# Extraction Processes on Reprocessing of Fast Reactor Fuel in a Purex Plant Constructed for Light Water Reactor Fuel

May. 1972

動力炉・核燃料開発事業団

東 海 事 業 所

TOKAI WORKS

POWER REACTOR AND NUCLEAR FUEL

DEVELOPMENT CORPORATION

Extraction Processes on Reprocessing of Fast Reactor Fuel
in a Purex Plant Constructed for Light Water Reactor Fuel

Kozo, GONDA*

Chuzaburo TANAKA**

Shozo YASU*

Nobuyoshi KAWASHIMA*

Tadaya HOSHINO*

Abstract

The extraction processes on reprocessing Japanese Experimental Fast Reactor (JEFR, named as JOYO) fuel have been studied with an unirradiated uranium and plutonium, supposing that the JEFR fuel will be treated with no modification of the facilities and the operation conditions except the solvent employed and the dilution of the fuel in the PNC Purex plant under construction for the exclusive use of light water reactor fuels.

The applicability of the extraction facilities in PNC plant to the extraction of JEFR fuel was verified, concerning the recoveries of uranium and plutonium in the co-decontamination cycle, even in the case that the solvent was changed to 10% TBP from 30% TBP employed in a light water reactor fuel reprocessing. Also, the validity of 30% TBP system on the partition cycle of uranium and plutonium was confirmed with regard to stripping plutonium.

* Tokai Works, Power Reactor and Nuclear Fuel Development Corporation Tokai, Ibaraki, Post No.319-11, Japan.

** Central Research Laboratory, Sumitomo Metal Mining Co. Ltd., 282-1, Nakakokubu, Ichikawa, Chiba, Post No.272, Japan.

## Introduction

It seems that, for the present, in European countries and the United States, irradiated fast breeder reactor fuels are scheduled to be reprocessed at the existing facilities[1]. The reprocessing of the irradiated fuel of Japanese Experimental Fast Reactor (JEFR) will be necessary from 1975. The amount of the core fuel to be discharged from this reactor will be ~1 ton/year and the blanket will be ~2 ton/year. The meat of core fuel is composed of 82% of 23% enriched $UO_2$ and 18% of $PuO_2$. The axial and radial blankets are made of natural or depleted $UO_2$. The meat and the blanket will be burnt up to 48,000 MW days/ton (MWD/T) with the fission ratio, Pu:U ; 1:0.91 and 860 MWD/T, respectively. The capacity of the Purex reprocessing plant of Power Reactor and Nuclear Fuel Development Corporation (PNC plant) is ~0.7 ton/day. The fuels to be treated in this plant are Magnox-clad natural uranium fuel burnt up to 3,900 MWD/T and Zircalloy-or stainless steel-clad enriched (up to 4%) uranium oxide fuel burnt up to 28,000 MWD/T, cooled for 155 and 180 days, respectively. From the capacity of PNC plant, the extraction processes of this plant are enough to treat the irradiated fuel of JEFR, since the throughput of the core fuel is calculated as 44.4 kg/day (4 elements/day), owing to the throughput of the Pu cycle and nuclear safety of the dissolver vessel and adjusting tanks. The problem left in abeyance for the extraction processes on reprocessing the irradiated JEFR fuel is an exact estimation of the fission products (FPs) and the decontamination factor (DF) through all the extraction processes. The FPs composition of the irradiated fuel, after 180 days' cooling is calculated according to the method of Burris and Dilton[2] and compared with that of the light water reactor fuel in Table 1. The amount of treatment, 44.4 kg, and the FPs dilution of irradiated JEFR fuel will be ~1/16 and ~1/6 of those of light water reactor fuels, respectively. Therefore, concerning the content of $^{106}Ru$ and $^{95}Zr$ difficult to release in the extraction processes, there will be no fateful problem to the extraction processes in reprocessing JEFR fuel.

It is a keynote not to modify the facilities and the operation conditions except the solvent to be employed and the dilution of the irradiated JEFR fuel in the extraction processes. Therefore, the followings are summarized as the limitations on the extraction processes.

Table 1 Fission products composition of irradiated fuel

(Total activity, $\beta$ Ci/kg: Cooling time 18 days)

| | LWR | JEFR (core) | JEFR (ax. blanket) |
|---|---|---|---|
| Cooling-time (d) | 180 | 180 | 180 |
| Total activity ( Ci/kg) | $4.6 \times 10^3$ | $1.4 \times 10^4$ | $2.3 \times 10^2$ |
| Kr-85 | $1.1 \times 10$ | $1.3 \times 10$ | $3.7 \times 10^{-1}$ |
| I-129 | | $5.3 \times 10^{-6}$ | $6.6 \times 10^{-8}$ |
| I-131 | $1.5 \times 10^{-4}$ | $1.3 \times 10^{-2}$ | $8.9 \times 10^{-5}$ |
| Ru-103 | $5.5 \times 10$ | $2.3 \times 10^2$ | 3.5 |
| Rh-103m | $5.5 \times 10$ | | |
| Ru-106 | $3.9 \times 10^2$ | $1.3 \times 10^3$ | 3.3 |
| Ce-141 | $3.7 \times 10$ | $1.1 \times 10^2$ | 1.9 |
| Ce-144 | $1.0 \times 10^3$ | $1.7 \times 10^3$ | $3.5 \times 10$ |
| Zr-95 | $2.5 \times 10^2$ | $9.5 \times 10^2$ | $2.0 \times 10$ |
| Nb-95 | $5.3 \times 10^2$ | $1.8 \times 10^3$ | $3.7 \times 10$ |
| Sr-89 | $1.4 \times 10^2$ | $3.1 \times 10^2$ | 7.8 |
| Sr-90 | $9.6 \times 10$ | $9.0 \times 10$ | 2.1 |
| Cs-137 | $9.0 \times 10$ | $1.5 \times 10^2$ | 2.9 |
| U-237 (g/kg) | $3.0 \times 10^{-11}$ | | |
| Np-237 (Ci/kg) | $8.2 \times 10^{-5}$ | | |
| (g/kg) | 0.12 | | |
| Am-241 (Ci/kg) | 0.15 | | |
| (g/kg) | 0.045 | | |
| Cm-242 (Ci/kg) | $6.5 \times 10^{-3}$ | | |
| (g/kg) | $2.0 \times 10^{-6}$ | | |
| Heat generation (watt/kg) | 14 | 61 | 0.92 |

(1) The throughput of fissile enriched uranium and plutonium does not exceed that in the extraction processes of light water reactor fuels from the criticality consideration of the dissolution vessel and extraction processes, namely, the same Pu cycle is considered for the JEFR fuel and light water reactor fuels.

(2) The species and concentrations of the various feed solutions and solvents except the solvent and the uranium concentration of the FPs co-decontamination cycle are same to those of the extraction processes for light water reactor fuels.

(3) No alteration is made on the numbers of the stages and the equipping points of the inputs/outputs for the various feeds/products in the extraction processes.

(4) The various feed solvents and solutions are supplied within the controllable capacities (±25%) of the pumps of PNC plant.

For the treatment of JEFR fuel in PNC plant, the following three cases are considered.

(1) To treat the core and the axial blanket separately in order to recover enriched uranium which remains in the core fuel.

(2) To treat the core and axial blanket simultaneously.

(3) To treat the core and axial blanket with the light water reactor fuels which will be treated routinely.

The first and second cases are independent campaigns for JEFR fuel. The more detailed prospect of treating the fuel in PNC plant, the specification of JEFR fuel and the planned operation conditions of JEFR have been reported in the previous papers[3][4][5], so here, we only give the amounts of treating JEFR fuel and the fuel composition in Table 2, with the chemical flow-sheets of the co-decontamination cycle and the partition cycle of light water reactor fuel shown in Figs. 1 and 2.

In the present paper, reprocessing technique and chemical flow-sheets of the co-decontamination cycle and the partition cycle of JEFR fuel established with the proving tests using unirradiated uranium and plutonium are described.

Table 2  Composition of JEFR fuel to be treated and Throughput per day

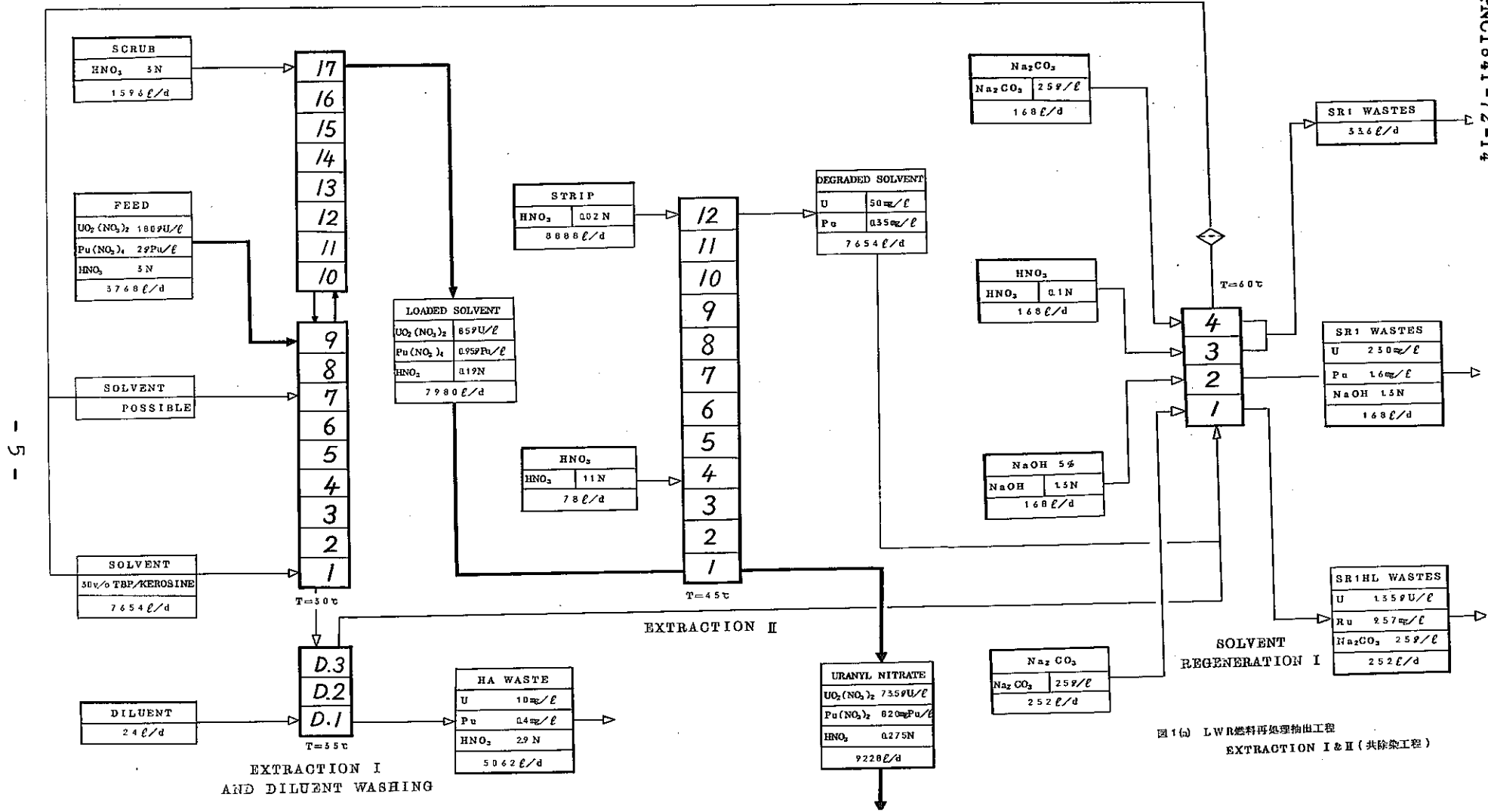| | First case | Second case |
|---|---|---|
| Form of fuel | Core | Core + axial blanket |
| | length      600 mm | length   600+2x400mm |
| | diameter    5.5 mm | diameter      5.5 mm |
| Composition of fuel | | |
| Pu-239 | 1.80 Kg/element | 1.80 Kg/element |
| 240 | 0.61 | 0.61 |
| 241 | 0.13 | 0.13 |
| 242 | 0.06 | 0.06 |
| U -235 | 1.68 | 1.78 |
| 236 | | |
| 238 | 6.82 | 21.52 |
| | 11.1  Kg/element | 25.9  Kg/element |
| Amount of treatment | 44.4  Kg | 103.60 Kg |

Note: Calculated from the composition before irradiation

- 5 -

**SCRUB**

| HNO₃ | 3 N |
|---|---|
| 1596 ℓ/d | |

**FEED**

| UO₂(NO₃)₂ | 1809U/ℓ |
|---|---|
| Pu(NO₃)₄ | 2 9Pu/ℓ |
| HNO₃ | 3 N |
| 3768 ℓ/d | |

**SOLVENT POSSIBLE**

**SOLVENT**

| 30 v/o TBP/KEROSINE |
|---|
| 7654 ℓ/d |

**DILUENT**

| 2 4 ℓ/d |
|---|

Column 17,16,15,14,13,12,11,10,9,8,7,6,5,4,3,2,1  T=30℃

D.3 D.2 D.1  T=35℃

**LOADED SOLVENT**

| UO₂(NO₃)₂ | 859U/ℓ |
|---|---|
| Pu(NO₃)₄ | 0.95 Pu/ℓ |
| HNO₃ | 0.19N |
| 7980 ℓ/d | |

**HA WASTE**

| U | 10 mg/ℓ |
|---|---|
| Pu | 0.4 mg/ℓ |
| HNO₃ | 2.9 N |
| 5062 ℓ/d | |

EXTRACTION I AND DILUENT WASHING

**STRIP**

| HNO₃ | 0.02 N |
|---|---|
| 8888 ℓ/d | |

**HNO₃**

| HNO₃ | 11 N |
|---|---|
| 7 8 ℓ/d | |

Column 12,11,10,9,8,7,6,5,4,3,2,1  T=45℃

EXTRACTION II

**URANYL NITRATE**

| UO₂(NO₃)₂ | 735 U/ℓ |
|---|---|
| Pu(NO₃)₂ | 820mg Pu/ℓ |
| HNO₃ | 0.275N |
| 9228 ℓ/d | |

**DEGRADED SOLVENT**

| U | 50 mg/ℓ |
|---|---|
| Pu | 0.35mg/ℓ |
| 7654 ℓ/d | |

**Na₂CO₃**

| Na₂CO₃ | 25 g/ℓ |
|---|---|
| 168 ℓ/d | |

**HNO₃**

| HNO₃ | 0.1 N |
|---|---|
| 168 ℓ/d | |

**NaOH 5%**

| NaOH | 1.5N |
|---|---|
| 168 ℓ/d | |

**Na₂CO₃**

| Na₂CO₃ | 25 g/ℓ |
|---|---|
| 252 ℓ/d | |

Column 4,3,2,1  T=60℃

**SR1 WASTES**

| 336 ℓ/d |
|---|

**SR1 WASTES**

| U | 230 g/ℓ |
|---|---|
| Pu | 1.6 mg/ℓ |
| NaOH | 1.5N |
| 168 ℓ/d | |

**SR1HL WASTES**

| U | 1.35 g U/ℓ |
|---|---|
| Ru | 2.57 mg/ℓ |
| Na₂CO₃ | 25 g/ℓ |
| 252 ℓ/d | |

SOLVENT REGENERATION I

図1(a) LWR燃料再処理抽出工程
EXTRACTION I & II (共除染工程)

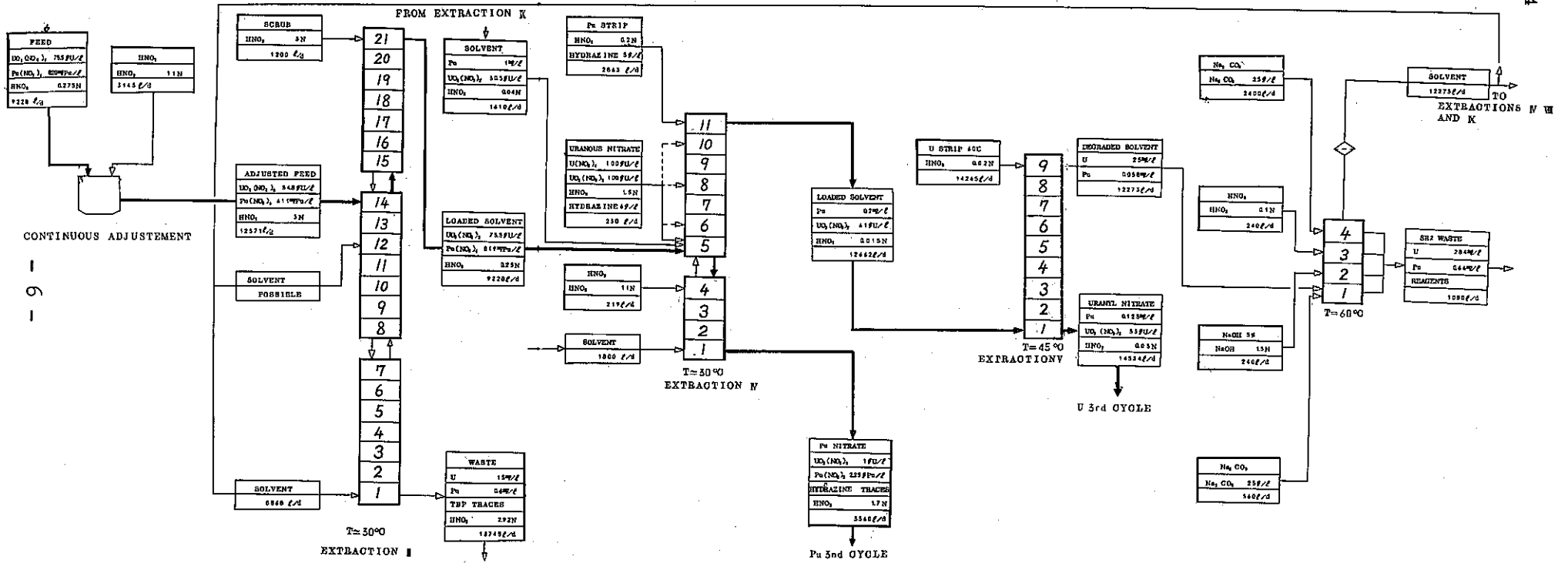Fig. 1  Co-decontamination cycle flow-sheet of Light Water Reactor Fuel in PNC plant

Fig. 2 Partition cycle flow-sheet of Light Water Reactor Fuel in PNC plant

Proving test

Principle of chemical flow-sheet

In Table 3, the number of stages employed and the operation conditions (flow-rate, flow-ratio of aqueous and organic phase, available volume of mixer and settler, and contact time of aqueous-organic phase in mixer) performed in this proving test are shown and compared with those of the extraction processes for the light water reactor fuel reprocessing calculated from the specified values in Figs. 1 and 2. The stages fewer than those specified in Figs. 1 and 2 results from the number of the stages of the miniature mixer-settler employed, 16. The other experimental conditions except the solvent employed in the co-decontamination cycle and uranium concentration were set scaling truly down the specified values in Figs. 1 and 2. The solvent recyclic process of the co-decontamination cycle is independent of those of the partition cycle and the plutonium cycle. On the other hand, in the latter two cycles, the solvent is recycled over each other cycle. Therefore, a modification of the solvent in order to raise the co-decontamination effect decreasing free TBP with the low TBP content solvent is possible in the co-decontamination cycle, but the modification is not acceptable in the partition cycle in so far as the plutonium cycle is employed without modification. In this proving test, 10 % TBP was used in the co-decontamination cycle, and 30 % TBP in the partition cycle.

Table 3  Operation conditions of JEFR fuel treatment
and comparison with those of reprocessing plant

| Extraction section and number of stages of plant | | | Flowrate in plant | | | Ratio of flowrate | | | Capacity of plant | | Contact time of Aq-Org phase in mixer | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Aq | Org | Total | Plant | First case | Second case | Mixer (ℓ) | Settler (ℓ) | Plant | First case | Second case (min) |
| I | 1~9 | EXTRN | 224 | 319 | 543 | 0.70 | 0.52 | 0.62 | 12 | 50 | 1.32 | 1.21 | 1.12 |
| | 10~17 | SCRUB | 67 | 319 | 386 | 0.21 | 0.12 | 0.12 | 12 | 70 | 1.86 | 1.63 | 1.62 |
| II | 5~12 | STRIP | 370 | 333 | 703 | 1.1 | 1.2 | 1.2 | 12 | 75 | 1.02 | 0.84 | 0.84 |
| III | 1~14 | EXTRN | 565 | 370 | 935 | 1.5 | 1.9 | 1.6 | 14.5 | 75 | 0.93 | 0.81 | 0.75 |
| | 15~21 | SCRUB | 50 | 370 | 420 | 0.14 | 0.16 | 0.15 | 12 | 50 | 1.71 | 2.06 | 1.69 |
| IV | 1~4 | EXTRN (U) | 138 | 75 | 213 | 1.8 | 2.1 | 2.1 | 3.6 | 16 | 2.88 | 2.91 | 2.94 |
| | 5~11 | STRIP (Pu) | 119 | 527 | 646 | 0.23 | 0.29 | 0.25 | 12 | 60 | 1.11 | 1.23 | 1.11 |

Equipment

Two miniature mixer-settlers and two feeding pumps for a uranium-plutonium solution and a solvent were installed in a glove box for plutonium. Other pumps were set up out the glove box. The mixer-settler (mélangeure-décanteurs 200-800 type Cyrano, made by Sonal in France) has 16 stages in a bank and each stage has a hold up of 25 ml (mixer 5.6 ml; settler 16.7 ml; dead volume 2.7 ml). As revealed in Table 3, the total volume of the bank, 400 ml is about 1/3,000 of that of EXTRACTION I, and 1/2,600 of that of EXTRACTION II. The volume of the mixer, 5.7 ml is about 1/2,000 of that of EXTRACTION I, and the ratio of the mixer to EXTRACTION II and III except EXTRACTION IV is almost the same. It can be used with an arbitrary number of stages within the total of 16, stopping the reverse aqueous stream with a stopper at the aqueous feed point. The bank is made of Plexiglass and is completely transparent; each stage has a screw to regulate the height of the two phase interfaces and an impeller which is composed of a shaft and a perforated plate of stainless steel. The detail of the mixer-settler will be illustrated in the next paper[6]. The pumps installed in the glove box, CV-1 were made by Tokyo Kagaku Seiki and the pumps set up out the box, micro pump series 2 by Hughes in England.

Procedure

The co-decontamination and the partition cycle process in the proving test were carried out individually. In these extraction processes, the solvent regeneration section was omitted and the solvent from EXTRACTION II or EXTRACTION IV was regenerated by a batchwise contact method and used in the next run. Prior to the feedings of the various solvents and solutions into the bank, fresh solvent and $3N$ $HNO_3$ or water were filled up in the bank. The flow rate of the pumps was measured every 30 min to readjust when the deviation exceeded $\pm 3\%$ of the fixed flow rate. Each height of the two phase interfaces in the settler was always checked to keep the inter-faces at the middle of the settler during the proving test run. The mixing of the two phases was performed with 2,500 r.p.m. The run was carried out at the room temperature. The composition of discharged streams was analyzed for checking of steady state during ~15 hours' operation. After steady state was obtained, the feedings and mixing were stopped and the stage samples of each bank were taken and analyzed to obtain the concentra-tion profiles of uranium, plutonium, and nitric acid in the bank.

Reagents

TBP and nitric acid used were of reagent grade and n-dodecane was obtained from Maruzen Oil Co. The solvent was prepared by washing with 10% $Na_2Co_3$, $H_2O$ and 3N $HNO_3$ in turn. The solvent containing uranium employed in EXTRACTION IV was adjusted with a pre-calculated method[7]. The plutonium used to prepare the uranium-plutonium solution was purified by the ordinary anion-exchange technique with Diaion SA-200 resin (Mitsubishi Chemical Co.) to remove americium. Sodium nitrite (reagent grade) equivalent to the plutonium was added into the prepared uranium-plutonium solution to adjust the valency of plutonium and the solution was left one night. U(IV) solution was employed to reduce Pu(IV) to Pu(III) and to release Pu(III) from the solvent stream in EXTRACTION IV, and hydrazine (extrapure grade) was added in U(IV) solution as a stabilizer.

Analysis

The methods adopted for each component were as follows:

Pu and Am: $\alpha$-counting or $\alpha$-spectrometry (if the separation of Pu from Am and/or U was necessary, an anion-exchange technique was used for this purpose).

U: colorimetry with Dibenzoylmethane (after the separation of uranium from plutonium with ethyl acetate).

U(IV): colorimetry with ferric complex of ortho-phenanthroline.

$NH_2NH_3NO_3$: titration with formaldehyde-NaOH.

Free $HNO_3$: titration with NaOH.

Obtained chemical flow-sheet

Figures 3 ~ 6 show the obtained chemical flow-sheets of the co-decontamination and the partition cycle of the first and the second cases, respectively, for the treatment of JEFR fuel. The loss of plutonium in the EXTRACTION I and III of this proving test exceeded the fixed value shown in Figs. 1 and 2 in the second case. This slight quantity of the loss is considered acceptable, since the loss in such a degree will be more or less caused in the actual extraction processes of PNC plant by a dissolution condition. The loss of plutonium in EXTRACTION II is not a problem peculiar to the extraction process for JEFR fuel, because the stripping of plutonium should be easier in this EXTRACTION II than in EXTRACTION II of PNC plant where 30% TBP is employed. The stripping loss of plutonium in EXTRACTION IV will be discussed with the loss in EXTRACTION I later. There was no problem concerning the extraction and stripping of uranium.
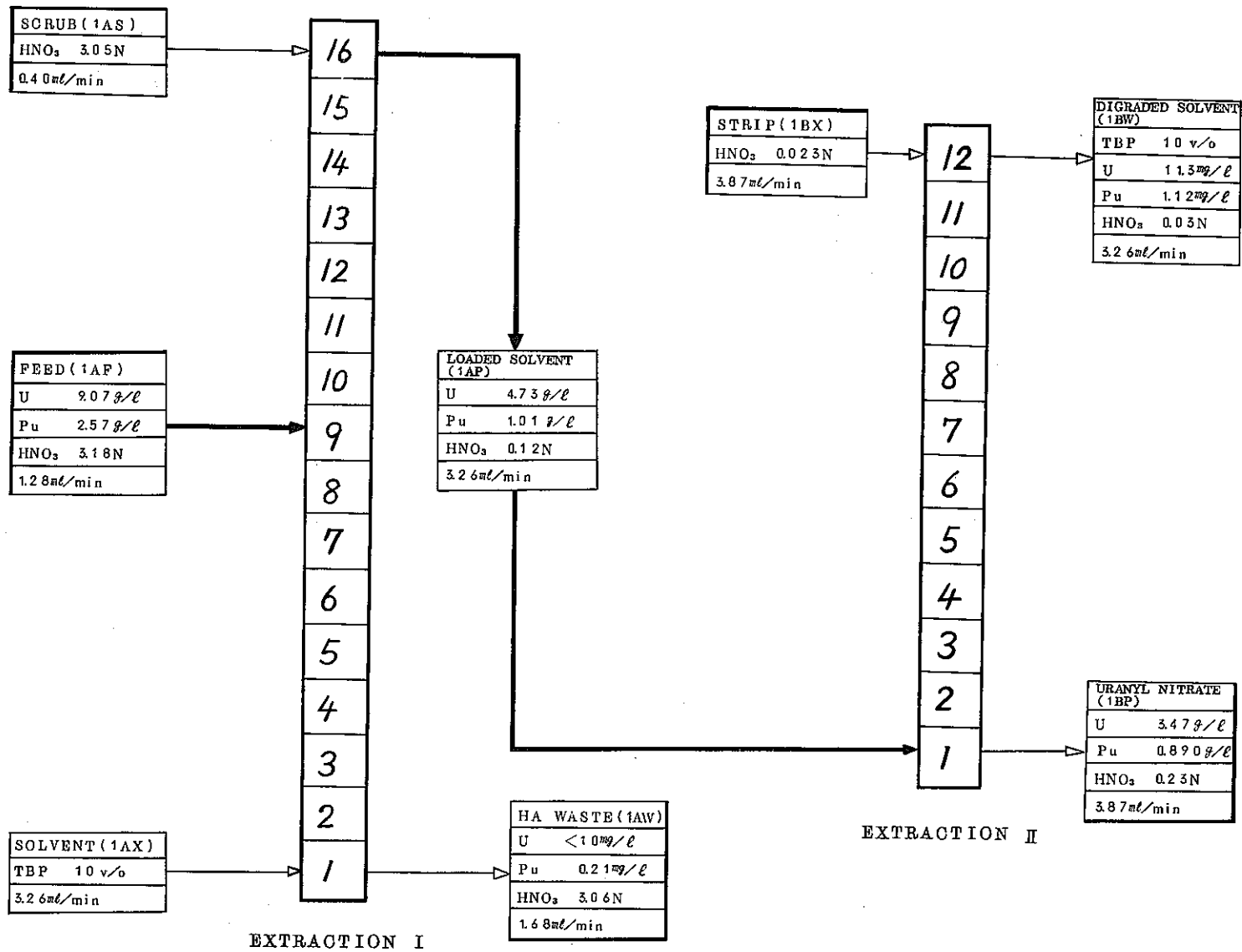
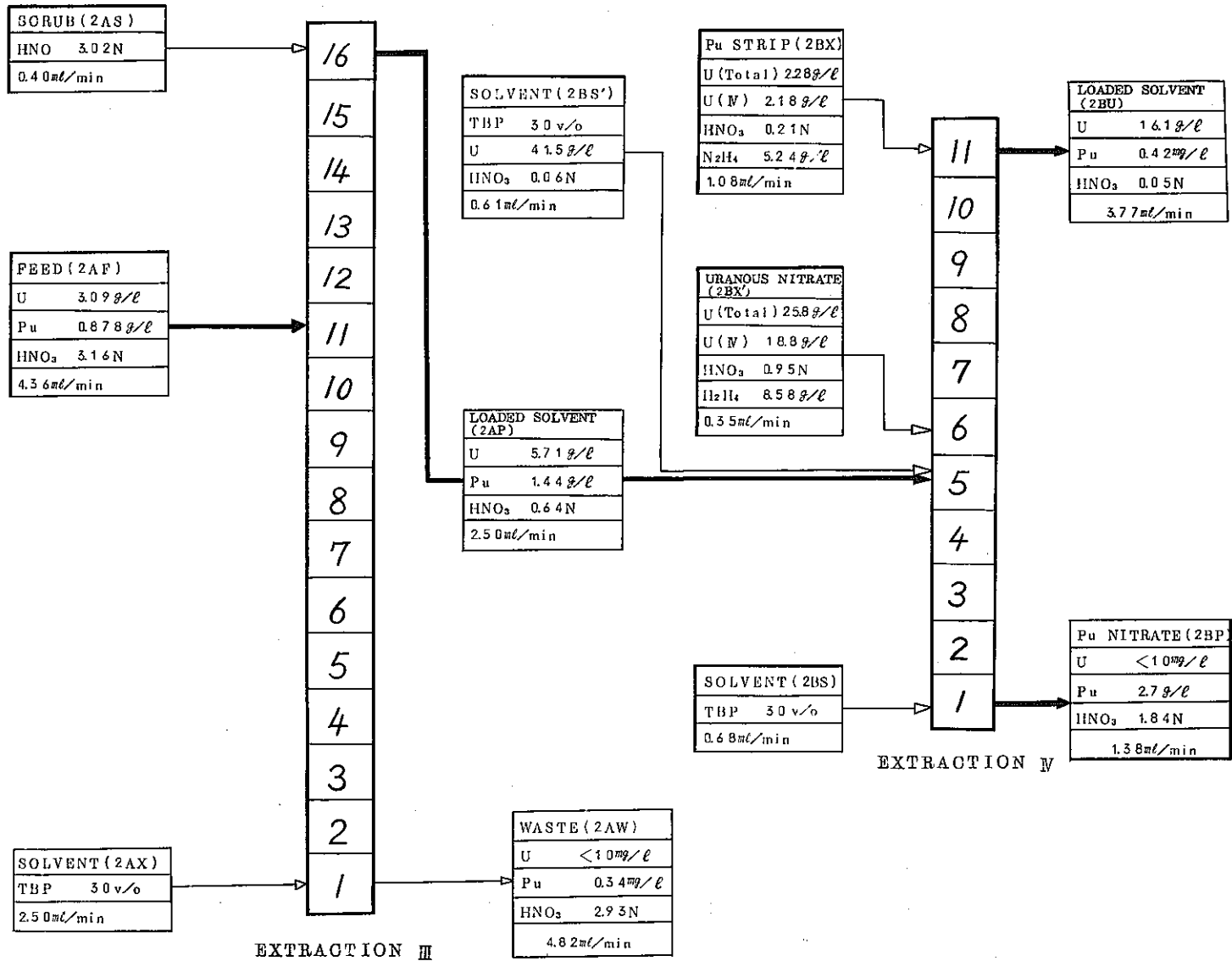Fig. 3 Co-decontamination cycle flow-sheet of JEFR fuel treatment (First case) EXTRACTION I & II

SCRUB (2AS)
| HNO | 3.02N |
| 0.40 mℓ/min | |

SOLVENT (2BS')
| TBP | 30 v/o |
| U | 41.5 g/ℓ |
| HNO₃ | 0.06N |
| 0.61 mℓ/min | |

Pu STRIP (2BX)
| U (Total) | 2.28 g/ℓ |
| U (Ⅳ) | 2.18 g/ℓ |
| HNO₃ | 0.21N |
| N₂H₄ | 5.24 g/ℓ |
| 1.08 mℓ/min | |

LOADED SOLVENT (2BU)
| U | 16.1 g/ℓ |
| Pu | 0.42 mg/ℓ |
| HNO₃ | 0.05N |
| 3.77 mℓ/min | |

FEED (2AF)
| U | 3.09 g/ℓ |
| Pu | 0.878 g/ℓ |
| HNO₃ | 3.16N |
| 4.36 mℓ/min | |

URANOUS NITRATE (2BX')
| U (Total) | 25.8 g/ℓ |
| U (Ⅳ) | 18.8 g/ℓ |
| HNO₃ | 0.95N |
| H₂H₄ | 8.58 g/ℓ |
| 0.35 mℓ/min | |

LOADED SOLVENT (2AP)
| U | 5.71 g/ℓ |
| Pu | 1.44 g/ℓ |
| HNO₃ | 0.64N |
| 2.50 mℓ/min | |

Pu NITRATE (2BP)
| U | <10 mg/ℓ |
| Pu | 2.7 g/ℓ |
| HNO₃ | 1.84N |
| 1.38 mℓ/min | |

SOLVENT (2BS)
| TBP | 30 v/o |
| 0.68 mℓ/min | |

EXTRACTION Ⅳ

SOLVENT (2AX)
| TBP | 30 v/o |
| 2.50 mℓ/min | |

WASTE (2AW)
| U | <10 mg/ℓ |
| Pu | 0.34 mg/ℓ |
| HNO₃ | 2.93N |
| 4.82 mℓ/min | |

EXTRACTION Ⅲ

Fig. 4 Partition cycle flow-sheet of JEFR fuel treatment
(First case) EXTRACTION III & IV

- 11 -

SCRUB (1AS)

| HNO₃ | 3.05N |
|---|---|
| 0.40 ml/min | |

STRIP (1BX)

| HNO₃ | 0.023N |
|---|---|
| 3.76 ml/min | |

DEGRADED SOLVENT (1BW)

| U | 11.5 mg/ℓ |
|---|---|
| Pu | 0.72 mg/ℓ |
| HNO₃ | <0.02N |
| 3.30 ml/min | |

FEED (1AF)

| U | 25.3 g/ℓ |
|---|---|
| Pu | 2.46 g/ℓ |
| HNO₃ | 3.27N |
| 1.64 ml/min | |

LOADED SOLVENT (1AP)

| U | 11.3 g/ℓ |
|---|---|
| Pu | 1.37 g/ℓ |
| HNO₃ | 0.18N |
| 3.30 ml/min | |

URANYL NITRATE (1BP)

| U | 8.35 g/ℓ |
|---|---|
| Pu | 1.28 g/ℓ |
| HNO₃ | 0.22N |
| 3.76 ml/min | |

SOLVENT (1AX)

| TBP | 10 v/o |
|---|---|
| 3.30 ml/min | |

HA WASTE (1AW)

| U | <10 mg/ℓ |
|---|---|
| Pu | 0.83 mg/ℓ |
| HNO₃ | 3.23N |
| 2.04 ml/min | |

EXTRACTION I

EXTRACTION II

Fig. 5 Co-decontamination cycle flow-sheet of JEFR fuel treatment
(Second case) EXTRACTION I & II

SCRUB (2AS)
HNO₃  3N
0.45 mℓ/min

SOLVENT (2BS')
TBP  30 v/o
U  40.0 g/ℓ
HNO₃  0.09 N
0.58 mℓ/min

Pu STRIP (2BX)
U (Total) 3.91 g/ℓ
U (Ⅳ)  2.67 g/ℓ
HNO₃  0.22 N
H₂H₄  0.33 g/ℓ
1.08 mℓ/min

LOADED SOLVENT (2BU)
U  20.9 g/ℓ
Pu  0.28 mg/ℓ
HNO₃  3N
4.30 mℓ/min

FEED (2AF)
U  9.66 g/ℓ
Pu  0.930 g/ℓ
HNO₃  2.9 N
4.35 mℓ/min

URANOUS NITRATE (2BX')
U (Total) 27.5 g/ℓ
U (Ⅳ)  24.6 g/ℓ
HNO₃  0.51 N
N₂H₄  2.0 g/ℓ
0.34 mℓ/min

LOADED SOLVENT (2AP)
U  14.1 g/ℓ
Pu  1.38 g/ℓ
HNO₃  0.54 N
3.10 mℓ/min

SOLVENT (2BS)
TBP  30 v/o
0.62 mℓ/min

Pu NITRATE (2BP)
U  30.0 mg/ℓ
Pu  2.76 g/ℓ
HNO₃  1.75 N
1.42 mℓ/mℓ

EXTRACTION Ⅳ

SOLVENT (2AX)
TBP  30 V/O
3.10 mℓ/min

WASTE (2AW)
U  <10 mg/ℓ
Pu  0.51 mg/ℓ
HNO₃  2.9 N
4.80 mℓ/min

EXTRACTION Ⅲ

Column Ⅲ stages: 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, 1

Column Ⅳ stages: 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, 1

Fig. 6  Partition cycle flow-sheet of JEFR fuel treatment
(Second case) EXTRACTION III & IV

Applicability of PNC plant for JEFR fuel treatment

Operating diagram

The calculations of the number of the theoretical stages in each extraction bank have been carried out using a graphical method. The operating diagram of each bank for uranium and plutonium is shown in Figs. 7 ~ 20. When the fixed value of an aqueous or an organic raffinate was not obtained in the proving test, the number of the theoretical stages was calculated using the result obtained from the other proving test. The results of the number of the theoretical stages obtained are summarized with the efficiency of each stage in Table 4 and compared with the actual number of stages of PNC plant. As shown in Table 4, every number of the actual stages was provided available except the plutonium stripping stage in EXTRACTION IV. However, the stripping loss of plutonium in EXTRACTION IV is not peculiar to the extraction process for JEFR fuel and will be avoidable by improving the optimum quantity of U(IV) to be added and the feeding point which are not fixed in PNC plant.

Table 4  Number of theoretical stages required for JEFR fuel treatment and stage efficiency

| Case | EXTRN section | Number of actual stages in PNC plant | Number of stages employed in test | Number of stages established | Stage efficiency |
|---|---|---|---|---|---|
| First case | EXTRN I | 17 {EXTRN 9 / SCRUB 8} | 16 {EXTRN 9 / SCRUB 7} | U EXTRN 3 / Pu EXTRN 7 | NOT ESTIMATED / 70 |
| | EXTRN II | 12 STRIP 12 | 12 STRIP 12 | U STRIP 3 / Pu STRIP 5~6 | 100 / NOT ESTIMATED |
| | EXTRN III | 21 {EXTRN 14 / SCRUB 7} | 16 {EXTRN 11 / SCRUB 5} | U EXTRN 2 / Pu EXTRN 5 | NOT ESTIMATED / 75 |
| | EXTRN IV | 11 {U EXTRN 4 / Pu STRIP 7} | 11 {U EXTRN 4 / Pu STRIP 7} | U EXTRN 2 / Pu STRIP ~7 | NOT ESTIMATED / 50 |
| Second case | EXTRN I | 17 {EXTRN 9 / SCRUB 8} | 16 {EXTRN 9 / SCRUB 7} | U EXTRN 4 / Pu EXTRN 8 | 75 / NOT ESTIMATED |
| | EXTRN II | 12 STRIP 12 | 12 STRIP 12 | U STRIP 4 / Pu STRIP 5~6 | 100 / 50 |
| | EXTRN III | 21 {EXTRN 14 / SCRUB 7} | 16 {EXTRN 11 / SCRUB 5} | U EXTRN 3 / Pu EXTRN 5 | 85 / NOT ESTIMATED |
| | EXTRN IV | 11 {U EXTRN 4 / Pu STRIP 7} | 11 {U EXTRN 4 / Pu STRIP 7} | U EXTRN 2 / Pu STRIP ~7 | NOT ESTIMATED / 50 |
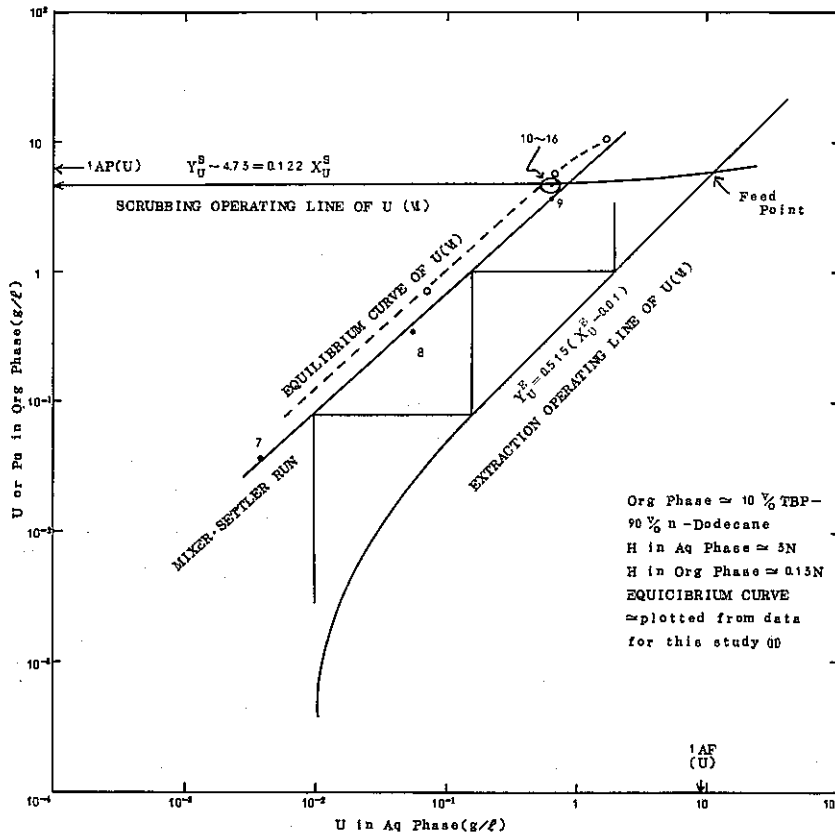
Fig. 7  Fist case, EXTRACTION I (U)
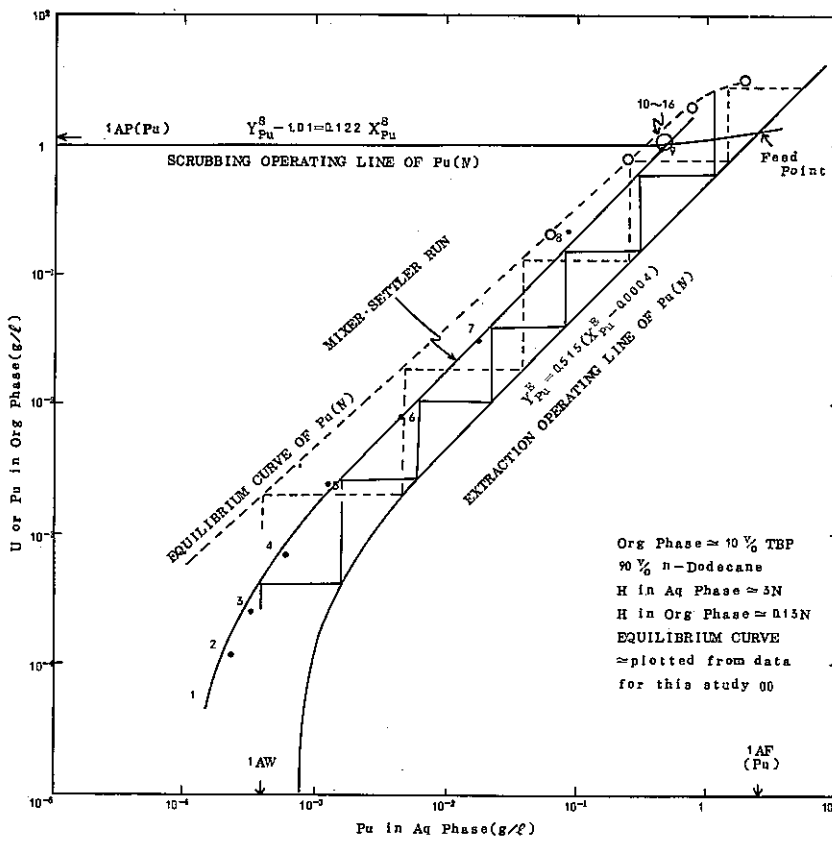


Fig. 8  First case, EXTRACTION I (Pu)
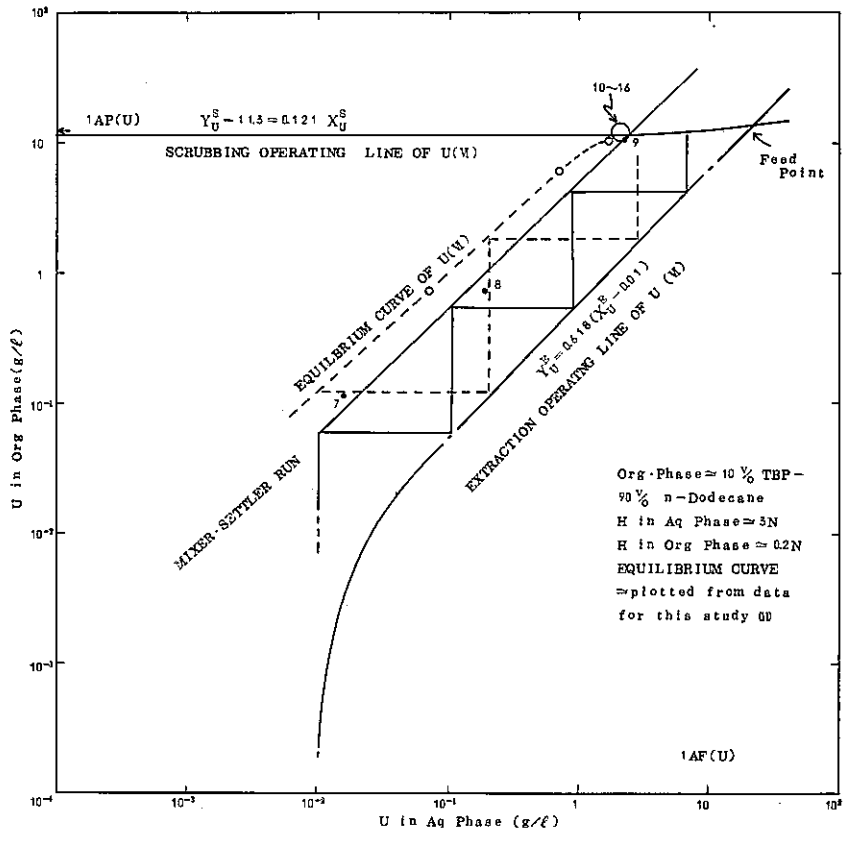
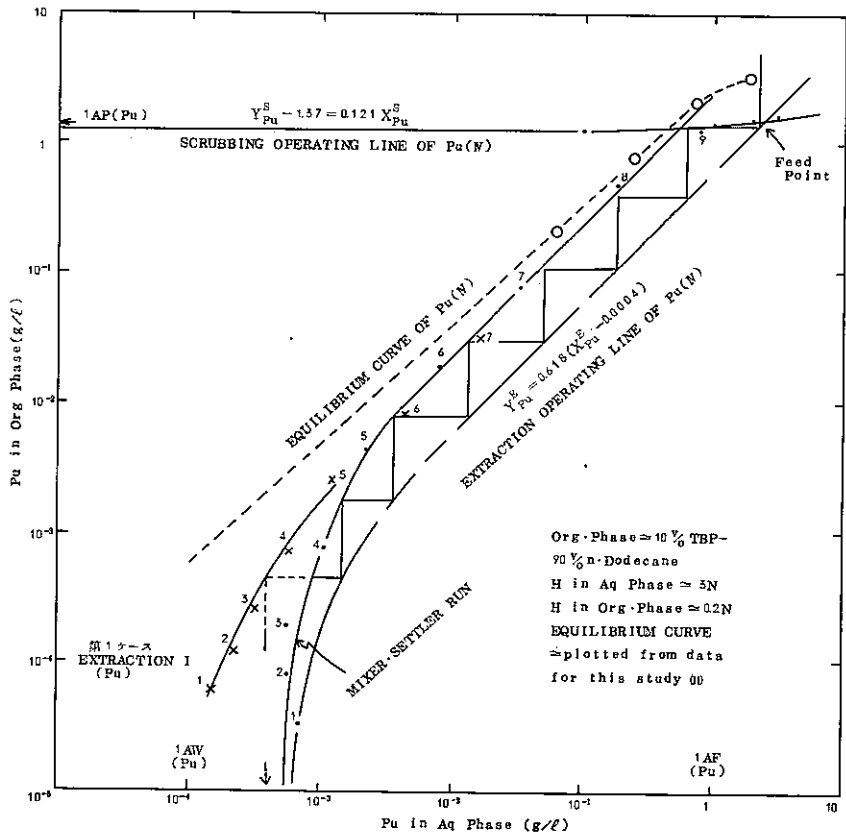Fig. 9　Second case, EXTRACTION I (U)



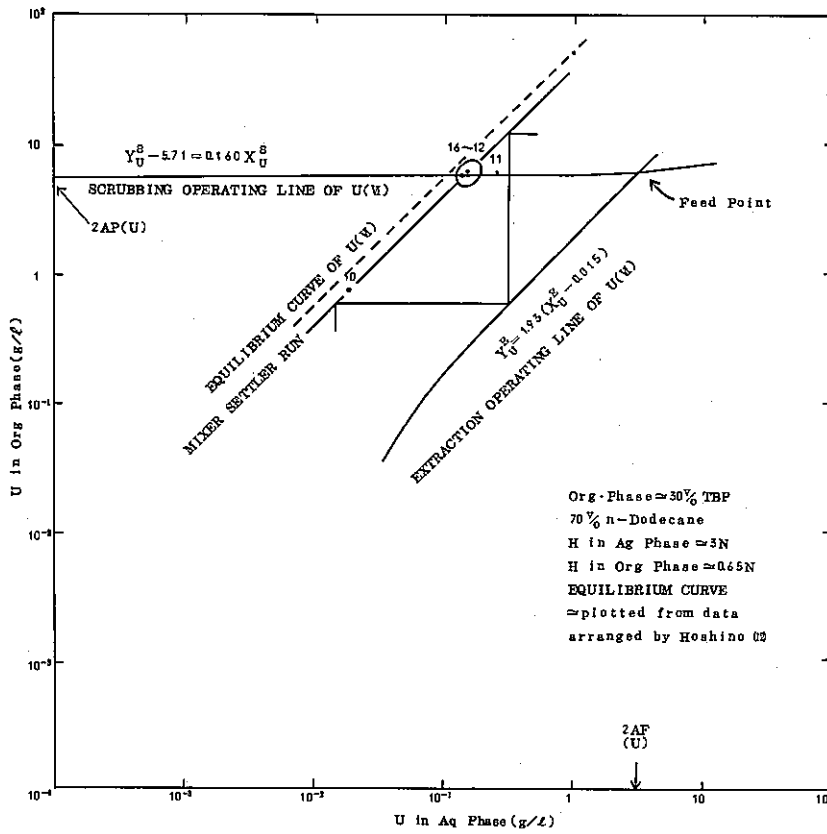Fig. 10　Second case, EXTRACTION I (Pu)
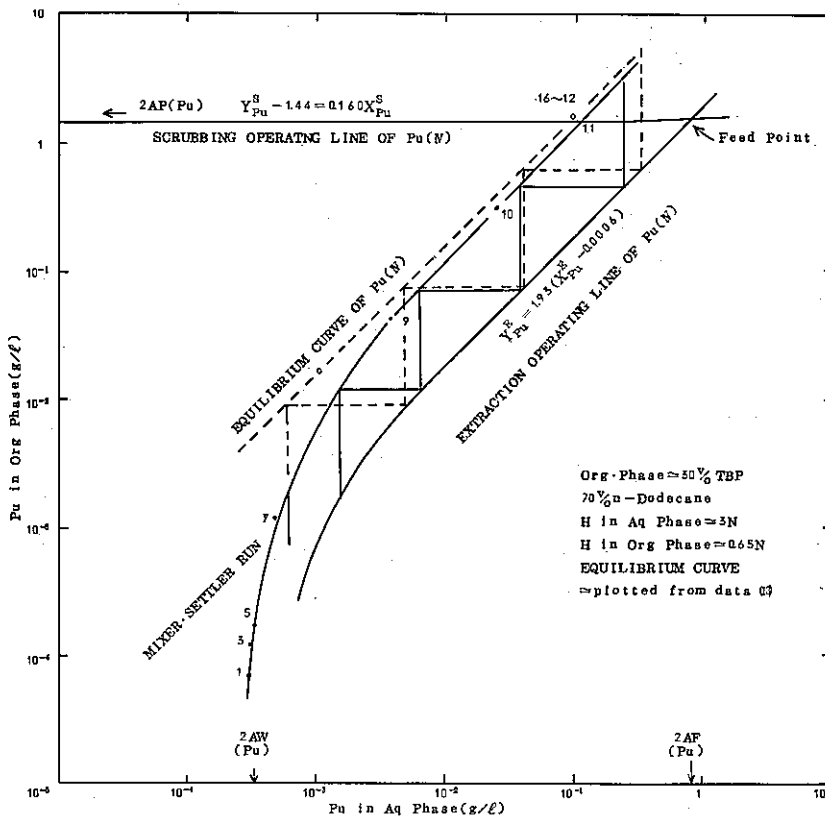
Fig. 11  First case, EXTRACTION III (U)



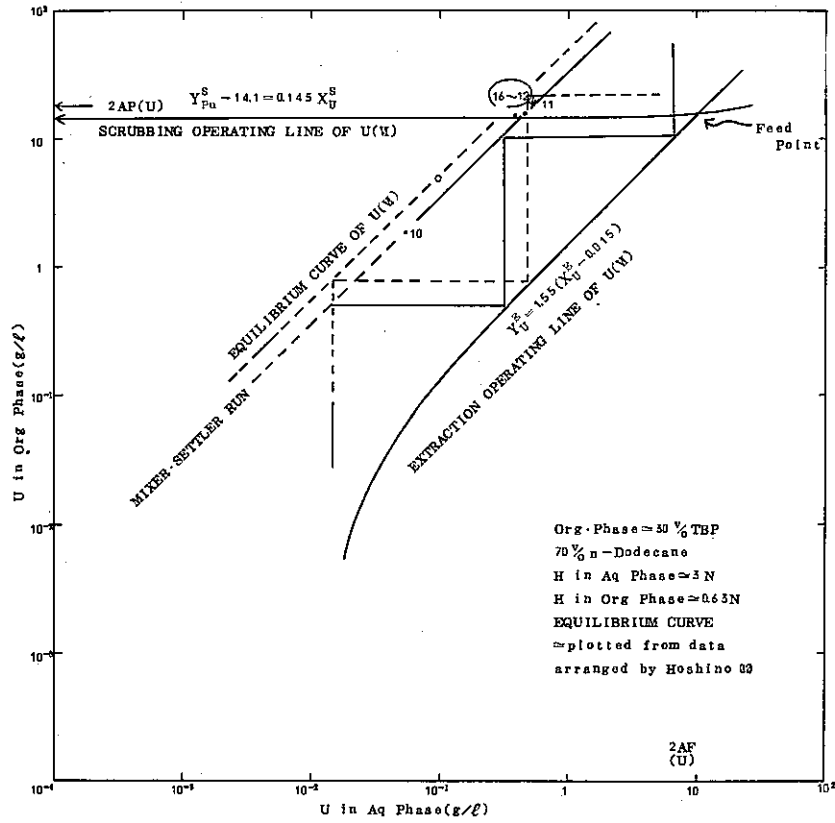Fig. 12  First case, EXTRACTION III (Pu)
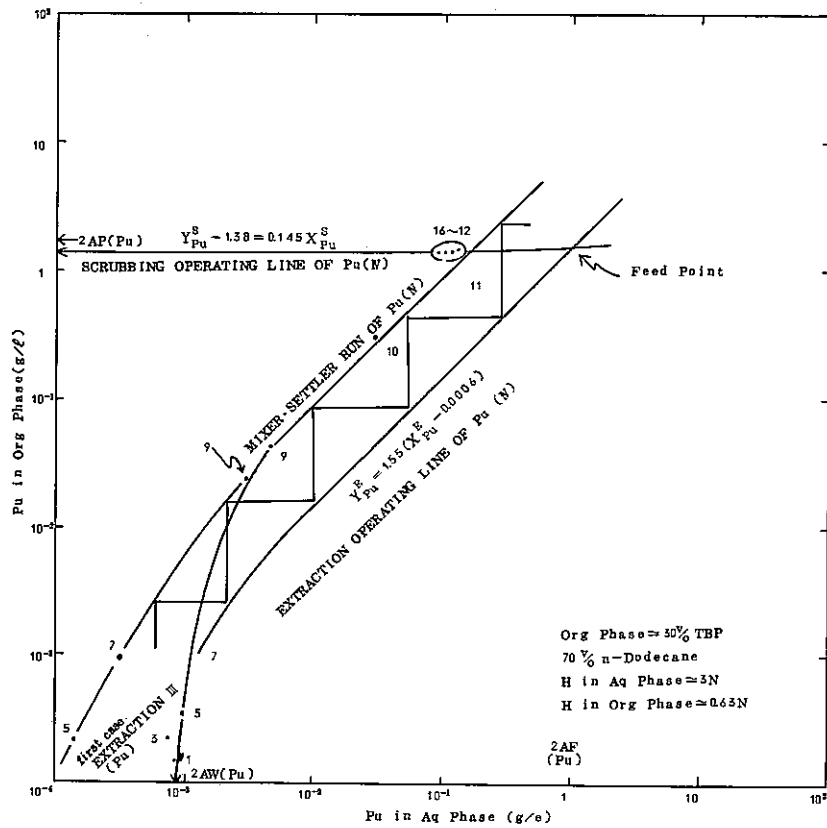
Fig. 13  Second case, EXTRACTION III (U)
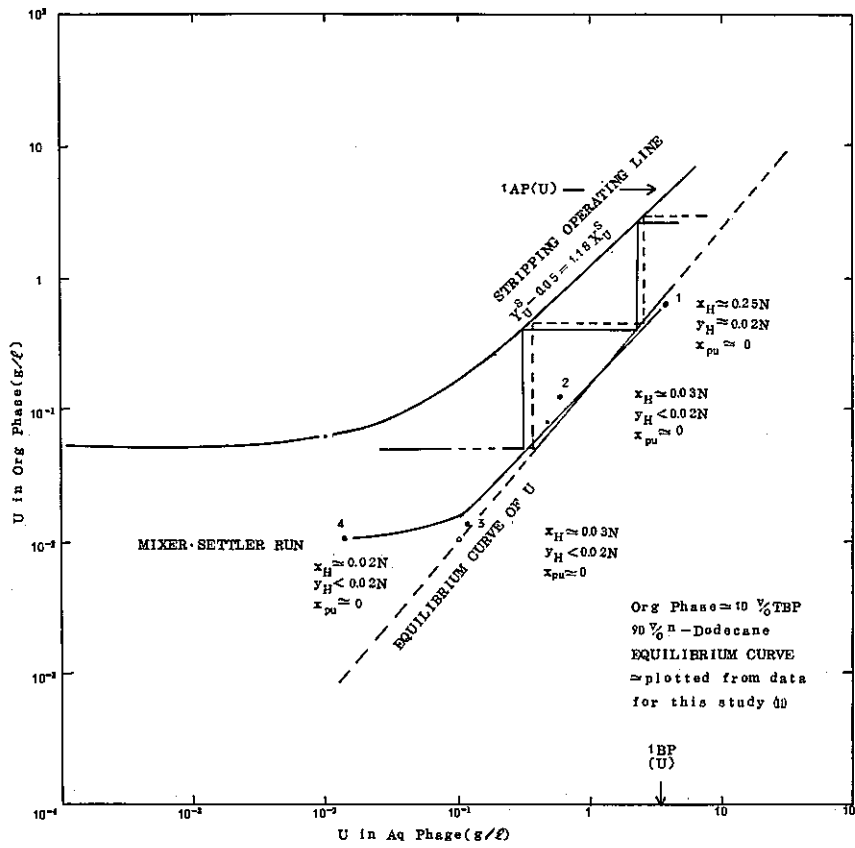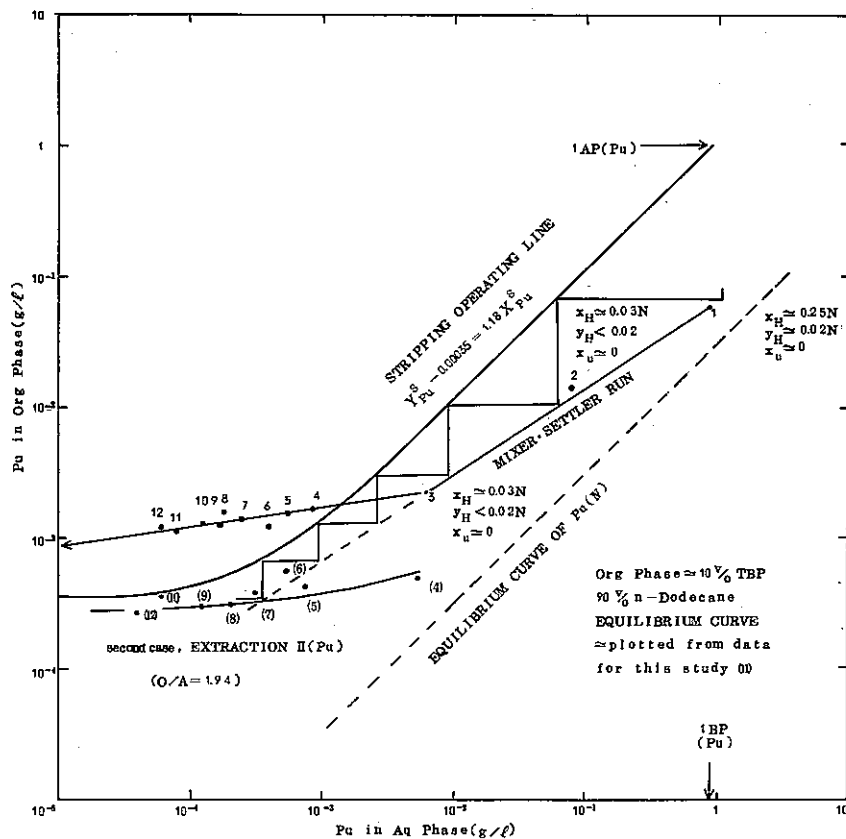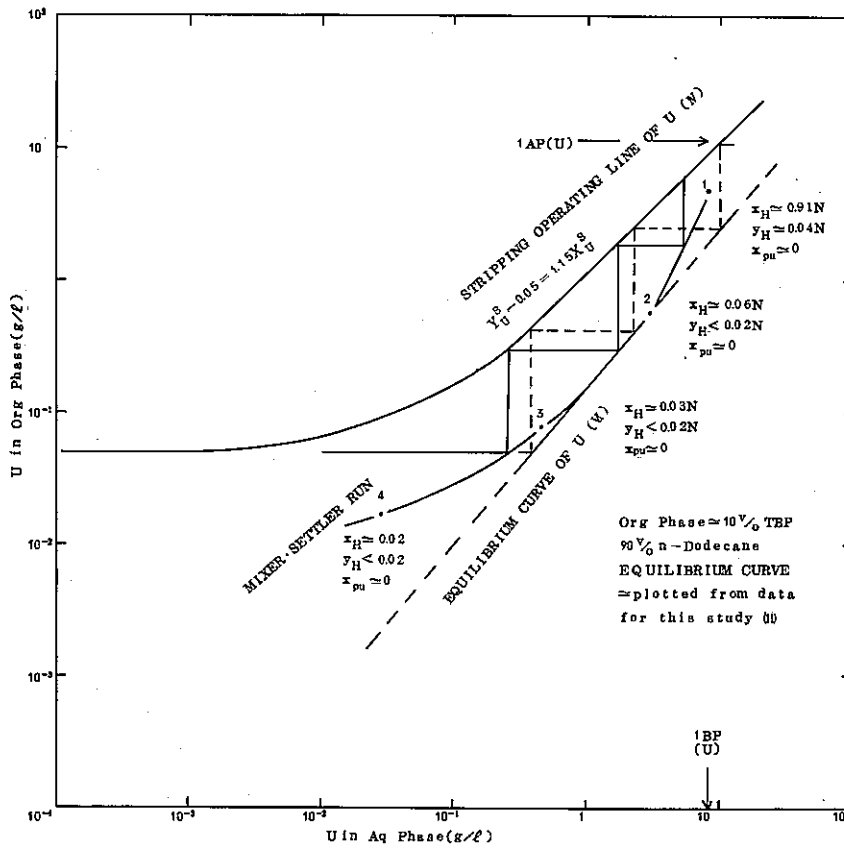


Fig. 14  Second case, EXTRACTION III (Pu)

Fig. 15  First case, EXTRACTION II (U)



Fig. 16  First case, EXTRACTION II (Pu)

Fig. 17  Second case, EXTRACTION II (U)
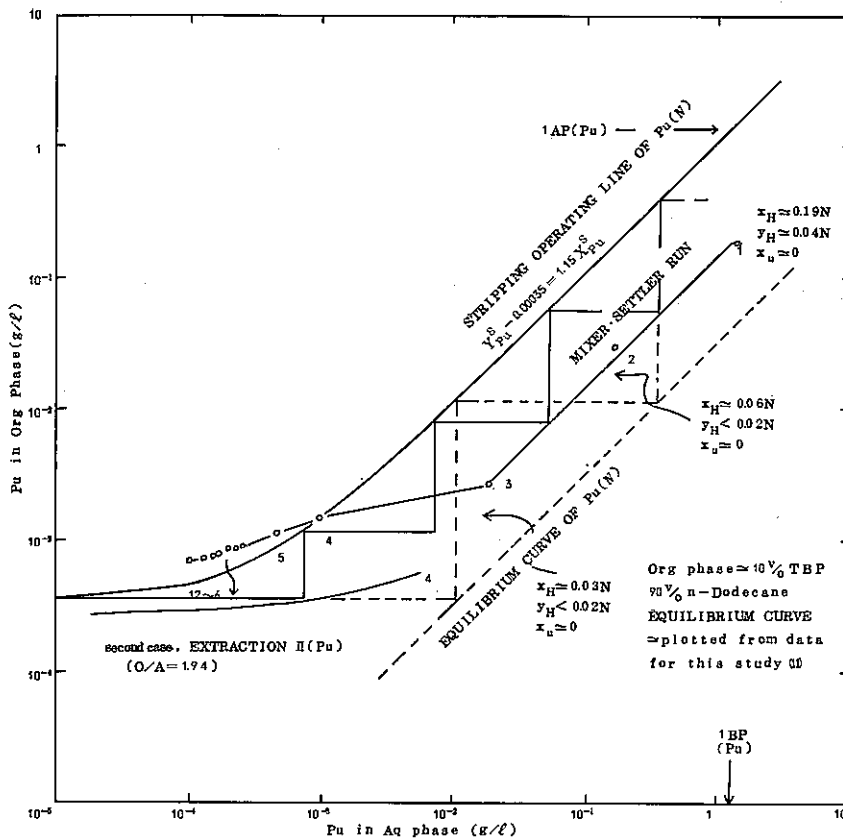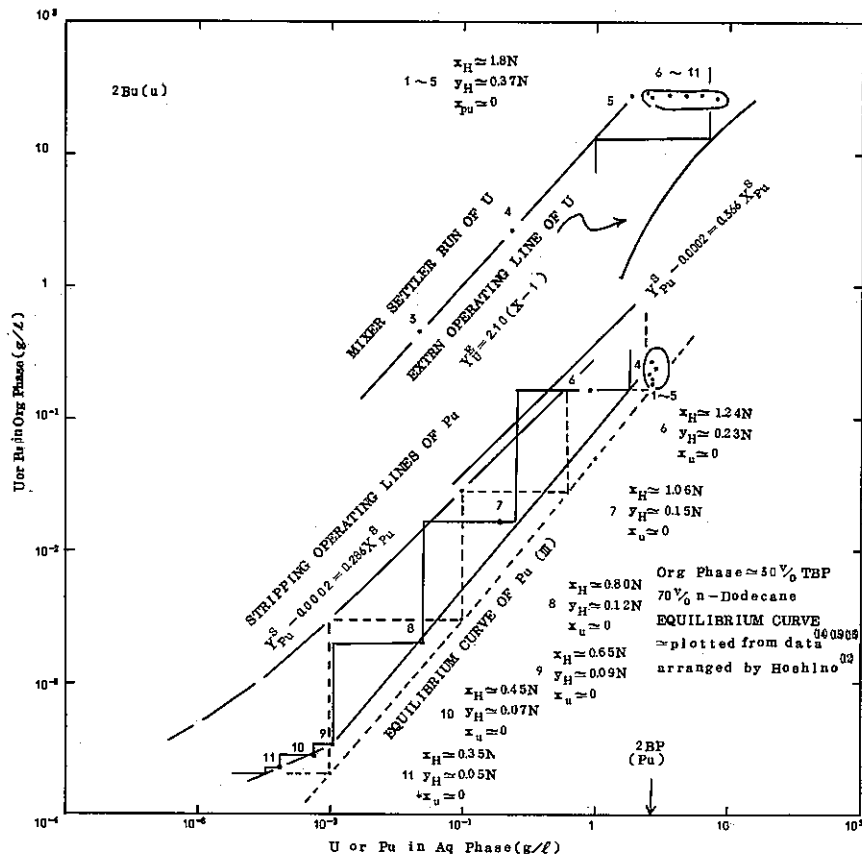


Fig. 18  Second case, EXTRACTION II (Pu)
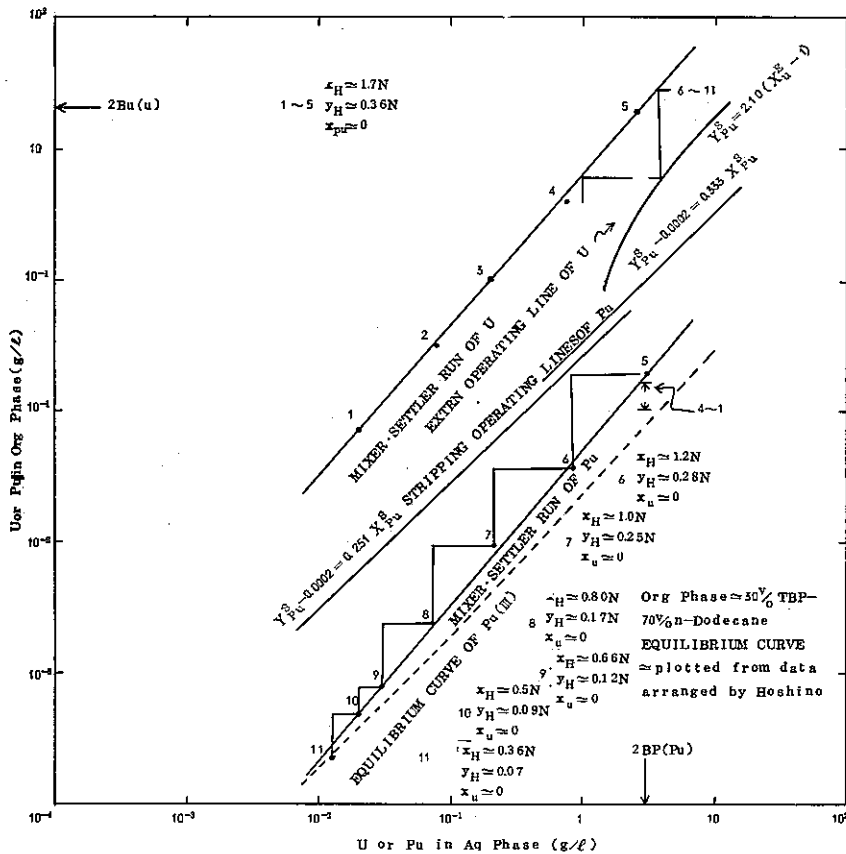
Fig. 19 First case, EXTRACTION IV (U or Pu)



Fig. 20 Second case, EXTRACTION IV (U or Pu)

Discussion

As shown in Figs. 8, 10, 12, and 14, the extractability of plutonium in EXTRACTION I and III turns lower with decreasing of plutonium concentration in the streams in the bank. On the other hand, the efficiency of stripping plutonium decreases in EXTRACTION II (Figs. 16 and 18). As these causes, the followings are considered.

(1) Existence of plutonium different from Pu(IV).

(2) Existence of hydrolyzed species of plutonium in the feed to EXTRACTION I and III.

(3) Existence of Dibutylphosphate (DBP) or Monobutylphosphate (MBP) in TBP.

The equilibrium constant for the disproportionation of Pu(IV) estimated from the data in dilute nitric acid solution[8] is $10^{-30}$-order in $3N$ $HNO_3$, so the existence of Pu(III) which is low extractive into TBP is not considered in the uranium-plutonium feed solution of $3N$ $HNO_3$. Also in 10% and 30% TBP phase, the notable disproportionation of Pu(IV) by which the unextractability can be explained was not found in a check in this proving test.

In the proving tests, the same plutonium was used over again purifying prior to each test run by the anion-exchange technique. In this anion-exchange procedure, plutonium was eluted with a dilute nitric acid ($\sim 0.2N$). The low extractability may be caused by a hydrolyzed species or a colloidal species which is not released during the recovery of plutonium from the test run or the anion-exchange procedure.

It is deduced that the leakage of plutonium at EXTRACTION II is due to DBP or MBP which is yielded by the radiation damage and the hydrolysis of ester bond of TBP and forms a complex with plutonium soluble into TBP phase. Some of DBP or MBP could be yielded in the procedure of the solvent preparation (washing with $3N$ $HNO_3$) and the long time of the stock after preparation, because even the solvent prepared by the steam distillation[9] behaves like the solvent which was not prepared by the steam distillation; and the solvent employed without delay had better efficiency of stripping plutonium. (See the data of mixer-settler run at D/A=1.94 in Figs. 16 and 18)

The stripping operation diagram of plutonium in EXTRACTION IV (Figs. 19 and 20) shows that the stripping effect falls off remarkably at the uranium product discharging side. The valency distribution of plutonium

in EXTRACTION IV has been calculated concerning the first and the second case of JEFR fuel treatment. The obtained results are shown in Table 5. In the first case, the reduction ratio of Pu(IV) to Pu(III) in the organic steam decreases rapidly at the last two stages (10 & 11) of the stripping stages, but no remarkable decrease was found in the second case. The re-oxidation of Pu(III) in the last two stages of stripping plutonium is surmised from the rapid lowering of the reduction ratio of plutonium expressly in the first case as shown in Table 5. The leakage of plutonium in the first case is slightly more than that in the second case; it seems to mean that the leakage of plutonium is caused by the re-oxidated plutonium, Pu(IV). U(IV) remaining in EXTRACTION IV was estimated as 2 equivalents of plutonium employed in EXTRACTION IV. U(IV) added was 6 equivalents of Pu(IV) and one equivalent of the 6 equivalents was consumed for reduction of Pu(IV) to Pu(III), therefore, 3 equivalent U(IV) of plutonium was spent as wasteful used in EXTRACTION IV. This amount of dissipation agreed with that which had been obtained from the plutonium free proving test[10].

Table 5  Reduction ratio of Pu(IV) in EXTRACTION IV

| Stage | Pu(III) in aqueous phase (%) | | Pu(III) in organic phase (%) | | U(IV) in organic phase (g/ℓ) | |
|---|---|---|---|---|---|---|
| | Fist case | Second case | First case | Second case | First case | Second case |
| 6 | 96 | 89 | 16 | 75 | 1.15 | 0.91 |
| 7 | 89 | 100 | 33 | 72 | 2.18 | 0.83 |
| 8 | 98 | 100 | 55 | 80 | 2.60 | 1.32 |
| 9 | 100 | 100 | 97 | 100 | 3.31 | 2.11 |
| 10 | 97 | 100 | 49 | 87 | 1.40 | 1.20 |
| 11 | 91 | 100 | 25 | 95 | 0.63 | 0.67 |

## Conclusion

To establish a technique for reprocessing irradiated JEFR fuel in the PNC plant for light water reactor fuels, the following studies of the extraction processes have been carried out:

(1)   Treatment of the core fuel;
(2)   Simultaneous treatment of the core fuel and axial blanket fuel.

As a result of these studies, it is concluded that the both cases of the reprocessing of JEFR fuel can be performed without important modification of the extraction processes in the PNC plant.   To obtain exact information on decontamination factors a series of $\gamma$-tests in $\alpha$-$\gamma$- cave using irradiated fuels will have to be carried out.

## Acknowledgment

The authors are indebted to Messrs K. Miyahara, M. Omine, Y. Ohuchi, S. Irinouchi, and K. Ueda for sample analysis.

## References

(1)  Auchapt, P., Chambon, M., et al.; A/CONF.49/P/601 4th U.N. Intern.
     Conf. Peaceful Uses of Atomic Energy, Geneva, Sept. 1971.
     Hughes, T. G., Waner, B. F., et al.; A/CONF.49/P/491 U.N. Intern.
     Conf. Peaceful Uses of Atomic Energy, Geneva, Sept. 1971.
     Sahiller, W., Baumgarther, F., et al.; A/CONF. 49/P/381 4th U.N.
     Intern. Conf. Peaceful User of Atomic Energy, Geneva, Sept. 1971.
     Nicholson, E. L.; Preliminary Investigation of Reprocessing Fast-
     Reactor Fuel in an Existing Plant. ORNL-TM-1784 (May 1967).

(2)  Burris, L., Jr., & Dilton, I. D.; Rep. Congr. atom. Energy Commn.
     U.S., 1957, ANE57.

(3)  Naito, K., Sato, S., & Hoshino, T.; Paper presented at I.A.E.A. Pannel
     on Reprocessing of Highly Irradiated Fuels, Viema, 1969.

(4)  Segawa, T., Hoshino, T., Kaya, A., Gondo, K., Tsuboya, T., & Tanaka, C.;
     Paper 139 presented at International Solvent Extraction Conference,
     1971.

(5)  Ishikawa, H.; J. Atomic Energy Soc. Japan 11, 691 (1969).

(6)  Gondo, K., Yasu, S. & Hiroki, T.; in publication

(7)  Sakakura, T.; unpublished.

(8)  Miner, F., J. et al.; B256 Chem. Rev. 3, 299-315 (1966).

(9)  Alcock, K., et al.; Trans. Faraday Soc. 52, 39 (1956).

(10) Hoshino, T.; unpublished.

(11) Tsuboya, T., et al.; unpublished.

(12) Hoshino, T., et al.; unpublished.

(13) Codding, J., W., et al.; Ind. Eng. Chem. 50, 145-52 (1958).

(14) Carleson, J.; Inorg. Nucl. Chem. 12, 136 (1959).

(15) Shevchenko ; Atomnaya Energ., 6, 426 (1958).

(16) Tsujno, T., et al.; unpublished.