

## DEVELOPMENT OF PARTITIONING PROCESS AT JAERI

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### ABSTRACT

One of the methods to manage HLLW generated from nuclear fuel reprocessing is to partition long-lived transuranic elements (TRU) and heat-generating nuclides such as Sr-90 and Cs-137 from HLLW, and then to transmute TRU to shorter-lived or stable nuclides by a fission process and to fix Sr-90 and Cs-137 in mineral-like stable compounds.

Since 1985, much efforts have been directed to developing an advanced partitioning process for separating elements in HLLW into four groups; TRU, Tc-platinum group metals (PGM), Sr-Cs and the other elements. The most recent studies on the separation of the TRU group with diisodecylphosphoric acid (DIDPA) will be described in our presentation.

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### 1. Introduction

Partitioning of nuclides such as transuranium elements (TRU), Tc-99, Sr-90 and Cs-137 in a high-level liquid waste (HLLW) generated in nuclear fuel reprocessing and transmutation of long-lived nuclides by nuclear reactions are expected to increase the efficiency of high-level waste disposal and will seek the utilization of existing resources in a spent fuel.

Since 1985, much efforts have been directed to developing an advanced partitioning process for separating elements in HLLW into four groups; TRU, Tc-platinum group metals (PGM), Sr-Cs and the other elements. The most recent studies on the separation of the TRU group with diisodecylphosphoric acid (DIDPA) will be described in the present paper.

### 2. Separation of TRU, especially Np

Effectiveness of DIDPA for the complete extraction of Am, Cm, Pu and U had been already demonstrated with an actual HLLW before 1984. Therefore much attention has been given to how to increase the Np extraction with DIDPA. The penta-valent Np, which is dominant in HLLW, is the TRU most difficult to extract with general organic solvents. We found that the addition of hydrogen peroxide to the TRU extraction with DIDPA accelerated the pentavalent Np extraction. Through the experiments on a counter-current continuous extraction using a mixer-settler, more than 99.96% of penta-valent Np was found to be extracted when hydrogen peroxide was fed at a level to compensate for its decomposition as shown in Fig. 1

The behavior of fission and corrosion products in the continuous counter-current extraction with DIDPA was experimented with a simulated HLLW. Table 1 shows that more than 99.99% of Nd, which was used as a stand-in for rare earths, Am and Cm, was recovered by back-extraction with 4 M nitric acid. Most of Fe was extracted with DIDPA, but not back-extracted with 4 M nitric acid. Therefore, Fe could be separated from Nd with a decontamination factor of about 100. About 6% of Ru and 11% of Rh were extracted in this experiment. Their extraction behavior might be changed in a practical application because their chemical forms seem to differ in an actual HLLW. Almost all Cs and Sr were transferred to the raffinate fraction and Nd was found to be completely separated from these heat generating nuclides in HLLW.

### 3. Separation of Am and Cm from rare earths

In order to develop a more continuous process, we are developing a preferential back-extraction of Am and Cm, leaving rare earths in the DIDPA solvent. Figure 2 shows the comparison of distribution ratio of Am with those of rare earths. The distribution ratio of Sm was the nearest to that of Am. Figure 3 shows the pH dependence of the distribution ratio of Am and Nd in the back-extraction from the DIDPA solvent with diethylenetriaminepentaacetic acid (DTPA). The separation factor between two elements became more than 10.

The pH of the stripping solution, which contains 0.05 M DTPA, 1 M lactic acid and ammonium, changes stage by stage in continuous back-extraction because of the extraction of ammonium. Figure 4 shows the pH profile in the continuous back-extraction. The profile could be estimated by calculation from batch experimental data of ammonium extraction. Good agreement between observed and calculated values makes it possible to design and optimize the process flow sheet from the batch experimental data of distribution ratio.

Figure 5 shows the Am concentration profile obtained in the continuous back-extraction experiment with the DIDPA solvent containing only Am. About 99.9% of Am was back-extracted from the solvent and the Am concentration profile was in fair agreement with calculated profile as shown in Fig. 6.

The continuous back-extraction experiment was performed with the DIDPA solvent containing Nd and La, which are stand-ins for Am-Cm and rare earths respectively. The experimental results as shown in Fig. 7 revealed that more than 99.9% of Nd was back-extracted, while more than 98% of La was left in the solvent, and the Nd and La concentration profiles agreed very closely with calculated profiles.

These findings show that the estimation of the back-extraction behavior of Am, Cm and rare earths by calculation would be valid enough for the optimization of the back-extraction process.

Figure 8 shows calculated concentration profiles of Am and Nd in the optimized process. The calculation shows that 99.995% of Am is back-extracted and only 1.7% of Nd or 1.1% of rare earths accompanies Am. As the weight ratio of Am and Cm to rare earths is only 3.3% in HLLW, it would become about 75% after separation.

We are going to confirm this estimation by performing the continuous experiment with the solvent containing both Am and rare earths.

#### 4. Back-extraction of U

After the Am and Cm back-extraction with DTPA solution, all of rare earths and other TRU (Np and Pu) can be back-extracted with 4 M nitric acid and 0.8 M oxalic acid solution, respectively. As for the back-extraction of U, effectiveness of phosphoric acid has been already studied. However, it has some difficulties in practical use.

We are developing on the application of a sodium carbonate solution to the U back-extraction from the DIDPA solvent.

Batch experiments showed that the distribution ratio of U became lower than 0.1 when the 0.8 to 2 M sodium carbonate solution was used as back-extractant. However, the emulsion was formed. We found that it could be avoided by increasing temperature and/or adding alcohol.

Table 2 shows required temperature to avoid the emulsion. If the temperature was increased to 65 °C, no emulsion was observed. If ethanol was added with a ratio of 15 vol%, the temperature can be lowered to 15°C.

Figure 9 shows the distribution ratio of U under the condition that no emulsion was formed. Because of the low distribution ratio, U would be back-extracted in a small number of stages in a mixer-settler.

#### 5. Construction of partitioning process

Through these fundamental studies, the advanced partitioning process for the TRU separation from HLLW has been constructed as shown in Fig.10 and its effectiveness will be demonstrated with an actual HLLW at the NUCEF (Nuclear Fuel Cycle Safety Engineering Research Facility) in the near future.

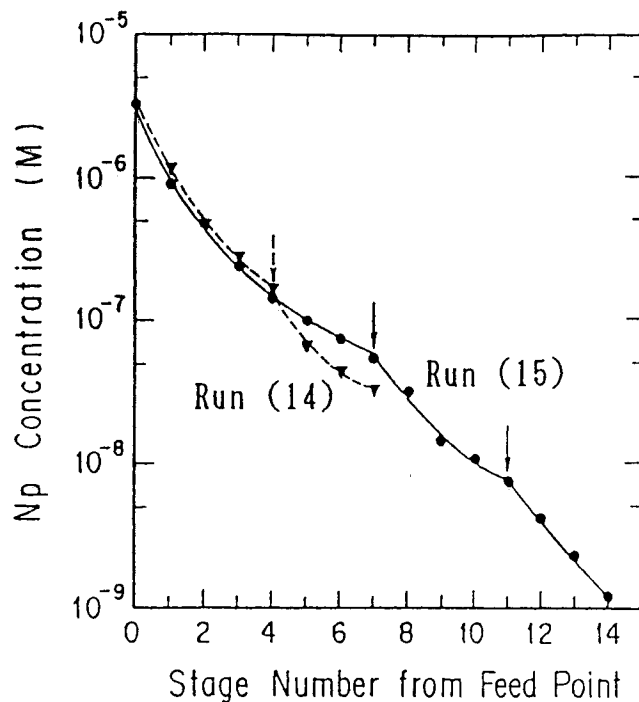


Fig.1 Concentration profile of Np in aqueous phase at the extraction section 45°C. Downward arrows show the points where H<sub>2</sub>O<sub>2</sub> is added.

Run (14) (7-stages) : 99.0% recovery.

Run (15) (14-stages): over 99.96% recovery

Table 1 Behaviors of fission and corrosion products

- Ratios (%) of each element in the three fractions. 45 °C.

7-stages extraction with the DIDPA solvent  
 4-stages scrubbing with 0.5M HNO<sub>3</sub> - 0.5M H<sub>2</sub>O<sub>2</sub>  
 5-stages stripping with 4M HNO<sub>3</sub>

Element	Raffinate	4 M HNO <sub>3</sub> (Product I)	Organic Fraction
Na	>99.6	0.31	<0.1
Cr	>99.5	0.38	<0.1
Fe	0.13	0.1	99.77
Ni	>99.5	0.44	<0.1
Rb	>99.97	<0.02	<0.01
Cs	>99.99	<0.005	<0.002
Sr	>99.95	0.042	<0.004
Ba	>99.94	0.027	<0.04
Ru	93.53	0.98	5.49
Rh	88.30	1.86	9.84
Pd	>99.5	0.43	<0.02
Nd	0.002	>99.99	<0.006

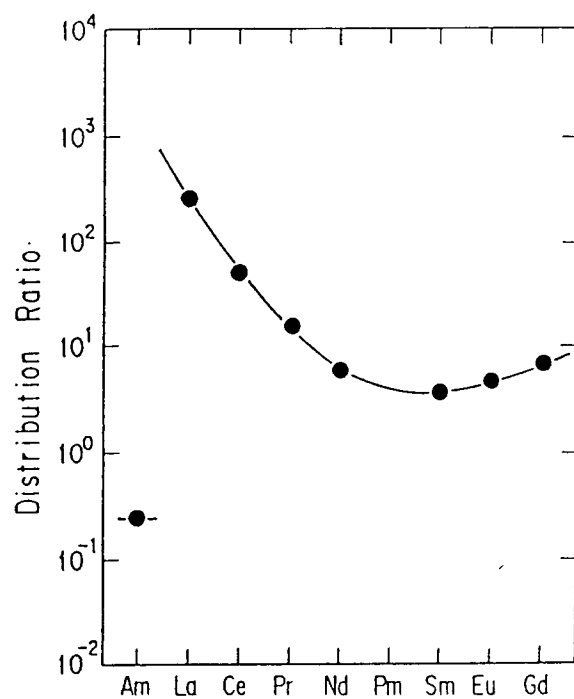


Fig.2 Comparison of Am distribution ratio with that of rare earths  
 Org. : 0.5 M DIDPA - 0.1 M TBP in n-dodecane  
 Aq. : 0.05 M DTPA - 1 M lactic acid - NH<sub>4</sub>OH, pH 3.6 after back-extraction

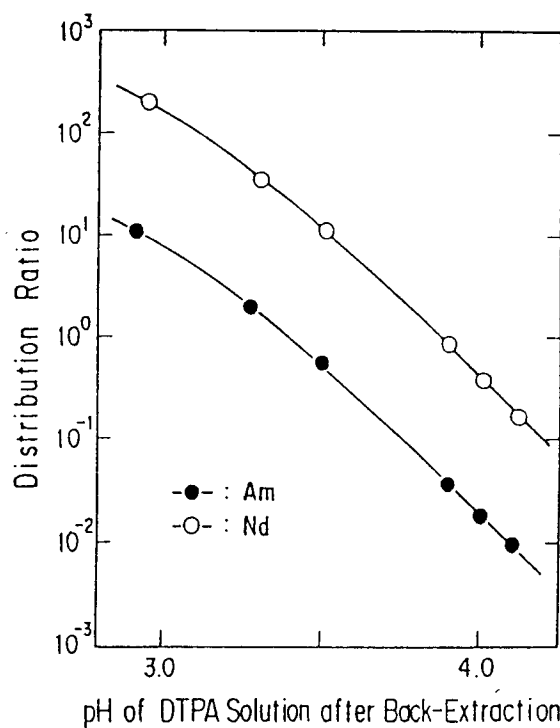


Fig. 3 pH dependence of distribution ratio of Am and rare earths in back-extraction from DIDPA solvent with DTPA  
 Org. : 0.5 M DIDPA - 0.1 M TBP in n-dodecane  
 Aq. : 0.05 M DTPA - 1 M lactic acid - NH<sub>4</sub>OH

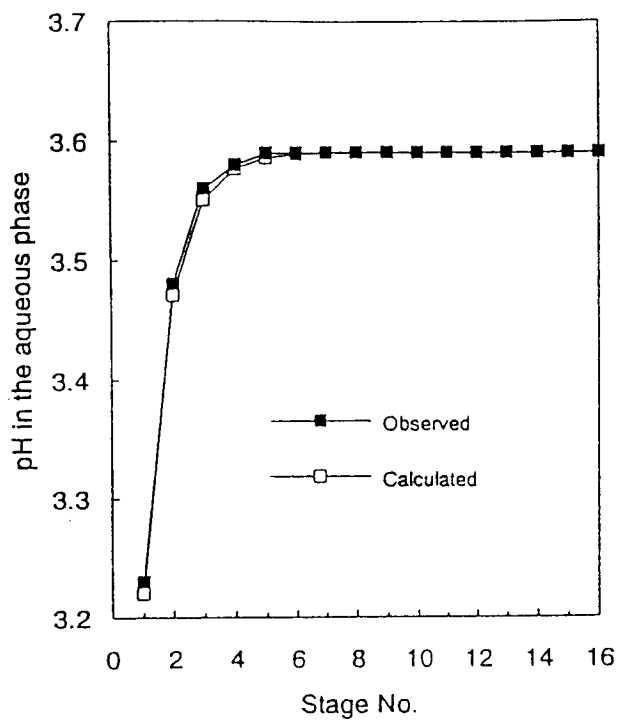


Fig. 4 pH profile in the back-extraction process with the DTPA solution

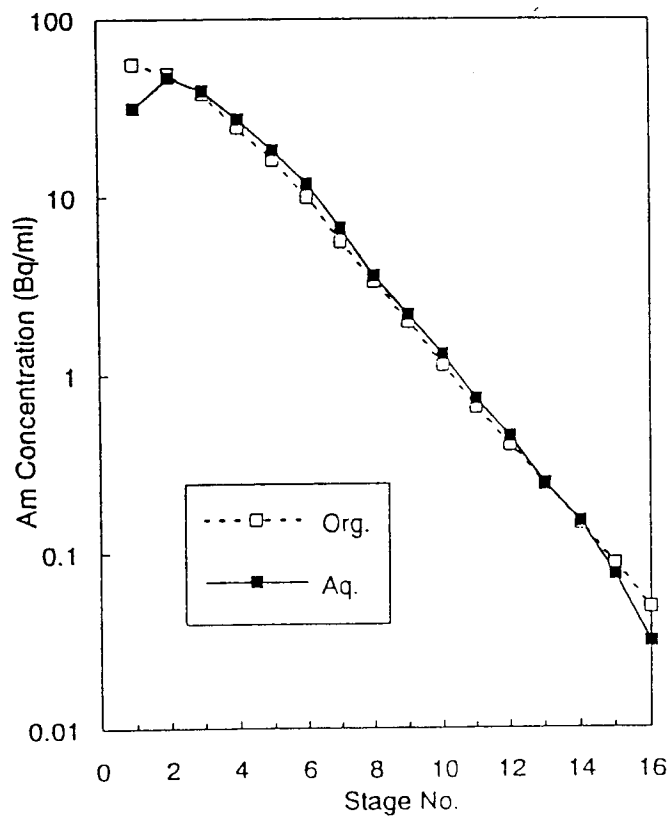


Fig. 5 Concentration profile of Am observed in continuous back-extraction with DTPA



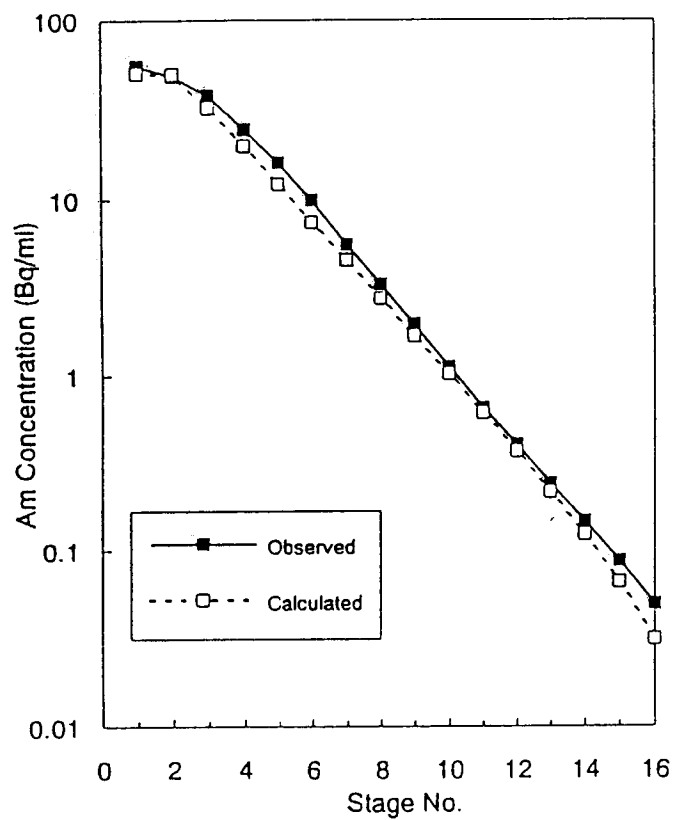


Fig. 6 Comparison of the observed and calculated concentration profiles of Am in the organic phase in continuous back-extraction with DTPA

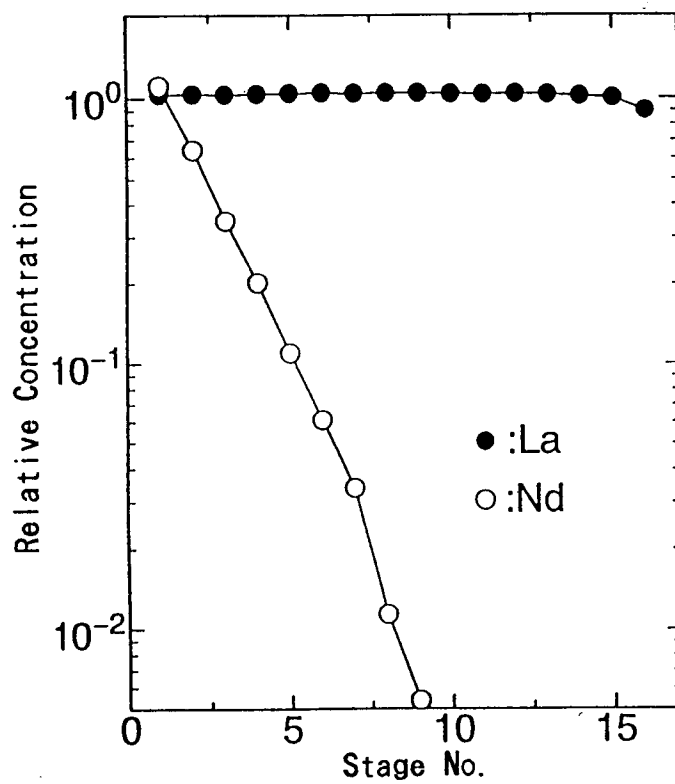


Fig. 7 Observed cocentration profiles of La and Nd in the organic phase in continuous back-extraction with DTPA

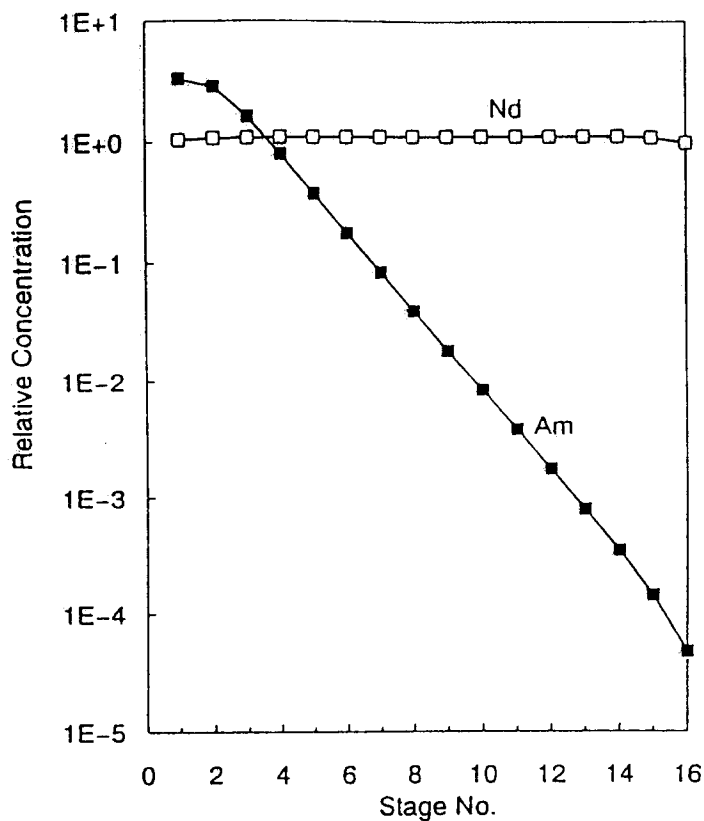


Fig. 8 Calculated concentration profiles of Am and Nd in the organic phase in continuous back-extraction with DTPA

Relative flow rate: Org. feed 100  
 DTPA soln. 100  
 Initial pH of DTPA soln.: 3.52

Table 2 Required temperature to avoid the formation of emulsion

Na <sub>2</sub> CO <sub>3</sub> (M)	Ethanol (v/o)	Temperature (°C)
0.5	0	> 70
1.5	0	> 65
1.5	5	> 45
1.5	10	> 30
1.5	15	> 15
0.5	15	> 25

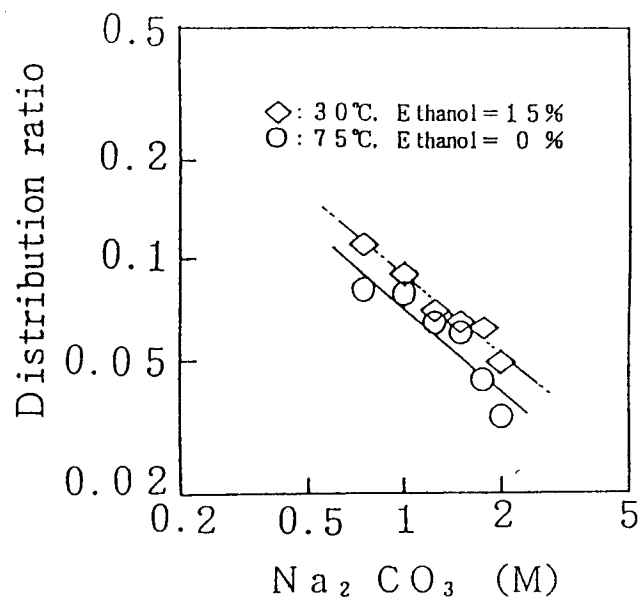


Fig. 9 Distribution ratio of U in back-extraction with Na<sub>2</sub>CO<sub>3</sub> from DIDPA solvent

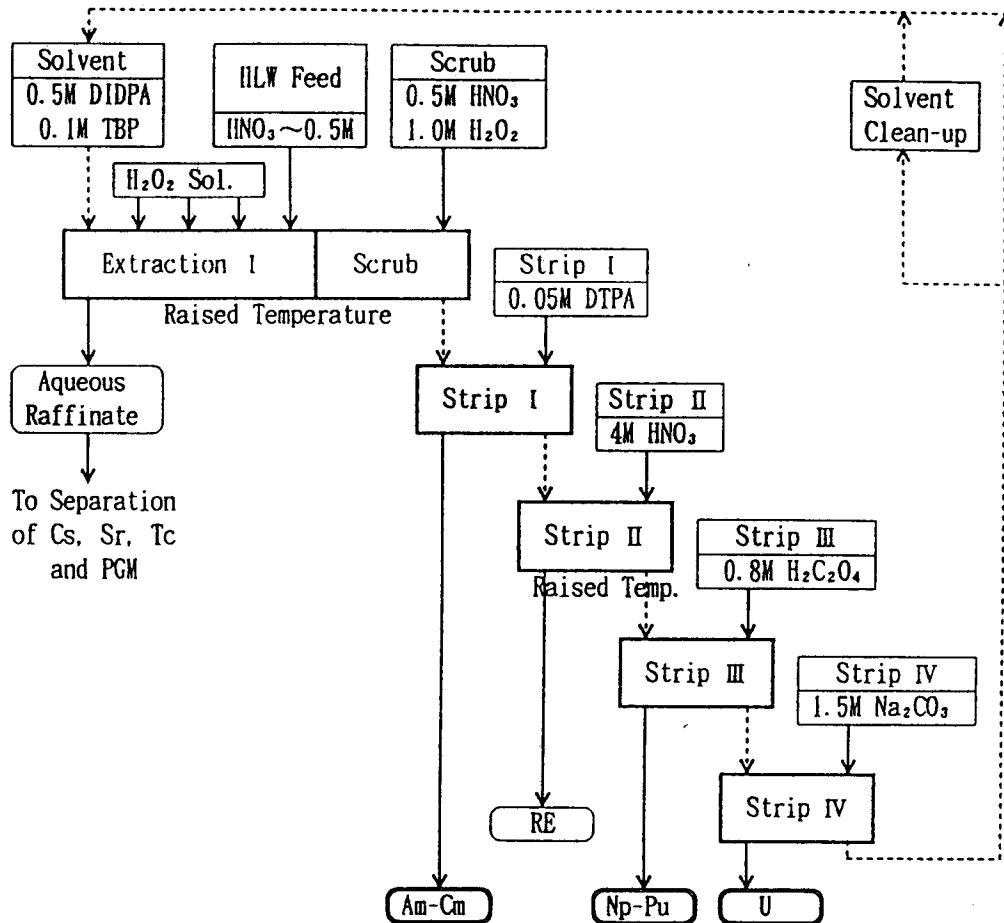


Fig. 10 Process flowsheet for TRU separation by extraction with DIDPA solvent followed by selective back-extraction