



**GOVERNMENT OF INDIA
ATOMIC ENERGY COMMISSION**

**COUNTER-CURRENT EXTRACTION STUDIES FOR THE RECOVERY OF
NEPTUNIUM BY THE PUREX PROCESS (PART II)**

by

N. Srinivasan, M. N. Nadkarni, S. V. Kumar, P. K. S. Kartha, R. R. Sonavane
Fuel Reprocessing Division

and

M. V. Ramaniah and S. K. Patil
Radiochemistry Division

**BHABHA ATOMIC RESEARCH CENTRE
BOMBAY, INDIA**

1974

GOVERNMENT OF INDIA
ATOMIC ENERGY COMMISSION

COUNTER-CURRENT EXTRACTION STUDIES FOR THE RECOVERY OF
NEPTUNIUM BY THE PUREX PROCESS (PART-II)

by

N. Srinivasan, M.N. Nadkarni, S.V. Kumer, P.K.S. Kertha, R.R. Sonavane
Fuel Reprocessing Division

and

M.V. Ramaniah and S.K. Petil
Radiochemistry Division

BHABHA ATOMIC RESEARCH CENTRE
BOMBAY, INDIA
1974

INIS subject category: B16

Descriptors:

PUREX PROCESS

COUNTER CURRENT

SOLVENT EXTRACTION

MIXER-SETTLERS

IRON COMPOUNDS

REDUCTION

NEPTUNIUM237

NEPTUNIUM COMPLEXES

URANIUM COMPLEXES

FLWSHEETS

ABSTRACT

Counter-current extraction experiments were carried out under the conditions relevant to the partitioning column (IBX) in the purax process to know the path of neptunium present as Np (VI) the organic phase during the partitioning step. The results obtained show that when Ferrous Sulphamate is used as the reducing agent, most of the neptunium continues to remain with uranium in the organic stream while with hydrazine stabilized uranous nitrate as the reducing agent, a major fraction of neptunium follows the aqueous stream. Mixer-settler experiments were also carried out under the conditions relevant to the uranium purification cycle (2D) to establish the conditions for forcing neptunium to the aqueous raffinate or for partitioning it from uranium if both neptunium and uranium are coextracted in this cycle and the results obtained are reported here.

COUNTER-CURRENT EXTRACTION STUDIES FOR THE RECOVERY OF NEPTUNIUM BY THE PUREX PROCESS

by

N. Srinivasan, M.N. Nadkarni, S.V.Kumar, P.K.S.Kartha, R.R.Sonavane
M.V. Ramaniah & S.K. Patil

PART - II

1. INTRODUCTION

Neptunium-237 is formed in nuclear reactors fuelled with natural or enriched uranium. This can be recovered as a valuable byproduct during the reprocessing of irradiated fuels. Neptunium-237 has assumed great importance recently as it is the starting material for the production of ^{238}Pu which is one of the most suitable isotopes used as heat source in the radio-isotopic power sources. Several kilograms of ^{237}Np are routinely recovered at present in the American reprocessing plants.

In the purex process, neptunium will accompany fission products, uranium or plutonium or be distributed between these three depending on the conditions selected or imposed. It was, however, felt that it would be most convenient to recover neptunium from the raffinate in the uranium purification cycle (2D cycle) as the fission product activity in this stream would be relatively low. It was, therefore, necessary to establish the conditions so that ^{237}Np is coextracted along with uranium and plutonium in HA and IA cycles of the purex process. The experimental data obtained to enable the coextraction of neptunium in HA and IA cycles are described in the earlier report ⁽¹⁾. In the present report the data are presented on the partitioning conditions of neptunium under the conditions relevant to 19X column conditions. The experimental work carried out to establish the conditions for forcing neptunium to the raffinate in the uranium purification cycle (2D cycle) or for partitioning it from uranium if both ^{237}Np and uranium are coextracted in this cycle and the results obtained are also reported.

2. MIXER-SETTLER UNIT

A 12-stage stainless steel permpox minimixer settler unit of Eurochemic type⁽²⁾ fabricated in Trombay was used in the present studies. The unit is described in the earlier report⁽¹⁾.

3. EXPERIMENTAL

Neptunium-237 was purified from its daughter ^{233}Pa extraction of the latter from 7M nitric acid into 0.5M TTA in xylene. The purified ^{237}Np was spiked with radiochemically pure ^{239}Np or ^{238}Np to enable the estimation of neptunium by gamma counting. Neptunium-239 for this purpose was periodically prepared from neutron irradiated natural uranium. Neptunium-238 used in a few experiments was prepared by neutron irradiation of ^{237}Np and separated from the fission products by TTA extraction⁽³⁾. The radiochemical purity of both ^{238}Np and ^{239}Np was checked by their half-life determination as well as the gamma-ray spectra. The mixture of ^{239}Np or ^{238}Np and ^{237}Np was then reduced to Np(IV) by ferrous sulphamate ($\approx 0.01\text{M}$) and was then added to uranium solution with known uranium and nitric acid concentration to prepare the feed, for the extraction experiments. The feed for the extraction experiments for Np(VI) was prepared in a similar way using 0.01M $\text{K}_2\text{Cr}_2\text{O}_7$ as the holding oxidant.

For the partitioning experiments, Np(VI) was prepared by oxidizing a mixture of ^{238}Np and ^{233}Pa -free ^{237}Np in dilute nitric acid by KMnO_4 . The Np(VI) was then extracted into 30% TBP in Shell Sol-T and the TBP solution containing Np(VI) was added to 30% TBP with a known concentration of uranium to prepare the feed for the partitioning experiment. The feed solution for the partitioning of Np(IV) from U(VI) was prepared in a similar way starting from Np(IV) obtained by reducing with ferrous sulphamate.

Uranyl nitrate solutions used were prepared by dissolving U_3O_8 or UO_3 in nitric acid. Uranium was estimated spectrophotometrically using the thiocyanate method⁽⁴⁾. The concentration of nitric acid in the uranium solution was determined by titration against standard sodium hydroxide solution after complexing uranium by oxalate⁽⁵⁾.

Uranium(IV) solution was prepared by the electrolytic reduction of uranyl nitrate solution in nitric acid containing hydrazine as a stabilizer using mercury cathode⁽⁶⁾. The estimation of U(IV) was carried out by reducing Fe(III) to Fe(II) by U(IV) and the resulting Fe(II) was estimated as O-phenanthroline complex⁽⁷⁾. Ferrous sulphamate was estimated⁽⁸⁾ volumetrically by titration against standard ceric sulphate solution and sometimes by spectrophotometry as O-phenanthroline complex⁽⁷⁾. Hydrazine was estimated colorimetrically⁽⁸⁾.

30% TBP in Shellisol-T was used in all the experiments after pre-equilibrating it with 2M nitric acid. The initial neptunium concentration in the feed was usually about 1-3/ μ g/ml. Other experimental details were same as described in the earlier report⁽¹⁾.

4. RESULTS AND DISCUSSION

When ferrous sulphamate is used as a reducing agent for the partitioning of plutonium from uranium, it is known that the coextracted neptunium-237 [$\sim$ as Np(VI)] could be made to follow either plutonium in the aqueous stream or uranium in the organic stream by careful choice of nitric acid concentration⁽¹⁰⁾. In the Tarapur Plant, U(IV) is proposed as a reducing agent for plutonium in the place of ferrous sulphamate. The reduction of Np(VI) to Np(V) by U(IV) is fast while further reduction of Np(V) to Np(IV) is slow⁽¹¹⁻¹³⁾. The use of U(IV) as a reducing agent can very much influence the path of neptunium during partitioning. A few experiments were carried out to study the same.

For the studies relevant to the uranium purification cycle (20-cycle) it was believed that the addition of oxidizing or reducing agent to the feed to enable the recovery of neptunium was compatible with the process as the plutonium main product was not involved in this cycle. Moreover, for the decontamination of uranium product in this cycle from residual plutonium contamination, addition of excess of ferrous sulphate to reduce plutonium to non-extractable trivalent state is usually followed. This causes the reduction of neptunium to Np(IV) and hence more emphasis was placed on the extraction behaviour of Np(IV) rather than Np(VI) in the present studies.

4.1 Path of Neptunium Under Partitioning Column Conditions

The results of the experiments carried out to know the behaviour of neptunium, present in the organic feed solution as Np(VI) under the partitioning column conditions with U(IV) as the reducing agent are given in Tables 1 and 2 and the corresponding neptunium and uranium profiles are shown in Figs 1 and 2. The concentration of U(IV) and the flow ratio used in these experiments are comparable to those proposed to be used in the Tarapur Plant. It is seen from these data that 50-65% of neptunium follows the aqueous stream and this is expected as the reduction of Np(V) to Np(IV) by U(IV) is slow. The reduction of Np(V) to Np(IV) by U(IV) is known to be favoured by exposure to light. In the present experiments although sufficient precautions were taken to limit the exposure to light, complete protection against it was not possible. The fraction of neptunium following the aqueous stream in the partitioning column would depend on several factors such as nitric acid and U(IV) concentrations, uranium saturation of the organic phase, temperature and residence time. Koch carried out the mixer settler experiments to study the path of neptunium during partitioning and observed⁽¹⁴⁾ that 7 to 70% neptunium accompanied the aqueous stream containing plutonium.

Two counter-current runs were also carried using ferrous sulphate as the reducing agent in a similar way. The results of these experiments are given in Tables 3 and 4 the corresponding neptunium and uranium profiles are shown in Figs 3 and 4. The reduction of Np(VI) to Np(IV) by Fe(II) is quite rapid and most of the Np(IV) thus formed is seen to continue to follow uranium in the organic stream. These results are in good agreement with those reported by Lewis⁽¹⁰⁾.

It can be concluded from these results that if U(IV) is used as a reducing agent in the partitioning column, neptunium will be distributed in both uranium and plutonium streams assuming neptunium to be present as Np(VI) in the entering organic stream containing Pu(IV) and U(VI) . Under similar conditions if Fe(II) is used as the reducing agent, most of the neptunium can be made to follow the organic stream containing uranium.

4.2 Behaviour of Neptunium under Uranium Purification cycle (20-cycle) conditions

Since it was felt that it would be most convenient to recover neptunium from the raffinate of the uranium purification cycle, it was necessary to establish the conditions required to force neptunium to the raffinate in this cycle or to partition it from uranium if both neptunium and uranium are coextracted. Although in the Hanford Plant⁽¹⁵⁾ neptunium is forced into the raffinate in this cycle, no details regarding feed acidity, uranium saturation etc. are available.

All but one counter-current experiment were carried out with Np(IV) only for the reasons explained earlier. Neptunium-239 was used in these experiments and the gamma activity of various samples was not corrected for the gamma activity due to uranium and its decay products which was usually less than 10% of ^{239}Np activity.

The results of the only experiment carried out on the extraction of Np(VI) are summarised in Table 5 and the corresponding

neptunium and uranium extraction profiles are shown in Fig.5. It is evident from these data that Np(VI) is quantitatively extracted in three stages from 2M nitric acid solution at uranium concentration of 85-90 g/l in the organic phase.

It is known that the extraction of Np(IV) is decreased by using low nitric acid concentration and by keeping high uranium concentration in the organic phase. A mixer-settler experiment was carried out with 1M nitric acid both in the feed and the scrub keeping the uranium concentration in the organic phase at about 80 g/l. Under these conditions, Np(IV) was expected to follow the aqueous raffinate. However, Np(IV) was found to reflux in the unit following neither the aqueous raffinate nor the uranium loaded organic stream. The run was continued for 25 hours. However this did not help in moving Np(IV) to the aqueous raffinate or the organic stream as is seen from the data given in Table 5. The corresponding neptunium extraction profile shown in Fig 6 clearly shows that neptunium is getting refluxed in the unit.

To avoid the refluxing of Np(IV) the extraction experiment was repeated at a higher concentration of nitric acid in the feed and the scrub and by decreasing the uranium loading of the organic phase. The data obtained are given in Table 7 and the corresponding neptunium and uranium extraction profiles are shown in Fig 7. It is seen from these data that under these conditions, Np(IV) is quantitatively extracted into TBP along with uranium. In the Tarapur Plant two scrubs are proposed in the 2D extraction column. A counter-current extraction experiment was carried out using two scrubs, as was relevant to 2D extraction column, and the data obtained are given in Table 8 and the corresponding profiles of neptunium, uranium and nitric acid are shown in Fig.8. It is seen from these data that Np(IV) is extracted quantitatively under these conditions also.

Assuming that Np(IV) and U(VI) are coextracted, experiments were carried out to establish the conditions to partition Np(IV)

from U(VI). This was attempted by scrubbing the uranium loaded organic phase containing Np(IV) with 0.5M and 1.0M nitric acid keeping 1:1 flow ratio and using 12 stages. It was observed that although Np(IV) followed the aqueous stream quantitatively, U(VI) losses to the aqueous raffinate were too high ($\approx 50\%$ with 0.5M HNO_3 and $\approx 35\%$ with 1M HNO_3 scrub) to be tolerated. The experiments were repeated using a smaller number of stages for partitioning and by changing the flow ratio. The results of an experiment with 1M HNO_3 as the scrub are given in Table 9 and the corresponding neptunium and uranium profiles in Fig 9. It is seen from these data that Np(IV) follows the aqueous stream almost quantitatively ($\approx 90\%$) and U(VI) accompanying Np(IV) to the aqueous stream is only about 7-8%. In a similar experiment with 0.5M nitric acid scrub, uranium losses to the aqueous stream were found to be about 10-11% although the Np(IV) partitioning was slightly more quantitative.

The results of the mixer-settler runs to force Np(IV) to the aqueous raffinate under 2D cycle conditions are given in Tables 10 and 11 and the corresponding extraction profiles are shown in Figs 10 and 11. It is seen from these data that by using 0.3M HNO_3 in the feed and 1-2M HNO_3 in the scrub and keeping the organic phase uranium loading about 85 g/l, most of the neptunium present as Np(IV) can be forced with the aqueous raffinate. It should however be noted that when 2M HNO_3 is used as the scrub the number of stages required for quantitative extraction of uranium can be limited to 4 whereas with 1M HNO_3 as the scrub about 6 stages are required for quantitative extraction of uranium.

Optimum chemical flow-sheet conditions for 2D cycle either to force Np(IV) to the aqueous raffinate or to partition it from U(VI) if both Np(IV) and U(VI) are coextracted can be chosen from these data.

4.3 Study of the Co-stability of Np(IV) and Pu(IV)

It is known that when U(IV) is used as the reducing agent in the partitioning column, a major fraction of neptunium accompanies plutonium to the aqueous stream assuming neptunium to be present as Np(VI) in the uranium loaded organic stream entering the partitioning column. This necessitates the recovery of neptunium from the plutonium purification cycle (2A-cycle) by forcing it to the aqueous raffinate. However if neptunium is present in the loaded organic stream entering the partitioning column as Np(IV), it can be made to follow uranium in the organic stream by a careful choice of nitric acid concentration irrespective of the reducing agent used for partitioning.

Champion and Cheene established ⁽¹⁶⁾ the conditions for stabilising Np(IV) and Pu(IV) together by controlling the Fe^{++}/Fe^{+++} ratio in the solution. A few preliminary experiments were carried out to study the stability of Np(IV) and Pu(IV). The data obtained showed that only a small fraction of Np(IV) was oxidized by Pu(IV). It was recently reported ⁽¹⁷⁾ that under the conditions relevant to the Pured Process at room temperature ($\approx 25^{\circ}C$) more than 80% Np(IV) remains unoxidized in the presence of Pu(IV) upto 8 hours. Thus the feed to 1A cycle can be conditioned to have both Np(IV) and Pu(IV) and subsequently the extracted Np(IV) can be made to follow uranium in the organic stream during partitioning even though U(IV) is used as the reducing agent.

5. CONCLUSIONS AND PROPOSED CHEMICAL FLOWSHEET CONDITIONS

It is considered that most of the relevant aspects of the process chemistry of neptunium are investigated in the present studies. From the data reported here and in the earlier report ⁽¹⁾ it can be said that by controlling the conditions it is possible to recover neptunium by its coextraction in the first (1A) and second (2A) cycles and forcing it to the raffinate (2DU) in the uranium purification cycle.

The schematic flowchart of the purex process to be used at Tarapur Plant is shown in Fig 12 and the conditions, for optimum recovery of neptunium, proposed are given below.

5.1 HA and 1A Column

(a) The following conditions may be used when nitrous acid is used to enhance the oxidation of Np(V) to Np(VI) to enable the coextraction of neptunium.

$$\begin{aligned}(\text{HNO}_3)_{\text{Feed}} &\geq 2.5 \text{ M} \\(\text{HNO}_3)_{\text{Scrub}} &\approx 3 \text{ M} \\(\text{U})_{\text{Org}} &= 70 \text{ g/l} \\(\text{HNO}_2)_{\text{Org}} &= 10^{-3} - 10^{-2} \text{ M}\end{aligned}$$

(b) When Vanadium (V) is used to oxidize Np(V) to Np(VI) which is subsequently coextracted.

$$\begin{aligned}(\text{HNO}_3)_{\text{Feed}} &\approx 2 \text{ M} \\(\text{HNO}_3)_{\text{Scrub}} &\approx 3 \text{ M} \\(\text{V(V)}) &= 10^{-3} - 10^{-2} \text{ M} \\(\text{U})_{\text{Org}} &= 70 \text{ g/l}\end{aligned}$$

It is preferable to introduce vanadium (V) to the extraction column below the feed stage rather than adding it to the feed solution.

(c) When the feed is conditioned to adjust neptunium to Np(V) which is coextracted.

$$\begin{aligned}(\text{HNO}_3)_{\text{Feed}} &\geq 2 \text{ M} \\(\text{HNO}_3)_{\text{Scrub}} &\geq 2 \text{ M} \\(\text{U})_{\text{Org}} &= 75 \text{ g/l}\end{aligned}$$

The feed may be conditioned by adding ferrous sulphamate or preferably by adding U(IV) and after allowing 1-2 hrs. for the reduction of neptunium to Np(IV), the Pu(III) formed may be oxidized to Pu(IV) by adding NaNO_2 to prepare the conditioned feed for the extraction columns.

5.2 1BX Column

When ferrous sulphamate is used as the reducing agent, if the concentration of HNO_3 in the outgoing aqueous stream containing Pu(III) is about 2M or more, most of the neptunium ($\approx 90\%$) will remain with uranium in the organic stream.

When U(IV) is used as the reducing agent and the conditions for 1A column are as described either in 5.1(a) or 5.1(b), major fraction of neptunium will accompany plutonium stream. This however may be avoided and neptunium may be made to follow uranium in the partitioning column by conditioning 1AF as described in 5.1(c).

5.3 2D Column

Neptunium present as Np(IV) may be coextracted with uranium under the following conditions and the extracted neptunium may be separated from uranium by 0.5M or 1M nitric acid scrub in an additional scrub column.

$$\begin{aligned} (\text{HNO}_3)_{\text{Feed}} &\geq 2 \text{ M} \\ (\text{HNO}_3)_{\text{Scrub}} &\geq 2 \text{ M} \\ (\text{U})_{\text{Org}} &\approx 75 \text{ g/l} \end{aligned}$$

Alternately Np(IV) may be forced to the raffinate (2DW) by using the following conditions:

$$\begin{aligned} (\text{HNO}_3)_{\text{Feed}} &\approx 0.3 \text{ M} \\ (\text{HNO}_3)_{\text{Scrub}} &= 1-2 \text{ M} \\ (\text{U})_{\text{Org}} &\approx 05 \text{ g/l} \end{aligned}$$

Neptunium separated from the uranium in this cycle can be concentrated and purified by ion exchange technique.

From the chemical flowsheet data used in the Plutonium Plant at Trombay⁽¹⁸⁾, it appears that the proposed chemical flowsheet conditions are quite compatible with the process. It is hoped that these data will prove useful to recover a valuable byproduct neptunium-237, along with plutonium and uranium in the fuel reprocessing plant at Tarapur.

References

- (1) N. Srinivasan, et al - BARC-734
- (2) E. Lopez-Menchero and J. Conteno - ETR-112 (1962)
- (3) F.L. Moore - Anal. Chem., 29 941 (1957)
- (4) ORNL Master Manual TID-7015 - Method No.1 - 215212
- (5) Ibid Method No.9 - 042200
- (6) N. Srinivasan et al - BARC-375 (1968)
- (7) E.N. Jenkins and R.J.W. Streaton - AERE-R-3158 (1959)
- (8) A.I. Vogel 'A Text-book of Quantitative Inorganic Analysis' 3rd Ed. P.322 (1961) Longmans.
- (9) C.W. Watts and J.D. Chirisp . Anal. Chem. 24 12 (1952)
- (10) W.H. Lewis "UN Int. Conference on Peaceful Uses of Atomic Energy" - Geneva Vol.17 236 (1958)
- (11) J.C. Sullivan, A.J. Zielen and J.C. Hindman J. - Am.Chem.Soc. 82 8288 (1960)
- (12) N.K. Sastri, E.S. Amis and J.O. Wear - J.Inorg.Nucl. Chem., 27 2413 (1965).
- (13) L. Salomon, E. Lopez-Menchero, J. Lopez de Monterola and G.G. Leynen - ETR-253 (1968)
- (14) G. Koch - KFK-976 (1969)

- (15) R.E. Isaacson and B.F. Judeon - Ind. Engg. Chem., Process Design Develop. 3 296 (1964)
- (16) J. Champion and A. Cheene - CEA-R-2607 (1964)
- (17) S.K. Patil, V.V. Ramakrishna and G.V.N. Avachany - DAE Chem. Symposium, Aligarh (1972)
- (18) N. Srinivasan, M.V. Rameniah, S.K. Patil and V.V. Ramakrishna - "J. Radioanal. Chem. 8 223 (1971).

Table 1

PATH OF NEPTUNIUM DURING PARTITIONING

Feeds:
 (Uranium loaded organic solvent) $[HNO_3] = 0.3 M$
 $[U] = 65 \text{ mg/ml}$
 $[^{238}Np] = 3.6 \times 10^4 \text{ C/50 Sec/ml}$

Reducing scrub:
 $[U(IV)] = 7 \text{ mg/ml}$
 $[N_2H_4] = 0.06 M$
 $[HNO_3] = 2 M$

Flow rates:
 (ml/min) Feed $[org] = 6.7$
 Reducing scrub = 0.9

No. of stages = 5

Time hrs.	Concentration of HNO_3 in the outgoing Aq stream	^{238}Np Activity in the outgoing streams $C/50 \text{ Sec/ml} \times 10^{-5}$		Concentration of Uranium in the out- going streams (mg/ml)	
		Aq	Org	Aq	Org
14	2.6	1.20	0.15	6.9	62
15	-	1.04	0.16	7.8	62
16	2.6	1.12	0.19	7.7	63
17	-	1.07	0.19	8.4	63
18	2.4	1.14	0.19	7.9	62
19	-	1.21	0.19	8.1	63
20	2.4	1.24	0.19	9.1	63

% of Neptunium following the organic stream = 48 - 53%

Table 2

PATH OF NEPTUNIUM DURING PARTITIONING

Feeds:	(HNO ₃)	= 0.3 M
(Uranium loaded organic solvent)	(U)	= 62 mg/ml
	(²³⁸ Np)	= 5.4 x 10 ⁴ C/50 sec/ml
Reducing scrub:	[U(IV)]	= 6.7 mg/ml
	(N ₂ H ₄)	= 0.06 M
	(HNO ₃)	= 2 M
Flow rates (ml/min):	Feed (org)	= 6.7
	Reducing scrub	= 0.7
	No. of stages	= 5

Time hrs.	²³⁸ Np activity in the outgoing streams C/50 sec/ml x 10 ⁻⁵		Concentration of Uranium in the outgoing streams (mg/ml)	
	Aq	Org	Aq	Org
15	2.39	0.16	9.0	59
16	3.09	0.18	9.5	59
17	2.92	0.19	9.5	60
18.30	3.37	0.21	10.0	58
19.30	3.25	0.22	9.8	61
20	-	0.21	-	58
20.30	-	0.21	-	58
21	-	0.21	-	57
21.15	-	0.21	-	-

% of Neptunium following the organic stream = 36%

Table 3

PATH OF NEPTUNIUM DURING PARTITIONING

Feed: (Uranium loaded organic solvent)	(HNO ₃)	= 0.3 M
	(U)	= 64 mg/ml
	(²³⁸ Np)	= 2.00 x 10 ⁴ C/50 sec/ml
Reducing scrub:	(Fe ⁺⁺)	= 0.12 M
	(HNO ₃)	= 1 M
Flow rates (ml/min):	Feed (org)	= 6.7
	Reducing scrub	= 0.5
	No. of stages	= 5

Time hrs.	Concentration of HNO ₃ in the out- going Aq stream	²³⁸ Np activity in the outgoing streams C/50 sec/ml x 10 ⁻⁴		Concentration of Uranium in the out- going stream (mg/ml)	
		Aq	Org	Aq	Org
15	-	2.28	0.9	10.6	62
16	2.2	1.90	1.63	6.4	61
17	-	1.86	1.78	6.0	61
18	2.0	1.85	1.78	6.8	62
19	-	1.87	1.84	7.5	61
20	2.4	1.83	1.83	7.4	60

% of Neptunium following the organic stream = 87

Table 4

PATH OF NEPTUNIUM DURING PARTITIONING

Feeds:		(HNO ₃)	=	0.3 M
(Uranium loaded organic solvent)		(U)	=	64 mg/ml
		(²³⁸ Np)	=	1.92 x 10 ⁴ C/50 sec/ml
Reducing scrub:		(Fe ⁺⁺)	=	0.12 M
		(HNO ₃)	=	2 M
Flow rates (ml/min):		Feed (org)	=	6.7
		Reducing scrub	=	0.7
		No. of stages	=	5

Time hrs.	Concentration of HNO ₃ in the out- going aq stream	²³⁸ Np activity in the outgoing streams C/50 sec/ml x 10 ⁻⁴		Concentration of Uranium in the out- going streams (mg/ml)	
		Aq	Org	Aq	Org
12	-	1.64	1.77	5.4	56
13	-	1.54	1.79	5.2	56
14	2.4	1.51	1.76	5.0	56
15	2.6	1.45	1.80	5.0	61
16	-	1.46	1.79	4.9	60
17	2.6	1.44	1.78	5.0	60

% of Neptunium following the organic stream = 93

Table 5

EXTRACTION OF Np(VI)

Feed : (HNO₃) = 1.9 M
 (U) = 270 mg/ml
 (²³⁹Np) = 6.4 x 10⁴ c/min/ml

Flow rates (ml/min): Feed = 1.1
 Solvent = 3.35

Time hrs.	²³⁹ Np activity in the outgoing streams		Concentration of Uranium in the outgoing organic stream (mg/ml)
	Aqueous	Organic	
	C/min/ml x 10 ⁻⁴		
15.15	N11	1.84	-
16.30	N11	1.84	78.5
17.30	N11	2.06	88
18.30	N11	2.02	88
19.30	N11	2.04	84.5
20.45	N11	2.10	92
22.00	N11	1.90	81
23.00	N11	1.98	89.5

Table 6

EXTRACTION OF Np(IV) IN EXTRACTION SCRUBBING CONDITIONS

Feed :	(HNO_3)	=	1 M
	(U)	=	330 mg/ml
	(^{239}Np)	=	1.09×10^6 c/min/ml
Scrub:	(HNO_3)	=	1 M
Flow rate (ml/min):	Feed	=	1.9
	Scrub	=	1.0
	Solvent	=	6.2

Time hrs.	^{239}Np activity in the outgoing streams c/min/ml $\times 10^{-3}$		Concentration of Uranium in the outgoing organic stream (mg/ml)
	Aqueous	Organic	
21.00	N11	4.7	62.0
22.15	N11	3.5	-
00.10	N11	1.57	75.0
1.15	N11	1.17	-
2.20	N11	1.36	-
3.30	N11	1.34	-
4.30	N11	1.24	96.0
5.45	0.02	1.15	-
7.15	0.04	1.12	91.0
8.30	0.03	1.29	91.0
10.20	0.03	1.33	91.0
11.15	0.03	1.59	93.0

Table 7

COEXTRACTION OF Np(IV)

Feed:	(HNO ₃)	= 2.2 M
	(U)	= 286 mg/ml
	(²³⁹ Np)	= 1.05 x 10 ⁵ C/50 sec/ml
Scrub:	(HNO ₃)	= 2 M
Flow rate (ml/min):	Feed	= 1.8
	Scrub	= 0.9
	Solvent	= 7.2

Time hrs.	²³⁹ Np activity in the outgoing streams C/50 sec/ml x 10 ⁻⁴		Concentration of Uranium in the out- going organic stream (mg/ml)
	Aqueous	Organic	
11	0.21	2.8	84
12	0.12	1.9	73
13	0.07	3.8	73
14	0.06	3.2	73
15	0.06	2.9	78

Table 8
EXTRACTION OF Np(IV)

Feed:	(HNO ₃)	=	2.15 M
	(U)	=	286 mg/ml
	(²³⁹ Np)	=	1.44 x 10 ⁵ c/50 ccc/ml
Scrub-1	(HNO ₃)	=	3 M
Scrub-2		=	Water
Flow rates (ml/min):	Feed	=	1.8
	Scrub-1	=	0.9
	Scrub-2	=	0.4
	Solvent	=	7.0

Time hrs.	²³⁹ Np activity in the outgoing streams C/50 ccc/ml x 10 ⁻⁴		Concentration of Uranium in the out- going organic stream (mg/ml)
	Aqueous	Organic	
13.30	0.12	3.04	79.4
14.30	0.09	2.65	82.0
15.30	0.10	2.69	78.8
16.30	0.10	3.57	72.6
17.30	0.110	3.58	75.6

Table 9

PARTITIONING OF Np(IV) FROM U(VI)-LOADED ORGANIC

Feed :	(U)	=	73 mg/ml
(org.)	(²³⁹ Np)	=	4.7 x 10 ⁴ cpm/ml
Scrub:	(HNO ₃)	=	1 M
No. of stages		=	4
Flow rates (ml/min): Feed (org)		=	4.3
Scrub		=	1.0

Time hrs.	²³⁹ Np activity in the outgoing streams cpm/ml x 10 ⁻³		Concentration of Uranium in Aqueous raffinate (mg/pl)
	Aqueous	Organic	
15.30	141	5.7	25
16.30	175	7.9	25
17.40	165	8.1	27
19.10	168	8.1	25
20.00	158	7.7	27

Table 10

**BEHAVIOUR OF Np(IV) UNDER EXTRACTION--
SCRUBBING CONDITIONS**

Feed:	(HNO_3)	= 0.3
	(U)	330 mg/ml*
Scrub:	(HNO_3)	= 1 M
Flow rates (ml/min):	Feed	= 1.7
	Scrub	= 0.9
	Solvent	= 6.0

Time hrs.	²³⁵ Np activity in the outgoing stream C/50 sec/ml $\times 10^{-4}$		Concentration of Uranium in the outgoing organic stream (mg/ml)
	Aqueous	Organic	
13	8.1	0.12	88
14	8.1	0.12	85
15	8.6	0.12	84
16	8.6	0.13	86

* Calculated from concentration of uranium in the outgoing organic stream.

Table 11

**BEHAVIOUR OF Np(IV) UNDER EXTRACTION-SCRUBBING
CONDITIONS**

Feed:	(HNO ₃)	= 0.3 M
	(U)	330 mg/ml*
Scrub:	(HNO ₃)	= 2 M
Flow rates (ml/min):	Feed	= 1.7
	Scrub	= 0.9
	Solvent	= 6.2

Time hrs.	239 Np activity in the outgoing streams C/50 sec/ml $\times 10^{-4}$		Concentration of Uranium in the outgoing organic stream (mg/ml)
	Aqueous	Organic	
12	1.6	0.16	86
13	5.5	0.17	83
14	7.1	0.15	88
15	6.9	0.15	88
16	6.9	0.16	90
17	7.4	0.15	90
18	7.2	0.24	86

* Calculated from concentration of uranium in the
outgoing organic stream.

FIG.1.
PROFILE OF NEPTUNIUM DURING PARTITIONING
USING URANIUM(IV)

(FOR EXPERIMENTAL CONDITIONS REF. TABLE I.)

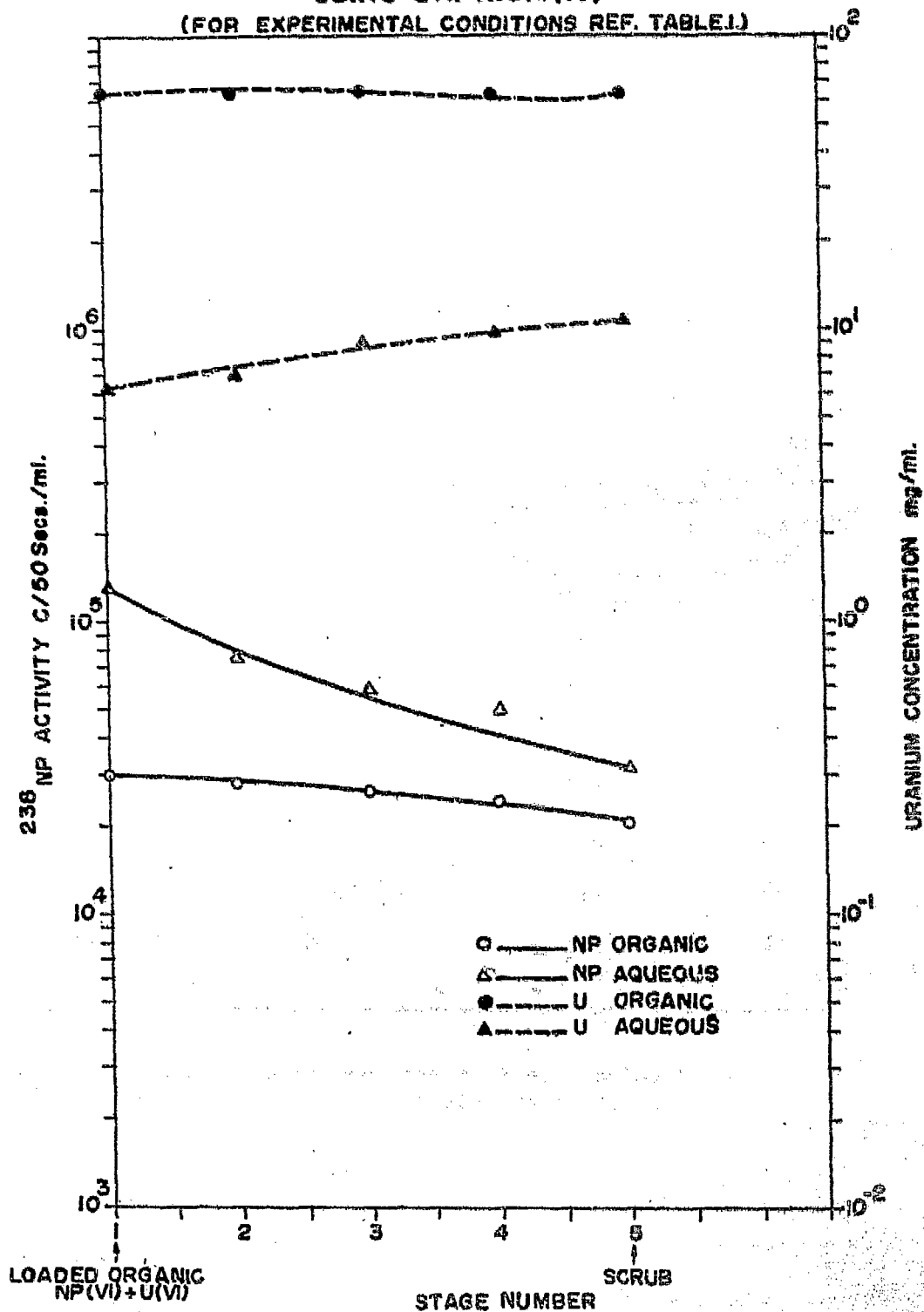


FIG. 2.
PROFILE OF NEPTUNIUM DURING PARTITIONING
USING URANIUM (IV)
 (FOR EXPERIMENTAL CONDITIONS REF. TABLE 2.)

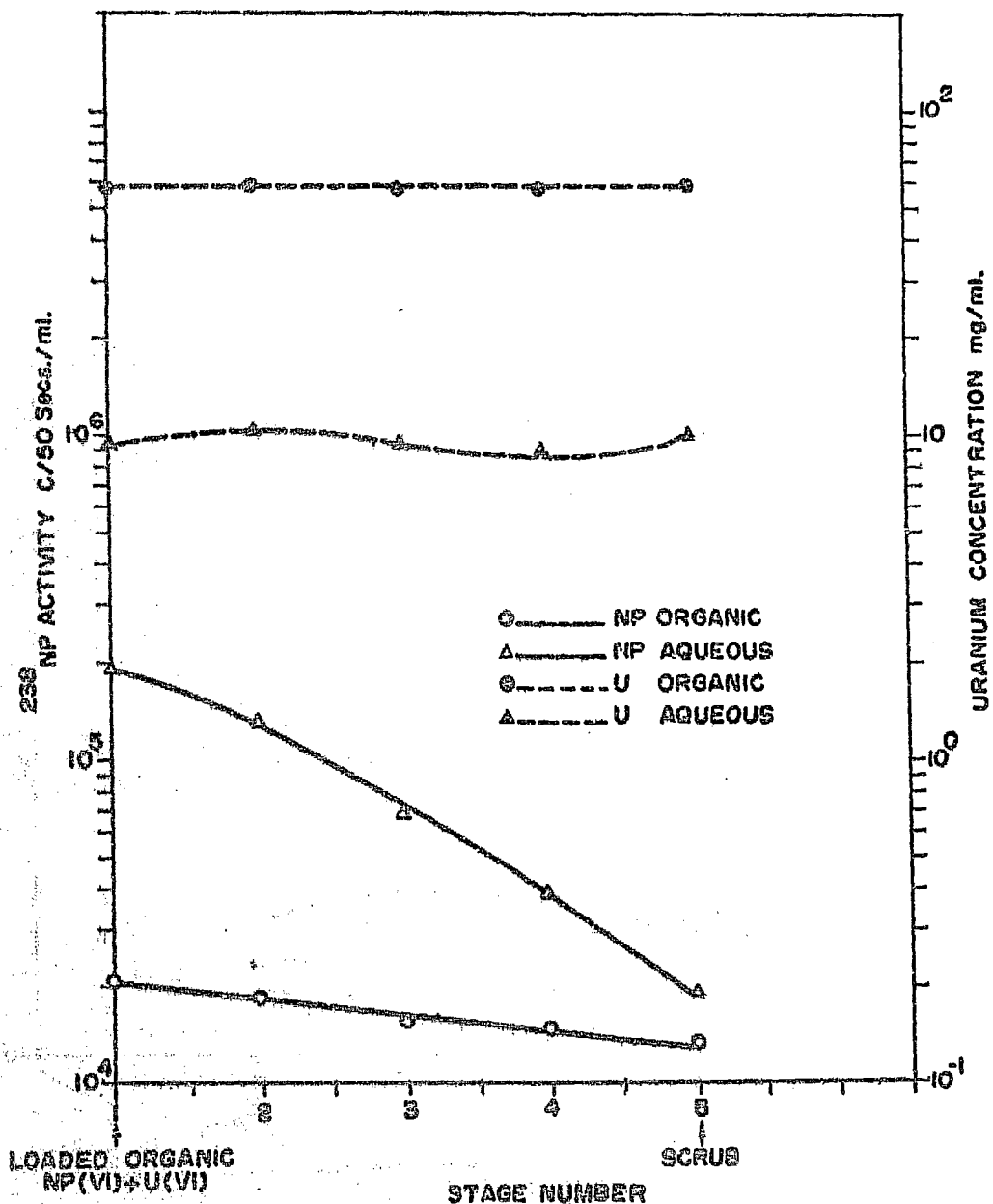


FIG. 3.
PROFILE OF NEPTUNIUM DURING
PARTITIONING USING F_2^{++}
(FOR EXPERIMENTAL CONDITIONS REF. TABLE 3.)

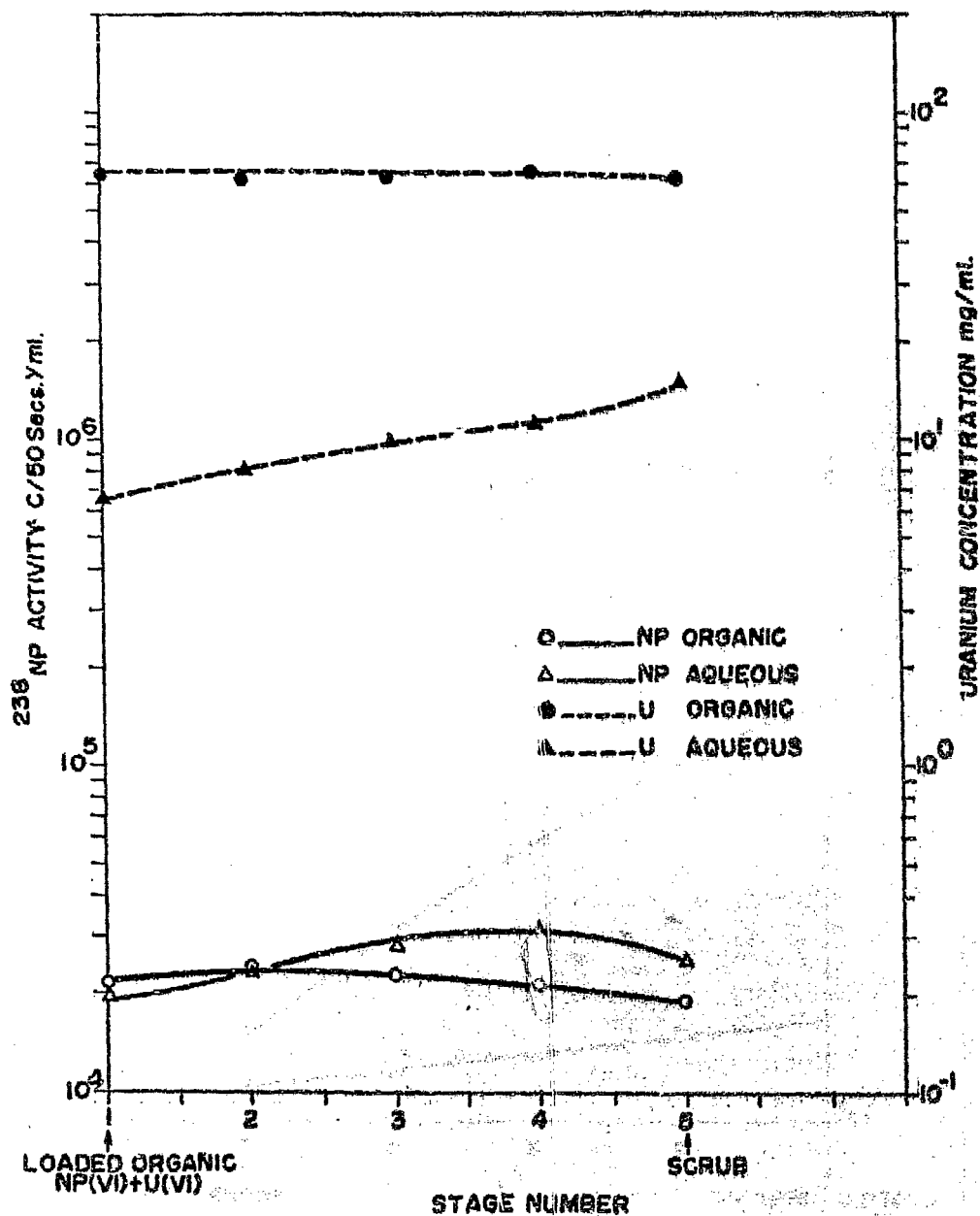


FIG. 4.
 PROFILE OF NEPTUNIUM DURING
 PARTITIONING USING F_0^{++}
 (FOR EXPERIMENTAL CONDITIONS REF. TABLE 4.)

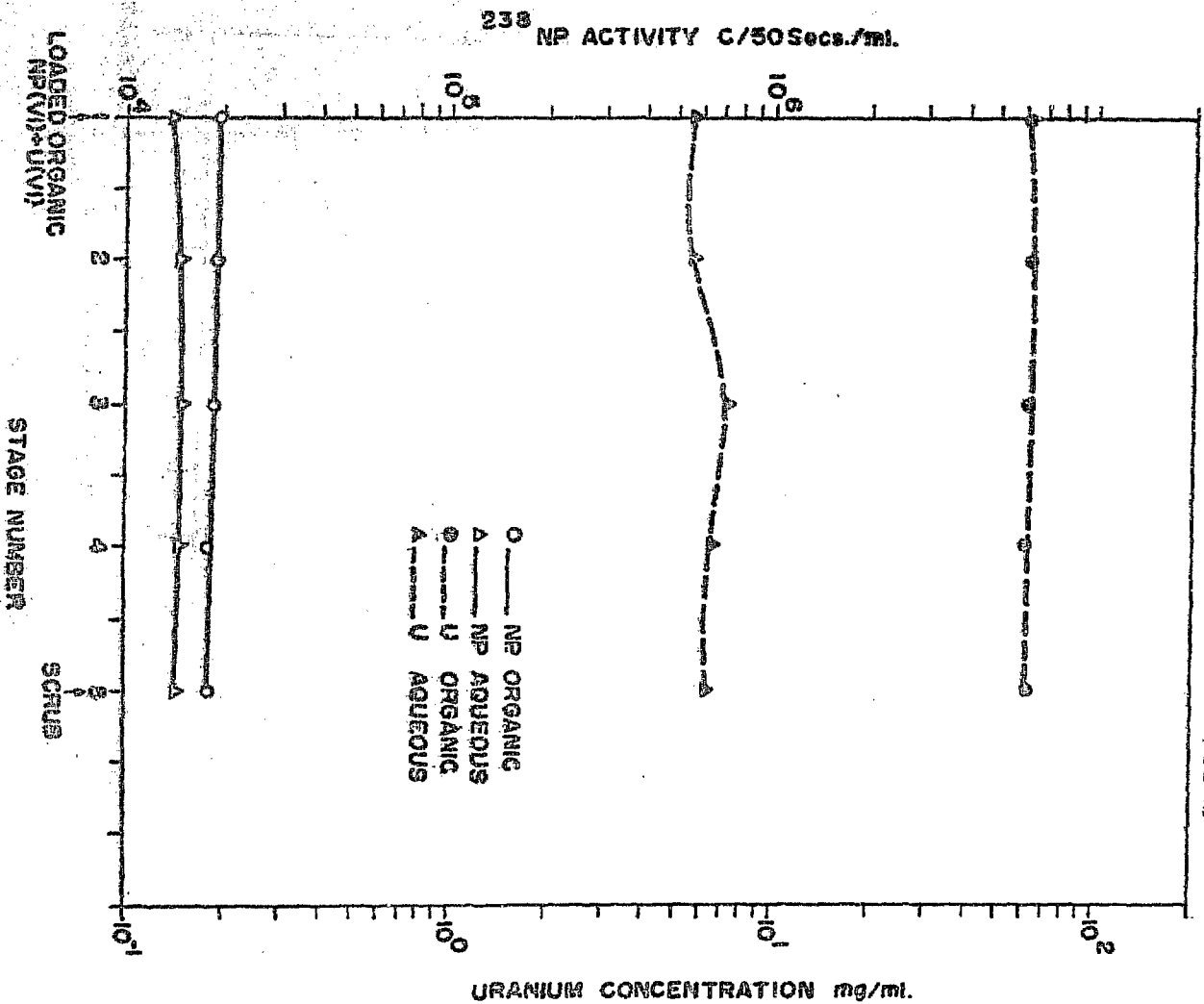


FIG.5.
PROFILE OF NEPTUNIUM(VI) AND URANIUM(VI)
IN THE EXTRACTION
 (FOR EXPERIMENTAL CONDITIONS REF. TABLE 5.)

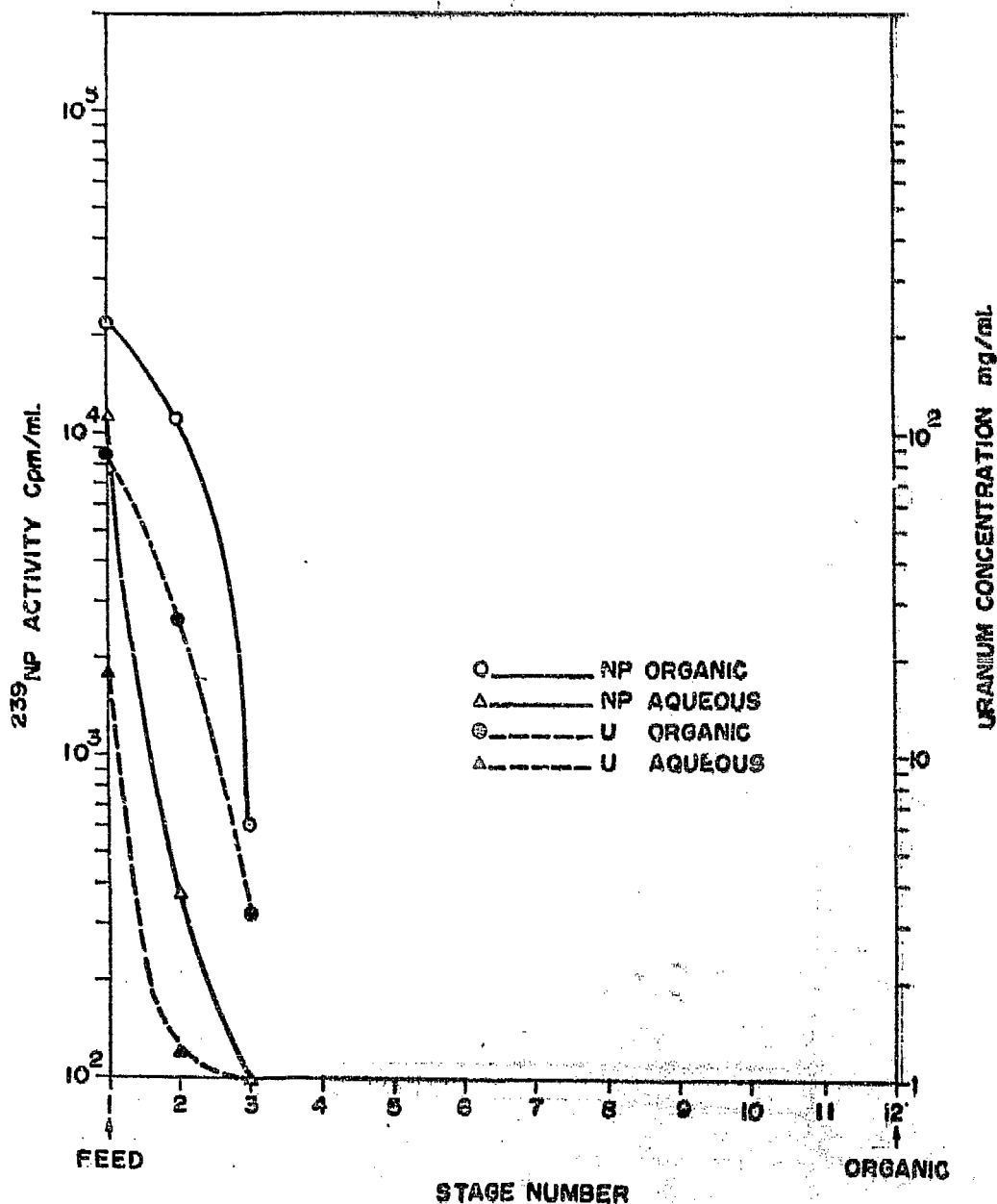


FIG.6.
PROFILE OF NEPTUNIUM (IV) IN THE EXTRACTION
 (FOR EXPERIMENTAL CONDITIONS REF. TABLE 6.)

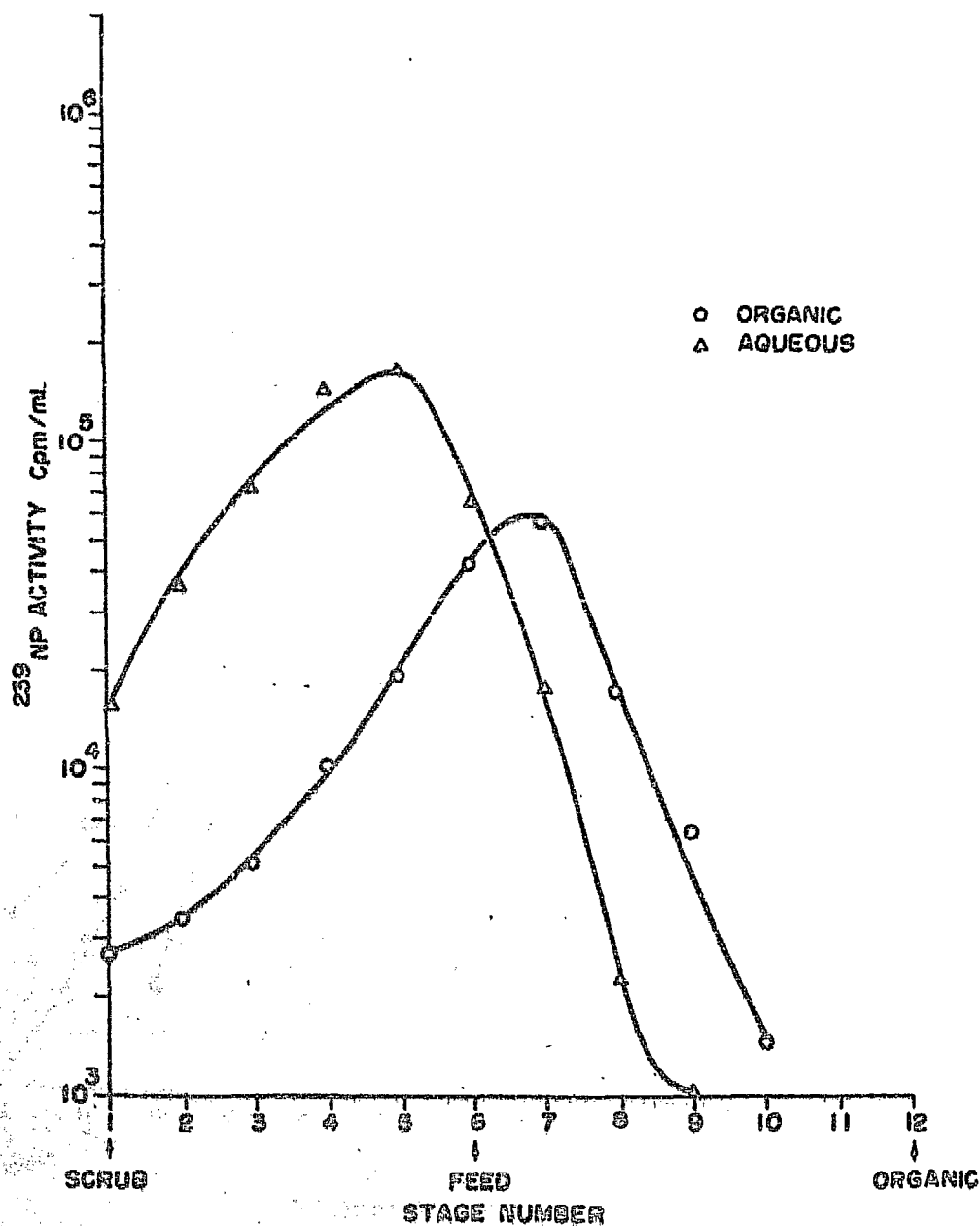


FIG. 7
PROFILE OF NEPTUNIUM(IV) AND URANIUM(VI) IN
THE EXTRACTION

(FOR EXPERIMENTAL CONDITIONS REF. TABLE 7.)

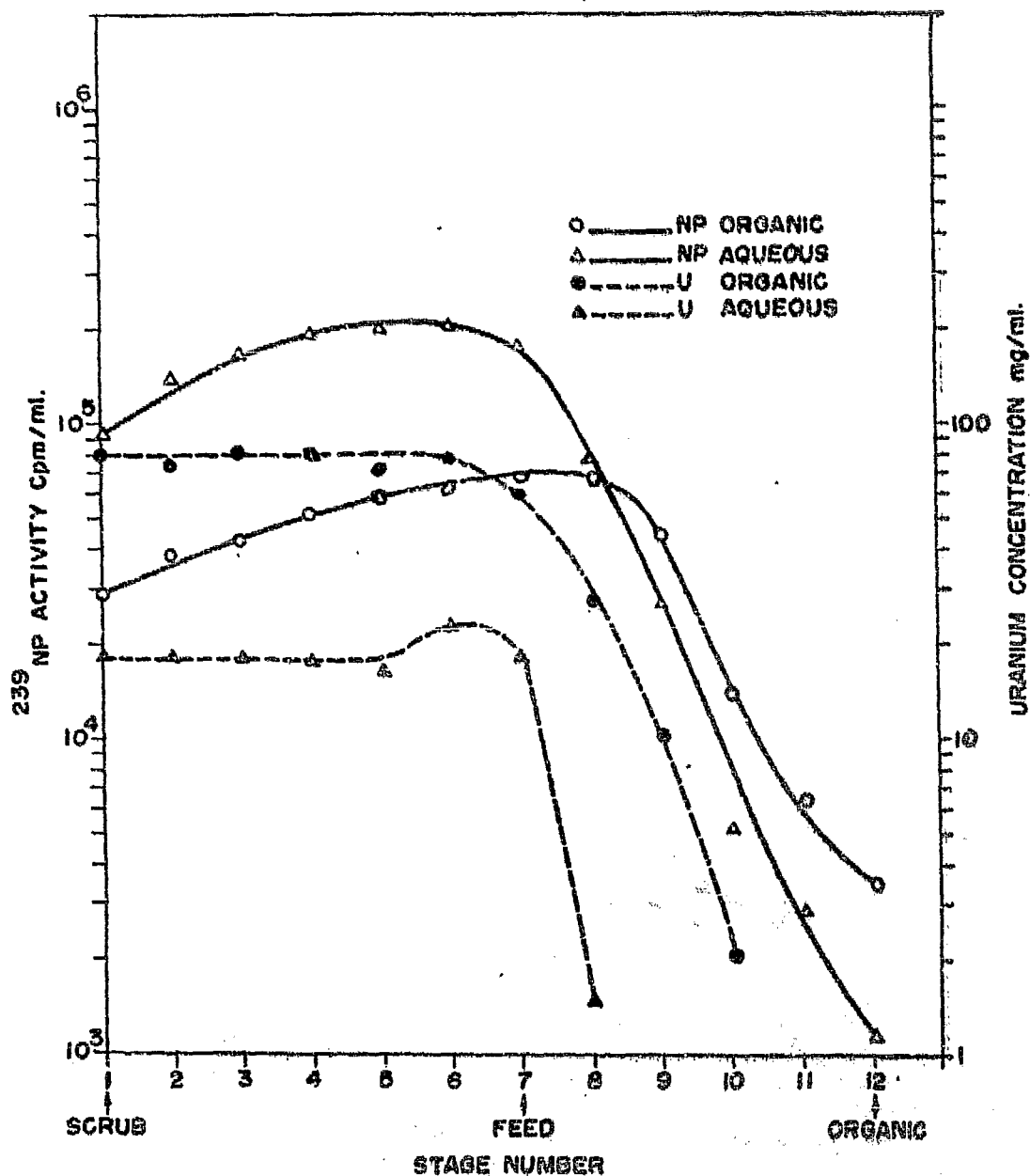


FIG. 8.
 PROFILE OF NEPTUNIUM(IV), URANIUM(VI)
 AND HNO_3 IN THE EXTRACTION
 (FOR EXPERIMENTAL CONDITIONS REF. TABLE 5.)

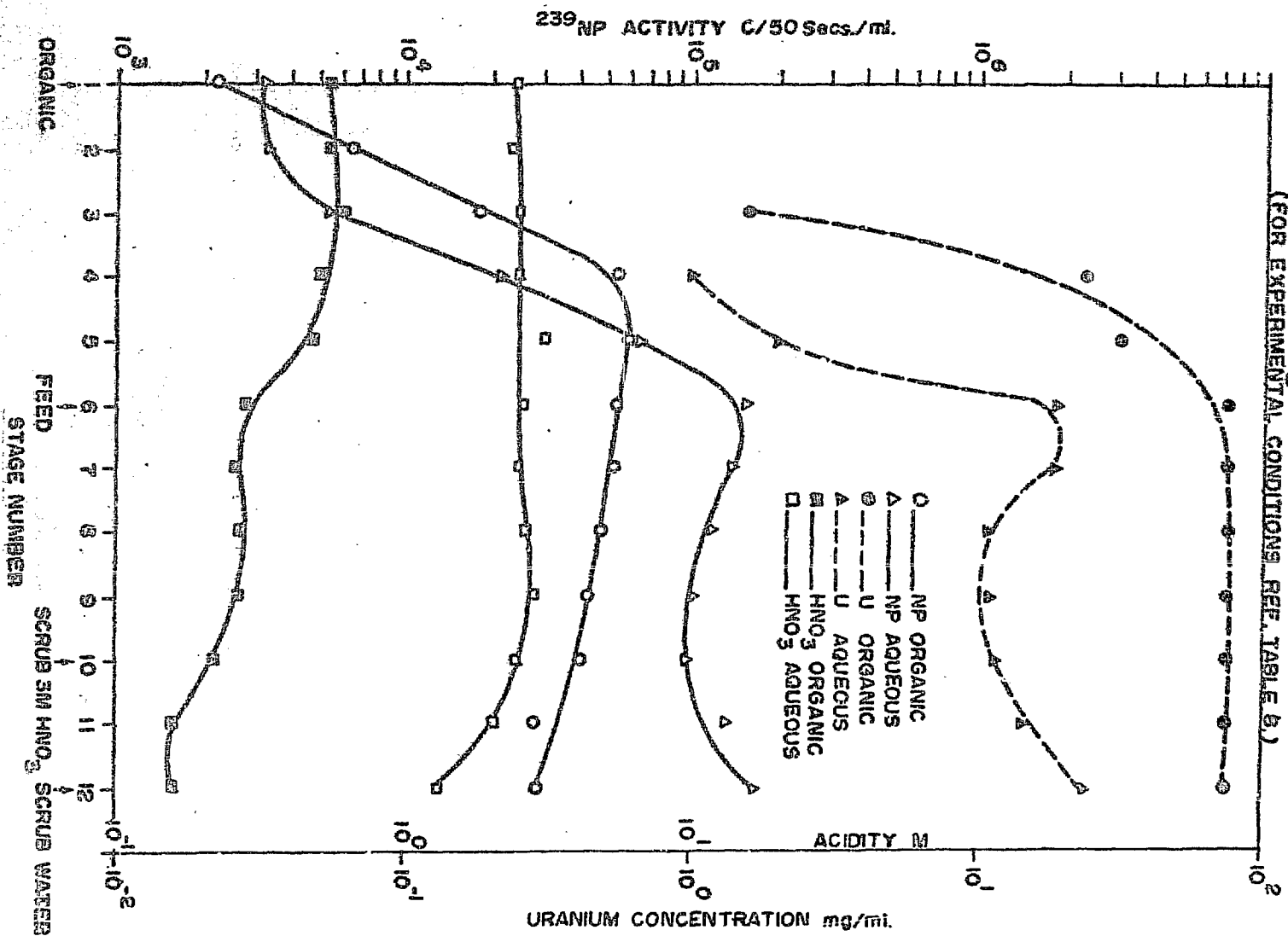


FIG.9.
PROFILE OF URANIUM (VI) AND NEPTUNIUM (IV)
IN THE SCRUBING

(FOR EXPERIMENTAL CONDITIONS REF. TABLE 9.)

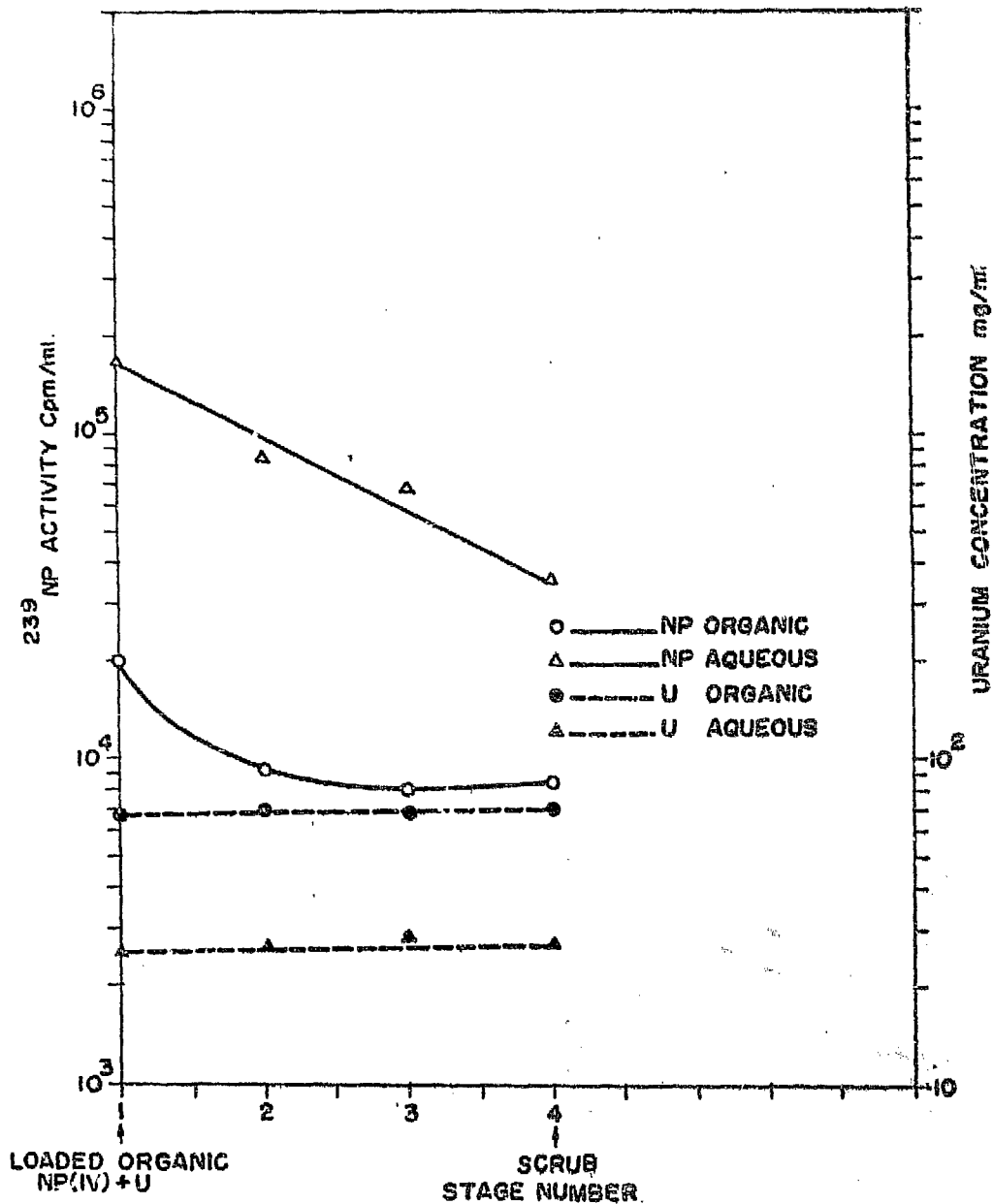


FIG.10.
 PROFILE OF NEPTUNIUM (IV), URANIUM (VI)
 AND HNO_3 IN THE EXTRACTION
 (FOR EXPERIMENTAL CONDITIONS REF TABLE 10.)

