(K_{\text{obs}}\) (s\(^{-1}\)) is defined as

\[
\ln \frac{[\text{Pu}]}{[\text{Pu}]_{0}} = - K_{\text{obs}} t
\]

where \([\text{Pu}]_{t}\) and \([\text{Pu}]_{0}\) denote the concentration of
plutonium at time \((t)\) and the initial concentration,
respectively and \(t\) is the time elapsed in second.

**Membrane support**

**FSSLM**

Throughout the study, “Enka” Accurel PP thin flat
sheet type hydrophobic microporous polymeric
membranes (\(\sim 300\) \(\mu\)m thick, a nominal porosity of
70\% with an average pore diameter of the order of 0.2
\(\mu\)m) coaded as 2E-PP were used.

Characteristics of the modules used in present study
for HFSLM are internal surface area 134 cm\(^2\), fiber i.d.
2.5 mm, fiber o.d. 2.8 mm, fiber wall thickness 300\(\mu\)m,
fiber length 17 cm, porosity 70\%, pore size 0.1\(\mu\)m
and the material being polypropylene. Specifications
of CSLMFC are: internal surface area 42 cm\(^2\), length
of flat-sheet support 7 cm, width of flat-sheet support 6 cm.

**Liquid-liquid extraction**

Equal volumes (1 mL) of ²³⁹Pu tracer in nitric acid (of desired molarity) and cyanex 923 dissolved in dodecane, were pipetted into a 15 mL glass stoppered tube and mechanically stirred for nearly half an hour at room temperature (25°C). After settling for about 30 min, aliquots from both the phases were withdrawn for radioassay. The distribution ratios (Dpu) of plutonium defined as the ratio of its concentration in the organic phase to that in the aqueous phase, was thus calculated.

For the back extraction studies, aliquots from the loaded organic phase were taken and equilibrated with the same volume of the reagents for about 10 min.

**Results and Discussion**

**Liquid-liquid extraction**

The extraction behaviour of an ionophore for a specific metal ion is normally carried out to understand the behaviour of the carrier. The, D value with cyanex 923 (TRPO, trialkyl phosphine oxide) at 0.1, 0.5, 1.0, 2.0, 3.0, 4.0, 5.0, 6.0 and 7.0 M nitric acid, are 0.80, 1.37, 4.79, 5.51, 11.02, 15.76, 14.65, 13.16 and 12.29 respectively under the experimental conditions (volume ratio (org./aq.):1, extractant 1% cyanex 923, diluent: dodecane, aqueous phase:Pu(IV) at varying nitric acid concentration). The contaminants, which generally accompany Pu such as ¹³⁷Cs, ¹⁰⁶Ru and ¹⁴⁴Ce, were negligibly extracted under the present experimental conditions.

The extraction of the tri-, tetra, penta and hexavalent actinides with cyanex 923 from nitric acid solutions is well-documented, the species extracted into the organic phase are respectively M(NO₃)₃·3TRPO, M(NO₃)₄·2TRPO, MO₂NO₃·TRPO and MO₂(NO₃)₂·2TRPO. The extraction equilibria for Pu(IV) with cyanex 923 from HNO₃ medium can be represented as :

![Fig. 1 - Effect of HNO₃ on transport of Pu(IV) across SLM using cyanex 923 as the carrier; receiving phase: 0.06 M U(IV) solution in 1.0 M HNO₃; carrier concentration: 0.1M cyanex 923 in dodecane](image-url)

<table>
<thead>
<tr>
<th>Source phase</th>
<th>Strippant: Pu(IV) in aqueous HNO₃</th>
<th>Strippant: Dilute solution of Uranous + 1 M HNO₃</th>
</tr>
</thead>
<tbody>
<tr>
<td>Varying HNO₃ conc., keeping 0.1 M cyanex 923</td>
<td>Varying carrier conc., keeping 1.0 M HNO₃</td>
<td></td>
</tr>
<tr>
<td>HNO₃ (M)</td>
<td>Kₓₓ×10⁻⁴</td>
<td>% Plutonium permeation after 6 h</td>
</tr>
<tr>
<td>1.0</td>
<td>6.19</td>
<td>96.3</td>
</tr>
<tr>
<td>2.0</td>
<td>7.93</td>
<td>95.2</td>
</tr>
<tr>
<td>3.0</td>
<td>8.82</td>
<td>91.3</td>
</tr>
<tr>
<td>4.0</td>
<td>7.14</td>
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<td>6.53</td>
<td>84.3</td>
</tr>
<tr>
<td>7.0</td>
<td>6.58</td>
<td>83.7</td>
</tr>
<tr>
<td>8.0</td>
<td>6.50</td>
<td>83.1</td>
</tr>
</tbody>
</table>
Table 2 — Transport of Pu (IV) across SLM from aqueous nitric acid medium as a function of strippant variation in receiving phase

<table>
<thead>
<tr>
<th>Strippants</th>
<th>Time elapsed (h)</th>
<th>Apparent rate constant ( K_{un}(\times 10^{-5} \text{ s}^{-1}) )</th>
<th>Recovery (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1 M ( \text{NH}_2\text{OH-HCl in 0.3 M HNO}_3 )</td>
<td>6</td>
<td>11.26</td>
<td>10.1</td>
</tr>
<tr>
<td>0.35 M ( \text{H}_2\text{C}_2\text{O}_4 ) in 0.3 M HNO(_3)</td>
<td>6</td>
<td>5.68</td>
<td>95.1</td>
</tr>
<tr>
<td>0.1 M EDTA</td>
<td>6</td>
<td>11.49</td>
<td>56.1</td>
</tr>
<tr>
<td>0.5 M ( \text{Na}_2\text{CO}_3 )</td>
<td>6</td>
<td>8.89</td>
<td>30.3</td>
</tr>
<tr>
<td>1 M HClO(_4)</td>
<td>6</td>
<td>11.10</td>
<td>29.6</td>
</tr>
<tr>
<td>0.06 M U(IV) solution in 1 M HNO(_3)</td>
<td>6</td>
<td>6.19</td>
<td>96.3</td>
</tr>
</tbody>
</table>

\[
\text{Pu}^{4+} + 4 \text{NO}_3^- + 2 \text{TRPO}_{org} \rightleftharpoons \text{Pu} (\text{NO}_3)_4 \cdot \text{TRPO}_{org}
\]  

(2)

SLM transport of Pu(IV)

Transport of Pu(IV) across SLM experiments carried out without carrier immobilized in the membrane indicated no transport of Pu(IV), suggesting thereby that the transport of only the plutonium-cyanex 923 complex (reaction 2) takes place.

Effect of nitric acid concentration on plutonium transport

Results for the transport of Pu from an aqueous feed adjusted to different concentrations of nitric acid through cyanex 923/dodecane SLM using dilute solution of uranous in 1.0 M HNO\(_3\) as strippant are summarised in Table 1. The maximum Pu recovery, ~96% (after 6h) and an apparent rate constant, \( K_{un} \approx 6.19 \times 10^{-5} \text{ s}^{-1} \) was obtained at a feed acidity of 1M. It is further clear from Fig. 1, that the transport of Pu(IV) from the feed to the product side decreases from 1.0 to 8.0 M of HNO\(_3\), which may be explained as due to the simultaneous transport of HNO\(_3\) into the strip compartment.

Effect of cyanex 923 concentration on Pu(IV) transport

Results of Pu(IV) transport from 1.0 M HNO\(_3\) by varying concentrations of ionophore cyanex 923/dodecane are also presented in Table 1. The maximum \( K_{un} \approx 9.23 \times 10^{-5} \text{ s}^{-1} \), was observed at 0.2 M cyanex 923 at elapsed time of 6 h. It can further be seen from Fig. 2, that by increasing the carrier concentration, plutonium transport gradually increased and was highest for 0.2 M of cyanex 923 at the elapsed of 1, 2, 3, 4 and 5 h, but at the end of the 6 h a maximum (~96%) was observed at 0.1 M cyanex 923. The optimum carrier concentration for the transport studies was thus chosen as ~0.1 M.

Effect of strippants on permeation behaviour of Pu(IV)

Transport of Pu(IV) ion across the cyanex 923/dodecane membrane is to a great extent dependent on the nature and type of the strippant used in the product side of the membrane. Several aqueous complexing strippants such as EDTA, \( \text{Na}_2\text{CO}_3 \), HClO\(_4\), \( \text{H}_2\text{C}_2\text{O}_4 \) in 0.3 M HNO\(_3\), \( \text{NH}_2\text{OH-HCl} \) in 0.3 M HNO\(_3\) and dilute solution of uranous in 1.0 M HNO\(_3\) were tested, of which the dilute solution of uranous in 1.0 M HNO\(_3\) was found to be the most efficient (Table 2). It can be seen that \( \text{NH}_2\text{OH-HCl} \) gave \( \text{Na}_2\text{CO}_3 \sim 10\), HClO\(_4\) ~ 30, EDTA ~
apparent rate constant, \( x \) flow rate of the feed solution (lumen side). The flow rates of HFSLM and CFSLMFC are varied between 0.03 to 0.42 \( \times 10^{-6} \) m\(^3\)/s. At optimum flow rates, the apparent rate constant \( K_{ohs} \) is maximum and a further increase in flow rate does not increase the \( K_{ohs} \) value. Furthermore, high feed velocities can be detrimental to the membrane stability, and also it can force the carrier solution out of the membrane pores by the excessive pressure exerted by the circulating liquid.

The data presented in Table 3 indicated that \( K_{ohs} \) increased from 3.06 \( \times 10^{-5} \) to 6.27 \( \times 10^{-5} \) s\(^{-1}\) by increasing flow rate from 0.03 \( \times 10^{-6} \) to 0.14 \( \times 10^{-6} \) m\(^3\)/s at 1 h and percentage permeation from 75 to 90% with HFSLM. When flow rate increased in the case of CFSLMFC from 0.27 \( \times 10^{-6} \) to 0.42 \( \times 10^{-6} \) m\(^3\)/s, \( K_{ohs} \) increased from 5.41 \( \times 10^{-5} \) to 5.74 \( \times 10^{-5} \) s\(^{-1}\) at 1 h and percentage permeation from 65 to 85%.

Effect of Pu concentration

Effect of initial plutonium concentration (3 to 76 mg dm\(^{-3}\)) in the feed on its transport across SLM impregnated with cyanex 923 as carrier was evaluated. After 6 h of permeation the \( K_{ohs} \times 10^{-5} \) s\(^{-1}\) are 8.02, 10.43, 7.09, 10.77, 5.24 and 4.51 and the % recovery of Pu(IV) are 96.3, 96.5, 95.1, 80.1, 72.1 and 52.7 at Pu(IV) concentrations of 3.4, 11.6, 17.1, 38.0, 45.8 and 76.0 mg dm\(^{-3}\) respectively. The experimental conditions for all the systems were kept same, i.e. in source phase various Pu concentration solution in 1.0 M HNO\(_3\), strippant dilute uranous solution in 1.0 M HN\(_3\) (Table 2) which suggests that the overall recovery across SLM remained practically unaffected with increasing plutonium concentration up to ca. 17 mg dm\(^{-3}\). However, further increase in plutonium concentration resulted in decrease in the permeation, possibly because of the saturation of the membrane with the permeant. Insufficient effective membrane area and incomplete plutonium stripping may be additional factors for poor permeation. It can be seen from Fig. 3 that at Pu(IV) concentration of 3.4 mg dm\(^{-3}\), above 95% permeation could be easily obtained with 0.1 M cyanex 923, whereas at around 76 mg dm\(^{-3}\), only about 50% permeation was observed.

Transport of Pu(IV) across HFSLM and CPSLMFC

The optimum hydrodynamic conditions for the modules used in this work were properly identified studying the plutonium permeability as a function of flow rate of the feed solution (lumen side). The flow rates of HFSLM and CFSLMFC are varied between 0.03 \( \times 10^{-6} \) to 0.42 \( \times 10^{-6} \) m\(^3\)/s. At optimum flow rates, the apparent rate constant \( K_{ohs} \) is maximum and a further increase of flow rate does not increase the \( K_{ohs} \) value. Furthermore, high feed velocities can be detrimental to the membrane stability, and also it can force the carrier solution out of the membrane pores by the excessive pressure exerted by the circulating liquid.

The data presented in Table 3 indicated that \( K_{ohs} \) increased from 3.06 \( \times 10^{-5} \) to 6.27 \( \times 10^{-5} \) s\(^{-1}\) by increasing flow rate from 0.03 \( \times 10^{-6} \) to 0.14 \( \times 10^{-6} \) m\(^3\)/s at 1 h and percentage permeation from 75 to 90% with HFSLM. When flow rate increased in the case of CFSLMFC from 0.27 \( \times 10^{-6} \) to 0.42 \( \times 10^{-6} \) m\(^3\)/s, \( K_{ohs} \) increased from 5.41 \( \times 10^{-5} \) to 5.74 \( \times 10^{-5} \) s\(^{-1}\) at 1 h and percentage permeation from 65 to 85%.

Separation of plutonium from oxalate supernatant generated during Pu reconversion operation

The recovery of plutonium from oxalate supernatant, without any feed adjustment, using cyanex 923/dodecane as the ionophore across the SLM gave ~ 93% recovery of Pu(IV) (Table 4) in 10 h, and an apparent rate constant, \( K_{ohs} \) 5.51 \( \times 10^{-5} \) s\(^{-1}\) at a feed acidity of 3.0 M HNO\(_3\). This recovery was achieved using dilute uranous solution in 1.0 M HNO\(_3\). The transportability of several long-lived fission products such as \(^{137}\)Cs, \(^{106}\)Ru and \(^{144}\)Ce was tested across the SLM. No detectable amount of these fission products permeated to receiving side even after 6 h of operation. Thus, selective
Table 3 — Apparent rate constants of Pu(IV) using SLM, Accurel hollow fiber SLM and Compact SLM flow cell

Source phase : Pu(IV) in aqueous 1.0 M HNO₃; Strippant : Dilute solution of uranous + 1 M HNO₃; Carrier concentration : 0.1M cyanex 923/dodecane

<table>
<thead>
<tr>
<th>Time, (h)</th>
<th>Apparent rate constant</th>
<th>Flow rate ( \times 10^{-5} ), m²/s</th>
<th>Flat sheet</th>
<th>Hollow fiber</th>
<th>Hollow cell</th>
<th>Compact cell</th>
<th>Compact cell</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( \times 10^{5} ) s⁻¹</td>
<td>( \times 10^{5} ) s⁻¹</td>
<td>( \times 10^{5} ) s⁻¹</td>
<td>( \times 10^{5} ) s⁻¹</td>
<td>( \times 10^{5} ) s⁻¹</td>
<td>( \times 10^{5} ) s⁻¹</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>11.37</td>
<td>3.06</td>
<td>6.27</td>
<td>5.41</td>
<td>5.74</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>11.01</td>
<td>2.41</td>
<td>4.43</td>
<td>2.75</td>
<td>4.91</td>
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<td></td>
</tr>
<tr>
<td>3</td>
<td>9.40</td>
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<td>3.51</td>
<td>2.31</td>
<td>3.81</td>
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<td></td>
</tr>
<tr>
<td>4</td>
<td>8.68</td>
<td>1.90</td>
<td>3.22</td>
<td>2.04</td>
<td>3.46</td>
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<td></td>
</tr>
<tr>
<td>5</td>
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<td>1.81</td>
<td>2.71</td>
<td>1.91</td>
<td>2.93</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>6.72</td>
<td>1.62</td>
<td>2.23</td>
<td>1.72</td>
<td>2.31</td>
<td></td>
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</tbody>
</table>

separation of plutonium from associated fission products is clearly established.

Conclusions

From this study it can be concluded that cyanex 923 has high permeation efficiency for Pu(IV), and its recovery from aqueous solutions. It suggests the potential application of this procedure in hydrometallurgy. The plutonium species formed with cyanex 923 is \( \text{Pu} (\text{NO}_3)_2 \cdot 2\text{cyanex 923} \).

Polypropylene proved to be quite suitable polymeric hydrophobic support for FSSLM and HFSLM applications. After obtaining the results with FSSLM batch mode, HFSLM and CSLMFC techniques were operated which were found to be important in terms of finding out the applicability in continuous mode. Around 95% of the Pu(IV) could be transported with the help of HFSLM and CSLMFC in about 6-7 h in a single run.

In the present study, feasibility of recovery of plutonium from oxalate supernatant without any feed adjustment using cyanex 923 carrier was explored across FSSLM, HFSLM and CSLMFC. The recovery of Pu(IV) from oxalate supernatant in presence of fission products, was above 95%.

Table 4 — Transport of Pu(IV) across SLM from oxalate bearing wastes

Source phase : Oxalate treated supernatant containing Pu(IV) at 3.0 M HNO₃; Strippant: Dilute uranous solution in 1.0 M HNO₃; Carrier concentration : 0.1M cyanex 923/dodecane

<table>
<thead>
<tr>
<th>Time elapsed (h)</th>
<th>Apparent rate constant ( K_{aw} \times 10^{-5} \text{s}^{-1} )</th>
<th>Recovery (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4.95</td>
<td>28.8</td>
</tr>
<tr>
<td>2</td>
<td>8.75</td>
<td>40.1</td>
</tr>
<tr>
<td>3</td>
<td>8.13</td>
<td>50.1</td>
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<td>70.9</td>
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<td>92.5</td>
</tr>
<tr>
<td>10</td>
<td>5.51</td>
<td>93.6</td>
</tr>
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</table>

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The authors wish to thank Shri. N K Bansal, Associate Director (Operation), Nuclear Recycle Group and Shri. R D Changrani, Chief Superintendent, Nuclear Recycle Group Facilities, Tarapur for their keen interest in this work.
References