

Distribution Category:
Defense Waste Management
(UC-721)

ANL-94/33

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, IL 60439

BASIC TRUEX PROCESS FOR ROCKY FLATS PLANT

by

R. A. Leonard, D. B. Chamberlain, J. A. Dow, S. E. Farley, L. Nuñez,
M. C. Regalbuto, and G. F. Vandegrift

Chemical Technology Division

August 1994

MASTER *ds*
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

| | <u>Page</u> |
|--|-------------|
| ABSTRACT..... | 1 |
| I. INTRODUCTION | 1 |
| II. SUMMARY AND CONCLUSIONS | 5 |
| III. BASE-CASE TRUEX FLOWSHEET | 6 |
| A. Design Criteria | 6 |
| 1. Process Feed | 6 |
| 2. Effluent Concentrations | 6 |
| 3. Other Considerations | 7 |
| B. Flowsheet Design..... | 8 |
| 1. Extraction and Scrub Sections | 8 |
| 2. Stripping Sections | 12 |
| 3. Solvent Cleanup Sections | 16 |
| C. Sensitivity to Process Variations | 16 |
| 1. Feed Flow Rates | 16 |
| 2. Feed Concentrations | 23 |
| 3. Number of Stages | 26 |
| IV. ALTERNATIVE TRUEX FLOWSHEET | 33 |
| A. Design Criteria | 33 |
| B. Flowsheet Design..... | 33 |
| C. Sensitivity to Process Variations | 35 |
| V. SOLVENT LOSSES AND REQUIRED VOLUME..... | 36 |
| A. Radiolysis and Hydrolysis of the TRUEX Solvent | 36 |
| B. Solubility Losses | 39 |
| C. Entrainment Losses..... | 40 |
| D. Solvent Inventory and Makeup | 40 |
| E. Removal of Solvent Components from Aqueous Effluents..... | 41 |
| VI. SAFETY INFORMATION | 43 |
| A. CMPO Safety Considerations | 43 |

TABLE OF CONTENTS (contd)

| | <u>Page</u> |
|--|-------------|
| B. Safety Considerations for Feeds to the Evaporator for Nitric Acid Recycle | 43 |
| VII. INTERACTION OF TRUEX PROCESS WITH NITRIC ACID RECYCLE | 45 |
| VIII. FEED ANALYSIS REQUIREMENTS | 46 |
| IX. OTHER PROCESS CONSIDERATIONS | 47 |
| A. Ammonium Oxalate Strip | 47 |
| 1. Benefits of an Ammonium Oxalate Strip | 47 |
| 2. Safety Concerns | 47 |
| B. Contactor Size | 48 |
| C. Filtration | 48 |
| X. FUTURE WORK | 49 |
| REFERENCES | 50 |
| APPENDIX A. GTM Reports for the Base-Case TRUEX Flowsheet | 53 |
| APPENDIX B. GTM Reports for the Alternative TRUEX Flowsheet | 60 |

LIST OF FIGURES

| <u>No.</u> | <u>Title</u> | <u>Page</u> |
|------------|---|-------------|
| I-1. | General TRUEX Flowsheet | 3 |
| III-1. | Base-Case Flowsheet for RFP Waste Developed by Using the GTM..... | 9 |
| III-2. | Effect of the Number of Extraction Stages and the O/A Flow Ratio (R) on the Am Concentration in the Aqueous (DW) Raffinate for $1 \times 10^{-10}M$ Am in the Organic (DX) Feed | 10 |
| III-3. | Effect of the Number of Extraction Stages and the O/A Flow Ratio (R) on the Am Concentration in the Aqueous (DW) Raffinate for $1 \times 10^{-11}M$ Am in the Organic (DX) Feed | 11 |
| III-4. | Effect of the Number of Scrub Stages and the O/A Flow Ratio (R) on the HNO_3 Concentration in the Organic Phase Exiting the Scrub Section | 12 |
| III-5. | Effect of the Total Number of Am Strip Stages on the Am Concentration Profile in the Aqueous Phase of the Am Strip Stages | 13 |
| III-6. | Effect of the Total Number of Am Strip Stages on the Am Concentration Profile in the Organic Phase of the Am Strip Stages..... | 14 |
| III-7. | Effect of Flow Rate for DX Feed on Am Concentration in DW Raffinate | 17 |
| III-8. | Effect of Flow Rate for DX Feed on Pu Concentration in EW Effluent | 17 |
| III-9. | Effect of Flow Rate for DX Feed on Am Concentration in FW Effluent | 18 |
| III-10. | Effect of Flow Rate for DF Feed on Am Concentration in DW Raffinate..... | 18 |
| III-11. | Effect of Flow Rate for DF Feed on Pu Concentration in EW Effluent..... | 19 |
| III-12. | Effect of Flow Rate for DF Feed on Am Concentration in FW Effluent..... | 19 |
| III-13. | Effect of Flow Rate for DS Feed on Am Concentration in DW Raffinate..... | 20 |
| III-14. | Effect of Flow Rate for DS Feed on Pu Concentration in EW Effluent..... | 20 |
| III-15. | Effect of Flow Rate for DS Feed on Am Concentration in FW Effluent..... | 21 |
| III-16. | Effect of Flow Rate for EF Feed on Am Concentration in DW Raffinate | 21 |
| III-17. | Effect of Flow Rate for EF Feed on Pu Concentration in EW Effluent | 22 |
| III-18. | Effect of Flow Rate for EF Feed on Am Concentration in FW Effluent..... | 22 |
| III-19. | Effect of Nitric Acid Concentration in DS Feed on Am Concentration in DW Raffinate | 23 |
| III-20. | Effect of Nitric Acid Concentration in DS Feed on Pu Concentration in EW Effluent | 24 |

LIST OF FIGURES (contd)

| <u>No.</u> | <u>Title</u> | <u>Page</u> |
|------------|--|-------------|
| III-21. | Effect of Nitric Acid Concentration in DS Feed on Am Concentration in FW Effluent | 24 |
| III-22. | Effect of Nitric Acid Concentration in EF Feed on Am Concentration in DW Raffinate | 25 |
| III-23. | Effect of Nitric Acid Concentration in EF Feed on Pu Concentration in EW Effluent | 25 |
| III-24. | Effect of Nitric Acid Concentration in EF Feed on Am Concentration in FW Effluent | 26 |
| III-25. | Effect of Number of Stages in Extraction Section on Am Concentration in DW Raffinate | 27 |
| III-26. | Effect of Number of Stages in Extraction Section on Pu Concentration in EW Effluent | 27 |
| III-27. | Effect of Number of Stages in Extraction Section on Am Concentration in FW Effluent | 28 |
| III-28. | Effect of Number of Stages in Scrub Section on Am Concentration in DW Raffinate | 28 |
| III-29. | Effect of Number of Stages in Scrub Section on Pu Concentration in EW Effluent | 29 |
| III-30. | Effect of Number of Stages in Scrub Section on Am Concentration in FW Effluent | 29 |
| III-31. | Effect of Number of Stages in Am Strip Section on Am Concentration in DW Raffinate | 30 |
| III-32. | Effect of Number of Stages in Am Strip Section on Pu Concentration in EW Effluent | 30 |
| III-33. | Effect of Number of Stages in Am Strip Section on Am Concentration in FW Effluent | 31 |
| III-34. | Effect of Number of Stages in Pu Scrub Section on Am Concentration in DW Raffinate | 31 |
| III-35. | Effect of Number of Stages in Pu Scrub Section on Pu Concentration in EW Effluent | 32 |
| III-36. | Effect of Number of Stages in Pu Scrub Section on Am Concentration in FW Effluent | 32 |
| IV-1. | Alternative Flowsheet for RFP Waste Developed by Using the GTM | 34 |
| V-1. | Calculated CMPO Loss due to Hydrolysis and Radiolysis as a Function of Temperature | 37 |
| V-2. | Effect of Process Temperature and Number of Cycles on the D Value for Am with No Solvent Cleanup When $[HNO_3] = 0.05M$ | 38 |
| V-3. | Effect of Process Temperature and Number of Cycles on the D Value for Am with No Solvent Cleanup When $[HNO_3] = 0.01M$ | 38 |
| A-1. | Concentration Profile Report from the GTM for the Base-Case TRUEX Process | 54 |
| A-2. | Americium Concentration Profile from the GTM for the Base-Case TRUEX Process | 57 |
| A-3. | Plutonium Concentration Profile from the GTM for the Base-Case TRUEX Process | 57 |
| A-4. | Hydrogen Concentration Profile from the GTM for the Base-Case TRUEX Process | 58 |

LIST OF FIGURES (contd)

| <u>No.</u> | <u>Title</u> | <u>Page</u> |
|------------|--|-------------|
| A-5. | Uranyl Concentration Profile from the GTM for the Base-Case TRUEX Process | 58 |
| A-6. | Oxalate Concentration Profile from the GTM for the Base-Case TRUEX Process..... | 59 |
| A-7. | Aluminum Concentration Profile from the GTM for the Base-Case TRUEX Process | 59 |
| B-1. | Concentration Profile Report from the GTM for the Alternative TRUEX Process | 61 |
| B-2. | Americium Concentration Profile from the GTM for the Alternative TRUEX Process | 64 |
| B-3. | Plutonium Concentration Profile from the GTM for the Alternative TRUEX Process | 64 |
| B-4. | Hydrogen Concentration Profile from the GTM for the Alternative TRUEX Process | 65 |
| B-5. | Uranyl Concentration Profile from the GTM for the Alternative TRUEX Process | 65 |
| B-6. | Oxalate Concentration Profile from the GTM for the Alternative TRUEX Process..... | 66 |
| B-7. | Aluminum Concentration Profile from the GTM for the Alternative TRUEX Process | 66 |

LIST OF TABLES

| <u>No.</u> | <u>Title</u> | <u>Page</u> |
|------------|--|-------------|
| III-1. | Composition of RFP Feed to the TRUEX Process | 7 |
| III-2. | Summary of Feed and Effluent Concentrations for the Base-Case Flowsheet Created by Using the GTM..... | 8 |
| III-3. | Effect of Nitric Acid and Oxalic Acid Concentrations on Pu Oxalate Solubility in the Aqueous Phase..... | 15 |
| IV-1. | Summary of Feed and Effluent Concentrations for the Alternative Flowsheet Created by Using the GTM..... | 35 |
| V-1. | Calculation of Dose Received by TRUEX Solvent after Processing RFP Nitric Acid Waste Stream for 3600 h | 37 |
| V-2. | CMPO Loss by Hydrolysis and Radiolysis Calculated by Using the GTM..... | 37 |
| V-3. | CMPO/TBP/Dodecane Loss in Base-Case Flowsheet Resulting from Solubility of Organics in the Aqueous Phase | 39 |
| V-4. | CMPO/TBP/Dodecane Loss in Base-Case Flowsheet Resulting from Entrainment of Organics in the Aqueous Phase | 40 |
| V-5. | CMPO/TBP/Dodecane Loss in Base-Case Flowsheet Resulting from Hydrolysis, Solubility, and Entrainment | 41 |
| V-6. | Solvent Inventory and Replacement Rate | 41 |
| V-7. | Concentration of CMPO, TBP, and Dodecane in Aqueous Effluents for Base-Case Flowsheet | 42 |

BASIC TRUEX PROCESS FOR ROCKY FLATS PLANT

by

R. A. Leonard, D. B. Chamberlain, J. A. Dow, S. E. Farley, L. Nuñez,
M. C. Regalbuto, and G. F. Vandegrift

Argonne National Laboratory
Chemical Technology Division
9700 S. Cass Avenue
Argonne, IL 60439

ABSTRACT

The Generic TRUEX Model was used to develop a TRUEX process flowsheet for recovering the transuranics (Pu, Am) from a nitrate waste stream at Rocky Flats Plant. The process was designed so that it is relatively insensitive to changes in process feed concentrations and flow rates. Related issues are considered, including solvent losses, feed analysis requirements, safety, and interaction with an evaporator system for nitric acid recycle.

I. INTRODUCTION

The major method for the recovery and purification of plutonium from fabrication and process residues from national security missions at Rocky Flats Plant (RFP) and at the Los Alamos National Laboratory (LANL) is dissolution in nitric acid and hydrofluoric acid, followed by an ion exchange and precipitation process. These processes generate nitric acid waste streams that contain 0.001 - 0.01 g plutonium per liter and similar amounts of americium. Over the years, decontamination of this waste (and the accompanying steam condensate, acid evaporator distillate, and caustic scrubber solutions) has resulted in the creation of millions of kilograms of transuranic (TRU) waste sludges (i.e., >100 nCi alpha emitters/gram) and tens of millions of kilograms of saltcrete (a cemented low-level waste [LLW] form). TRU wastes will require geologic disposal in the Waste Isolation Pilot Plant (Carlsbad, New Mexico). The saltcrete is currently disposed of at the Nevada Test Site. Current disposal costs for low-level waste are \$10.6/L (\$300/cu ft), while TRU waste disposal costs are about \$24/L (\$5,000/drum). Future plutonium recovery facilities will encounter a similar or worse situation.

A joint research effort is underway by LANL, RFP, the Oak Ridge Y-12 Plant, academia, and private industry to develop a process to recover nitric acid from these waste solutions. If such a process reduced saltcrete generation by 50%, the expected savings at RFP would be \$12M. One promising candidate is the TRUEX process (described below). Use of TRUEX as a pretreatment process greatly facilitates the recycling of nitric acid by providing a feed stream to the evaporators that has very low concentrations of plutonium and americium.

Besides simplifying nitric acid recycling, other benefits would result from implementing the TRUEX process. RFP would be able to (1) recycle hundreds of grams of plutonium for reuse in production facilities, (2) eliminate the need to recycle liquid effluents that now only slightly exceed discard

limits, saving hundreds of person-hours per year, (3) reduce operator exposure by keeping both americium and plutonium in properly shielded facilities instead of the unshielded waste treatment facility, (4) recover backlog residues now limited because of restrictions on the amount of americium that can be transferred to waste treatment, and (5) greatly reduce the volume of TRU waste, saving an estimated \$5-10M per year.

The TRUEX process was invented in the Chemistry (CHM) Division and is being developed in the Chemical Technology (CMT) Division of Argonne National Laboratory. TRUEX is a solvent extraction process that is very efficient at extracting and recovering both americium and plutonium from acidic waste solutions. The key ingredient in TRUEX is octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide, which is abbreviated O ϕ D[iB]CMPO but generally called simply CMPO. This extractant is combined with tributyl phosphate (TBP) and a diluent to formulate the TRUEX solvent. The diluent is typically a normal paraffinic hydrocarbon (NPH).

The potential for successfully implementing TRUEX processes at the various U.S. Department of Energy nuclear production and processing facilities is substantially enhanced by carrying out the processes in centrifugal contactors. The small size, high efficiency, and easy maintenance of centrifugal contactors allow a TRUEX process to be retrofitted in an on-line facility in existing space.

In a typical TRUEX process flowsheet, shown in Fig. I-1, the organic solvent passes from left to right and the aqueous solute from right to left in countercurrent flow. In this example, the flowsheet is divided into five sections.* Each section contains a number of stages, the specific number depending upon the TRU waste solution to be processed and the specific process goals.** A label consisting of a two-letter code is placed on each entering stream and on each effluent stream. In addition, labels shown in parentheses are placed on some streams leaving one section and entering another section.

The first section, shown on the left side of Fig. I-1, is the extraction section, in which the TRU elements are extracted from the TRU-containing waste solution. As the TRU-containing waste solution travels through the extraction section (right to left in Fig. I-1), its TRU content decreases while that of the solvent (traveling in the opposite direction, left to right) increases.

Next, the solvent encounters the scrub section. The objectives of this section are (1) to wash back impurities, which are either entrained or dissolved in the solvent coming from the extraction section, and (2) to lower the concentration of nitric acid in the solvent so that americium can be effectively stripped in the first strip section. Typically, dilute (0.04M) nitric acid is used as feed to the scrub section. Note that no effluent stream leaves the process from this section; the aqueous stream from the scrub section mixes with the aqueous (DF) feed as the two streams enter the extraction section.

The next series of stages makes up the first (or americium) strip section. In this section, americium, curium, and rare-earth elements are removed from the solvent and exit in the americium product

*A section is identified by the incoming aqueous phase that enters it at its last stage (the stage on the right side) and either exits at the first stage (the stage on the left side) or continues on to the next section. In the example presented in Fig. I-1, there are five sections, identified in terms of the five incoming aqueous feed streams.

**A stage in a countercurrent solvent-extraction process performed in a centrifugal contactor is a physical unit where a single equilibration between two phases takes place.

base-case flowsheet developed to meet these criteria. This flowsheet generates both an americium product stream and a plutonium product stream. In addition to discussing the various sections of the flowsheet, we evaluate how sensitive the base-case flowsheet is to variations in feed concentrations, flow rates, and number of stages in various sections. In Section IV, we present a second TRUEX flowsheet in which the extraction, scrub, and americium strip sections were modified in order to concentrate the americium in its product stream by a factor of 50. In Sections V through X, information is presented on solvent losses, solvent make-up, safety, the interaction of the TRUEX process with nitric acid recycling, feed analysis requirements, and other process considerations such as filtration and contactor size. Work that still needs to be completed or verified is also discussed.

II. SUMMARY AND CONCLUSIONS

1. The Generic TRUEX Model (GTM) was used to develop a viable TRUEX process flowsheet (the base-case flowsheet) for a typical Rocky Flats Plant (RFP) feed composition generated from the conditioning of waste residues for final disposal. This flowsheet will work over the expected range of variations in the feed composition. A 32-stage centrifugal contactor was suggested to implement this base-case flowsheet.
2. The base-case flowsheet has been optimized to achieve a process that is relatively insensitive to changes in process flow rates and feed compositions. As a design criterion, the base-case flowsheet will tolerate changes of 10% in these process variables over the expected range of feed compositions (see Table III-1).
3. The base-case flowsheet should give a TRUEX process that will convert nitrate TRU waste streams at RFP to nonTRU waste containing less than 0.01 nCi/mL of americium and plutonium. The plutonium will be recovered in a separate effluent from the americium, so it should be possible to recycle the plutonium to the plant.
4. Possible interactions between the TRUEX process and the nitric acid recycling process have been examined, and it appears that implementing the TRUEX process will not have any detrimental impact on nitric acid recycle.
5. Because of the robustness of the base-case flowsheet design, routine chemical analysis of the TRU waste feed to the TRUEX process may not be required.
6. Solvent losses have been estimated, and ways to minimize these losses are discussed.
7. The GTM was used to develop an alternative TRUEX flowsheet (high americium flowsheet) for a typical RFP feed composition that will also work over the expected range of feed compositions. The difference between the base-case and the alternative flowsheet is that in the alternative flowsheet, the americium concentration in the americium product stream will be greater by a factor of 50. A 40-stage centrifugal contactor will be required to implement this alternative flowsheet. Further work is needed to optimize the alternative flowsheet.

III. BASE-CASE TRUEX FLOWSHEET

The base-case TRUEX flowsheet for processing of TRU waste at RFP is presented here. First, the design criteria used to develop the flowsheet are discussed. Second, the flowsheet design is given, along with a discussion of how each section contributes to the design criteria. Finally, the base case is shown to be relatively insensitive to variations in the process feed concentrations and flow rates as well as to the number of process stages. The Generic TRUEX Model (GTM), which has been developed at Argonne over the last five years [VANDEGRIFT-1993], was the key tool used to design this base-case TRUEX flowsheet for RFP. The GTM version used here was Version 2.4.*

A. Design Criteria

The design criteria for the base-case TRUEX process include (1) the concentration range for the various components in the TRU waste stream that is the aqueous (DF) feed to the TRUEX process, (2) the desired component concentrations in the various process effluents, and (3) other process considerations relating to solvent cleanup and to process robustness when flow rates and component concentrations vary.

1. Process Feed

On the basis of discussions with people at the Los Alamos Technical Office at Rocky Flats Plant (in particular, with Anthony Muscatello and Jeff Hatchell), we developed a typical concentration for the nitrate waste feed. This feed concentration is shown in Table III-1, along with the maximum and minimum values. This range of feed compositions was used to develop the TRUEX flowsheets presented here. Several components present in typical RFP wastes (Na, Ca, and Cr) were eliminated from this list because their concentrations are low enough that their presence does not affect the process. Several components were also eliminated because they are not included in the GTM (NO_2 , Cl, NH_4 , and Si). The effect of these latter components on the TRUEX process is discussed below. The addition of caustic wastes to the nitrate waste feeds was not considered because such wastes could present a problem if they caused the formation of any plutonium polymer.

2. Effluent Concentrations

The basic design criterion for the effluent concentrations is to have less than 0.01 nCi/mL of TRU elements (Am, Pu) in the extraction section (DW) raffinate. This removal of TRU elements from an acid feed solution is the basic function of the TRUEX process. Note that 0.01 nCi/mL corresponds to $1.22 \times 10^{-11} \text{M}$ for ^{241}Am and to $6.1 \times 10^{-11} \text{M}$ for Pu that is 96% ^{239}Pu and 4% ^{240}Pu . A second design criterion is to have less than 1% Pu in the Am (EW) product stream from the Am strip so that greater than 99% of the Pu is recovered in the Pu (FW) product stream. The Pu recovered in the FW stream can then be recycled back to the plant. A third design criterion is to have less than 0.1% Am in the FW stream from the Pu strip so that greater than 99.9% of the Am is recovered in the EW stream.

The third design criterion, less than 0.1% Am in the Pu product, must be met to prevent $\text{Am}_2(\text{C}_2\text{O}_4)_3$ from precipitating on contact with the Pu strip solution, which contains oxalic acid. The solubility limit of $\text{Am}_2(\text{C}_2\text{O}_4)_3$ is reached when the concentration of Am exceeds about $1 \times 10^{-6} \text{M}$. The solubility of $\text{Am}_2(\text{C}_2\text{O}_4)_3$ is not affected much by variations in the concentration of oxalic and nitric acid in the Pu strip section.

*Version 3.1.1 was released September 30, 1994.

Table III-1. Composition of RFP Feed to the TRUEX Process

| Component | Feed Composition, <u>M</u> | | |
|---------------------------------|----------------------------|---------|---------|
| | Minimum | Typical | Maximum |
| Nitric Acid HNO ₃ | 5.0 | 7.5 | 7.5 |
| TRU Elements | | | |
| Pu ⁴⁺ | 4.2E-06 | 5.9E-06 | 1.3E-04 |
| Am ³⁺ | 2.1E-06 | 6.6E-06 | 2.1E-04 |
| UO ₂ ²⁺ | 0 | 3.4E-09 | 8.4E-06 |
| Metal Ions | | | |
| Mg ²⁺ | 0 | 0.24 | 0.24 |
| Al ³⁺ | 0 | 0.03 | 0.15 |
| Fe ³⁺ | 0 | 2.2E-03 | 0.04 |
| Anions | | | |
| NO ₃ ⁻ | 5.0 | 8.1 | 8.6 |
| F ⁻ | 0 | 0.03 | 0.03 |
| SO ₄ ²⁻ | 0 | 0.04 | 0.04 |

In an initial base-case flowsheet, both the Am and Pu would have been recovered in a single strip section. Because of the low solubility limit for Am in oxalic acid, this flowsheet had to be abandoned. By splitting the strip into two sections, one for Am and one for Pu, we avoid the formation of an Am₂(C₂O₄)₃ precipitate in the Pu strip section.

3. Other Considerations

In order to achieve very low concentrations of TRU elements in the extraction section, the recycled solvent must contain very low concentrations of the TRU elements. Our design criterion is for the organic (DX) feed to contain less than 0.01 nCi/mL of the TRU elements. To reach this goal, the stripping sections were designed so that the concentration of the organic phase leaving the Pu scrub section should be approximately 0.01 nCi/mL for the TRU elements. Some TRU elements may be held in the organic phase by degradation products of CMPO and TBP. These degradation products, typically organic acids, and the TRU elements contained in them must be removed in a solvent cleanup section such as the one identified in this report.

For the process to be robust, the TRUEX flowsheet should work over the entire range of concentrations expected in the aqueous (DF) feed. In addition, the process should achieve the desired effluent criteria even if (1) the feed rate of any process stream varies by $\pm 10\%$ and (2) the component concentrations in the various feed streams, except the DF feed, vary by $\pm 10\%$. In the DF feed the variations in component concentrations are much greater than $\pm 10\%$; that is, they were assumed to have the range given in Table III-1.

For general process operability, a second organic phase should not form, and no solid phase should be generated by precipitation. To prevent the formation of a second organic phase, the nitric acid concentration in the process is kept below 6M. The oxalic and nitric acid concentrations in the Pu strip are adjusted so the Pu does not precipitate in the contactors as Pu(C₂O₄)₂. This adjustment is discussed below in Section III.B.2.b. The criterion to prevent the formation of Am₂(C₂O₄)₃ as a precipitate is discussed below in Section III.B.2.a.

B. Flowsheet Design

By using the design criteria above with the GTM, a base-case flowsheet was developed for TRU processing of TRU wastes at RFP. This TRU processing flowsheet, summarized in Table III-2 and shown in Fig. III-1, achieves all of the design criteria by means of a 32-stage centrifugal contactor. Additional GTM reports and charts for the flowsheet are given in Appendix A. The design considerations for (1) the extraction and scrub sections, (2) the stripping sections, and (3) the solvent cleanup sections are discussed here.

Table III-2. Summary of Feed and Effluent Concentrations for the Base-Case Flowsheet Created by Using the GTM

| Section Name | Stage No. | Phase ^a | Flow Direction | Stream Identity ^b | Flow Rate, L/h ^c | Component Concentration, M | | | | | |
|--------------|-----------|--------------------|----------------|------------------------------|-----------------------------|----------------------------|------------------|------------------|-------------------------------|------------------|---|
| | | | | | | H ⁺ | Pu ⁴⁺ | Am ³⁺ | UO ₂ ²⁺ | Al ³⁺ | C ₂ O ₄ ²⁻ |
| Extraction | 1 | O | In | DX | 150 | 1E-15 | 1E-15 | 1E-15 | 1E-15 | 1E-15 | 1E-15 |
| Extraction | 10 | A | In | DF | 200 | 7.5 | 1.3E-04 | 2.1E-04 | 8.4E-06 | 0 | 0 |
| Scrub | 15 | A | In | DS | 100 | 0.25 | 0 | 0 | 0 | 0.03 | 0 |
| Am Strip | 23 | A | In | EF | 150 | 0.04 | 0 | 0 | 0 | 0 | 0 |
| Pu Strip | 24 | A | In | FF | 10 | 1.0 | 0 | 0 | 0 | 0 | 0.5 |
| Pu Scrub | 28 | A | In | FS | 140 | 9.0E-03 | 0 | 0 | 0 | 0 | 4.0E-03 |
| Extraction | 1 | A | Out | DW | 299.2 | 5.0 | <1E-16 | 1.22E-14 | <1E-16 | 9.9E-03 | 5.0E-16 |
| Am Strip | 16 | A | Out | EW | 150 | 0.184 | 7.1E-07 | 2.8E-04 | 1.26E-07 | 1.81E-04 | <1E-16 |
| Pu Strip | 24 | A | Out | FW | 150.8 | 0.0663 | 1.72E-04 | 3.6E-09 | 1.10E-05 | <1E-16 | 0.0315 |
| Pu Scrub | 28 | O | Out | FP ^d | 150 | 0.01087 | 3.0E-16 | <1E-16 | <1E-16 | <1E-16 | 5.395E-3 |

^aO = organic; A = aqueous.

^bOnly streams calculated by GTM are included here.

^cThese GTM flows differ from Fig. III-1 flows in that they include the effect of 0.5% other-phase carryover in all interstage flows.

^dThis stream is not marked in Fig. III-1 as it is neither a feed nor an effluent. It is the interstage organic-phase flow going from the plutonium scrub section to the first carbonate wash section.

1. Extraction and Scrub Sections

a. Extraction

The extraction section removes the TRU elements from the aqueous (DF) feed so that the aqueous (DW) raffinate emerges as a nonTRU waste. The single-stage options of the GTM were used to calculate the distribution ratios for Pu⁴⁺, UO₂²⁻, and Am³⁺ over the range of processing conditions in the extraction section. These distribution ratios, typically called D values, represent the concentration of a component in the organic phase divided by its concentration in the aqueous phase when the two phases are at equilibrium. For nitric acid concentrations ranging from 1 to 7.5M, the lowest D values obtained were for Am at 7.5M HNO₃: D_{Am} was between 19 and 20 at 7.5M HNO₃. Thus, for initial design purposes, we assumed a constant D_{Am} value of 18.

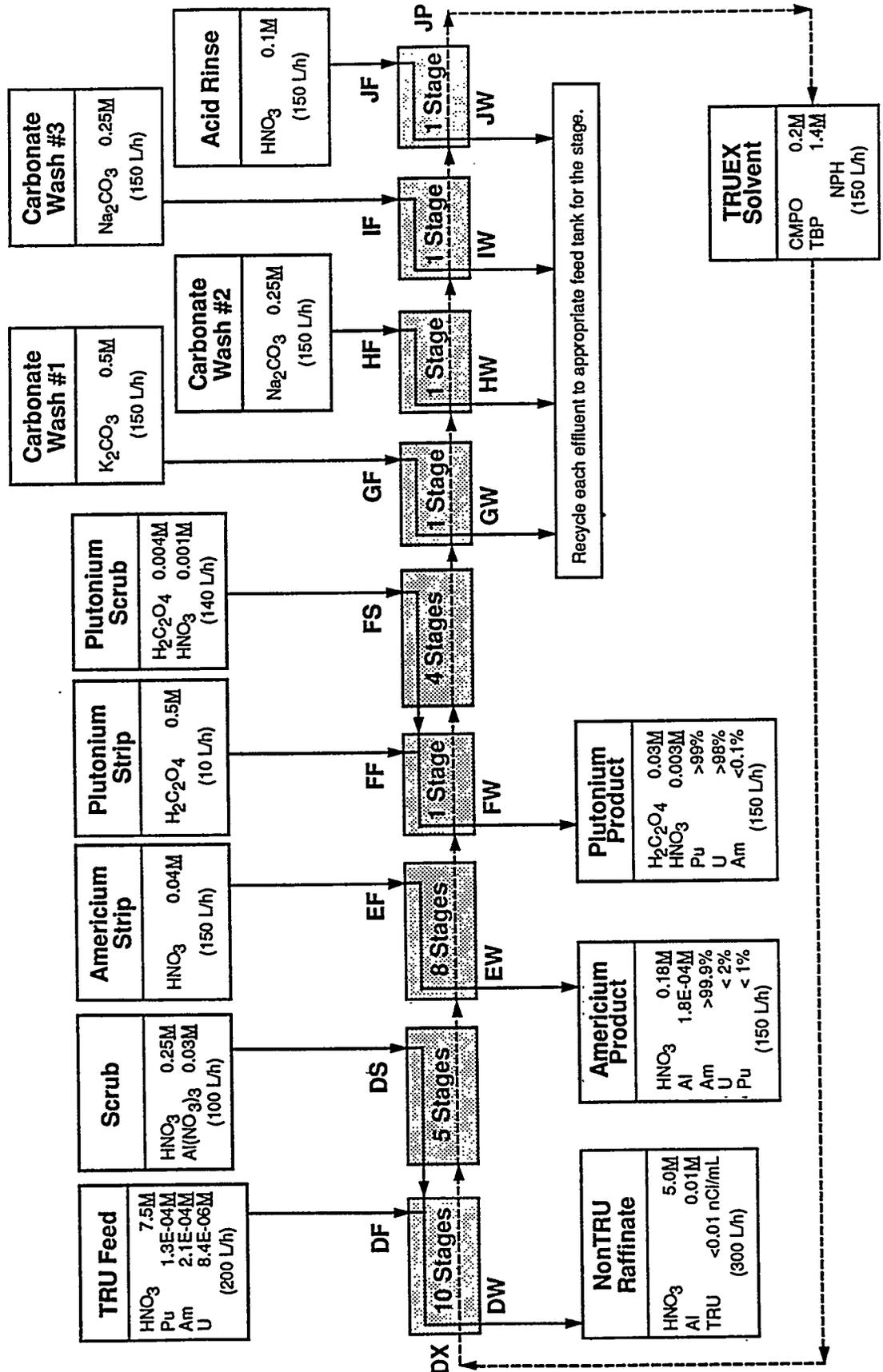


Fig. III-1. Base-Case Flowsheet for RFP Waste Developed by Using the GTM

Using the minimum D_{Am} value (18) and the maximum Am concentration ($2.1 \times 10^{-4}M$) in the aqueous (DF) feed and assuming no other-phase carryover, a simple extraction section model was developed to show the interaction of the organic-to-aqueous (O/A) flow ratio (R), the number of extraction stages, and the Am concentration in the organic feed ($[Am]_{DX}$)[LEONARD]. The results for $[Am]_{DX}$ values of $1 \times 10^{-10}M$ (10 times higher than the upper bound set by the design criteria) and $1 \times 10^{-11}M$ (right at the design limit) are shown in Fig. III-2 and III-3, respectively. Note that no matter how many stages there are or how high the R value, the minimum americium concentration in the aqueous (DW) raffinate is $[Am]_{DX} / D_{Am}$. Thus, for both figures, $[Am]_{DW}$ bottoms out below the design value of $1.22 \times 10^{-11}M$ Am in DW as long as the $[Am]$ in DX is less than $1 \times 10^{-10}M$. This sets the upper limit for the TRU concentration in the recycled solvent no matter how many stages are used in the extraction section. We needed an R value low enough to get a satisfactory aqueous flow in the scrub and high enough to reduce the number of extraction stages, so we chose an extraction section R value of 0.5 for our base-case design. Figure III-3 shows that we can meet our design goal of 0.01 nCi/mL if we use eight stages. We added two more stages to allow for process variations, for a total of 10 extraction stages for the base-case design.

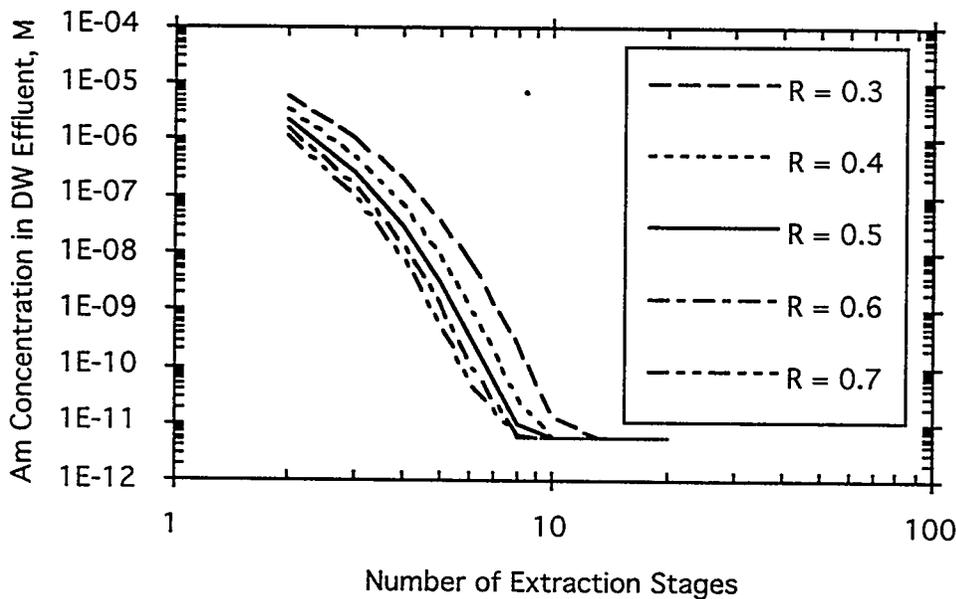


Fig. III-2. Effect of the Number of Extraction Stages and the O/A Flow Ratio (R) on the Am Concentration in the Aqueous (DW) Raffinate for $1 \times 10^{-10}M$ Am in the Organic (DX) Feed. These calculations for the extraction section are based on $2.1 \times 10^{-4}M$ Am in the aqueous (DF) feed.

As shown in Fig. III-1, no nonTRU metal ions are present in the TRU (DF) feed used in the GTM model of the TRU EX process. Since all metal ions would act to increase D_{Am} , leaving them out represents a worst-case situation. The omitted ions include not only Mg^{2+} , Al^{3+} , and Fe^{3+} , but also Na^+ , Ca^{2+} , and Cr^{3+} . As these elements generally are not extracted, they will all end up in the aqueous (DW) raffinate. The other components of the TRU feed (F^- , SO_4^{2-} , NO_2^- , Cl^- , NH_4^+ , and SiO_2) generally do not affect D_{Am} either and will also end up in the DW raffinate. The one exception is F^- without Al^{3+} present. In this case, D_F is close to 1.0, so uncomplexed F^- will move into the scrub section,

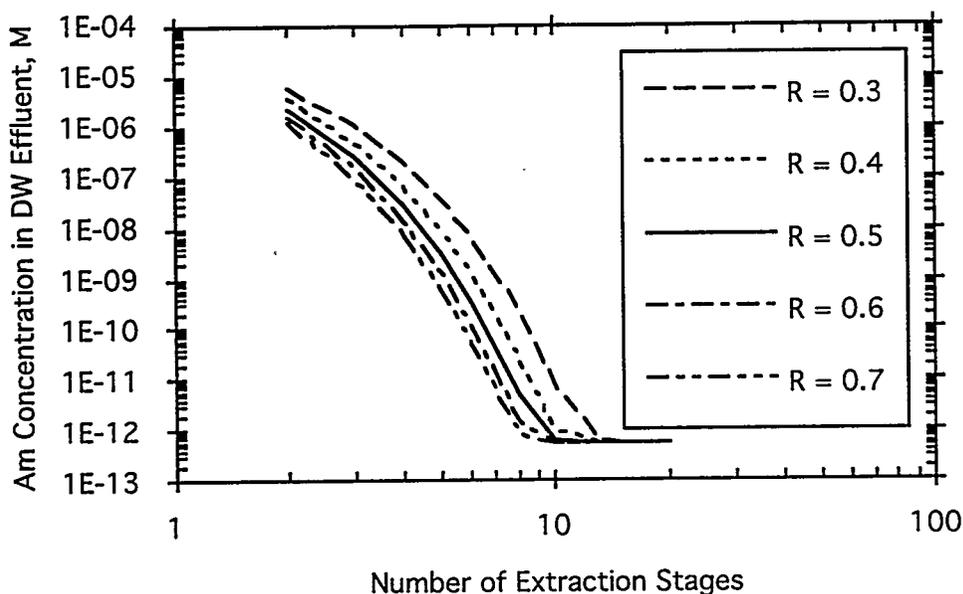


Fig. III-3. Effect of the Number of Extraction Stages and the O/A Flow Ratio (R) on the Am Concentration in the Aqueous (DW) Raffinate for $1 \times 10^{-11} \text{M}$ Am in the Organic (DX) Feed. These calculations for the extraction section are based on $2.1 \times 10^{-4} \text{M}$ Am in the aqueous (DF) feed.

where it can cause corrosion or precipitate AmF_3 as the acid concentration drops. To prevent this, $\text{Al}(\text{NO}_3)_3$ is added to the scrub (DS) feed so that the aluminum concentration will be 0.03M . This aluminum will complex the small amounts of fluoride that might be in the TRU (DF) feed and keep the fluoride ion from being carried through the scrub section by the organic phase.

b. Scrub

The purpose of the scrub section is to remove most of the nitric acid from the organic phase before it enters the Am strip section so that the Am can be stripped easily. By running the single-stage options of the GTM (options 4 and 5), as well as some scrub section cases (option 7), we determined that D_{HNO_3} was approximately 0.29 in the DF feed stage and 0.34 ± 0.05 in the scrub section.

Using this typical D_{HNO_3} value for the scrub section (0.34) and the maximum HNO_3 concentration in the aqueous (DF) feed, and assuming no other-phase carryover, a simple scrub section model was developed to show the interaction of the O/A flow rate (R), the number of scrub sections, and the HNO_3 concentrations in the aqueous (DF) and scrub (DS) feeds [LEONARD]. The results for $[\text{HNO}_3]_{\text{DF}} = 7.5 \text{M}$ and $[\text{HNO}_3]_{\text{DS}} = 0.25 \text{M}$ are shown in Fig. III-4. Note that no matter how many stages or how low the R value or how high $[\text{HNO}_3]_{\text{DF}}$, the minimum nitric acid concentration in the organic phase leaving the section is $[\text{HNO}_3]_{\text{DS}} \cdot D_{\text{HNO}_3}$. Thus, for this case, the limit is 0.075M HNO_3 . We needed an R value high enough that the contribution of scrub (DS) flow to the aqueous (DW) raffinate is small, yet low enough that the number of scrub stages is small, so we chose a scrub section R value of 1.5 for the base-case design. Figure III-4 shows that we can meet our design goal of 0.13M HNO_3 in

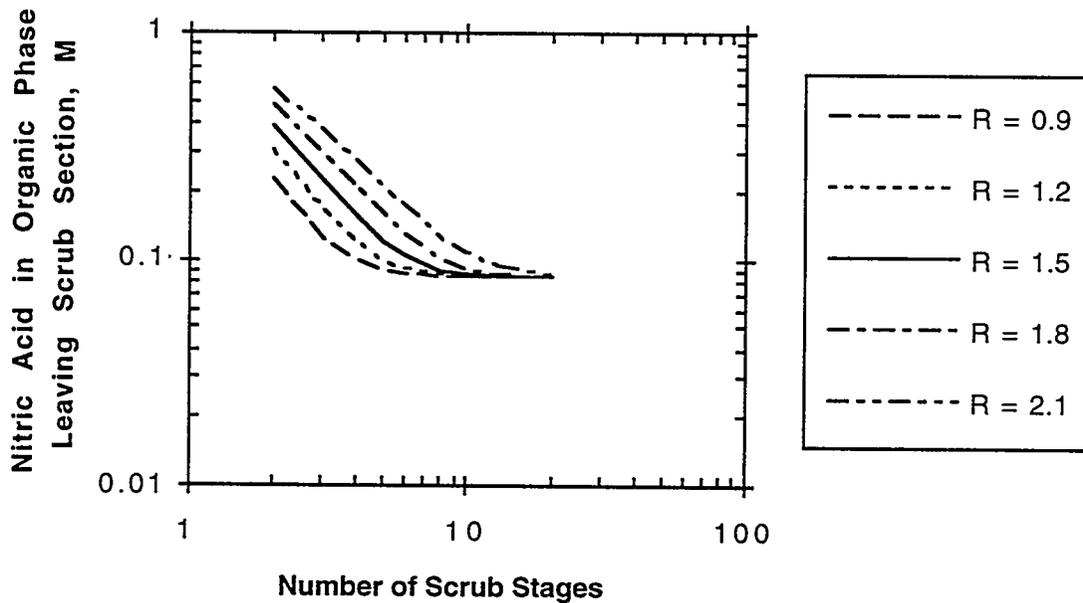


Fig. III-4. Effect of the Number of Scrub Stages and the O/A Flow Ratio (R) on the HNO_3 Concentration in the Organic Phase Exiting the Scrub Section. These calculations are based on 7.5M HNO_3 in the aqueous (DF) feed and 0.25M HNO_3 in the scrub (DS) feed.

the organic phase leaving the scrub section if we use five stages. In this case, we did not add extra scrub stages because this HNO_3 concentration is balanced between lower values that will allow too much Pu into the Am (EW) product and higher values that will make the Am harder to strip in the Am strip section.

With the exception of the Al^{3+} added to scrub out any fluoride ion, the metal ions in the scrub section will be the Am^{3+} , Pu^{4+} , and UO_2^{2-} ions. They will exit the scrub section and enter the Am strip section along with a small amount of HNO_3 , which can be determined from Fig. III-4. As the concentration of nitric acid in the aqueous (DF) feed ($[\text{HNO}_3]_{\text{DF}}$) drops, the slope of the curves in Fig. III-4 will also drop. However, the minimum nitric acid concentration that is possible in the organic phase exiting from the scrub section ($[\text{HNO}_3]_{\text{DP}}$) will stay the same. Thus, since the design range for $[\text{HNO}_3]_{\text{DP}}$ is 0.07 to 0.2M , one can see from Fig. III-4 that $[\text{HNO}_3]_{\text{DP}}$ will not drop below its design value even if $[\text{HNO}_3]_{\text{DF}}$ drops all the way to 1.0M .

2. Stripping Sections

a. Am Strip

Because the solubility of Am in an oxalic acid solution was thought to be low, the need for an Am strip was considered. Assuming that the solubility of La oxalate [LIDE] to be the same as that for Am oxalate, the solubility of Am oxalate is estimated to be $1 \times 10^{-6}\text{M}$. Thus, since Am concentrations can be as high as $2.1 \times 10^{-4}\text{M}$ in the aqueous (DF) feed, an Am strip is clearly needed. The design criterion for the Am strip is that it should recover 99.9% of the Am. As a result, the maximum Am concentration in the Pu strip will be about $3 \times 10^{-7}\text{M}$, well below the solubility limit for Am oxalate.

Since the Am strip is too complex for a simple model, in that the D_{Am} value drops sharply as the $[HNO_3]$ drops, the GTM (option 7) was used to model this section for a variety of process conditions. The effect of the total number of stages for the Am strip on the $[Am]$ in each stage is shown in Figs. III-5 and III-6 for the aqueous and organic phases, respectively. In these figures, $[HNO_3]$ in the organic phase leaving the strip section and going to the Am strip section is $0.15M$, the $[HNO_3]$ in the Am strip (EF) feed is $0.04M$, and the O/A flow ratio (R) is 1.0. Note that the $[Am]$ in the aqueous phase first increases, then decreases. This happens because the $[HNO_3]$ in the organic phase entering this section is high. This high acid concentration keeps the distribution coefficient for Pu high in the first Am strip stage so that less than 1% of the Pu is lost in the EW effluent.

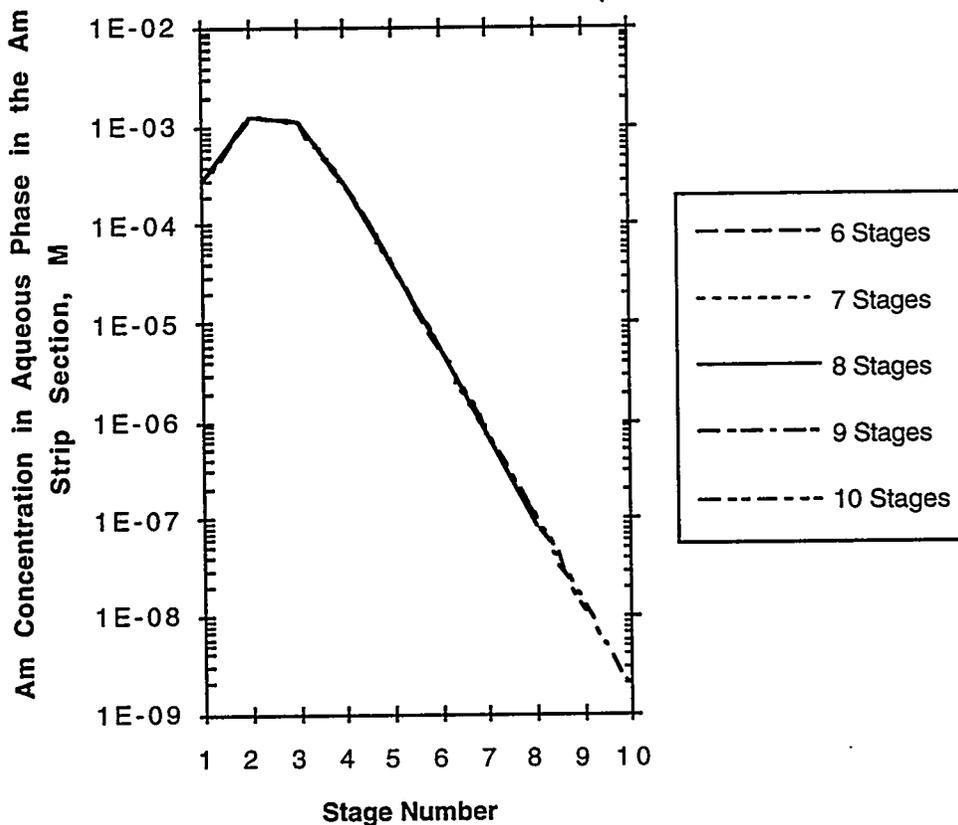


Fig. III-5. Effect of the Total Number of Am Strip Stages on the Am Concentration Profile in the Aqueous Phase of the Am Strip Stages

For design purposes, we focus on the $[Am]$ in the organic phase leaving the Am strip section, which we can get from Fig. III-6. This figure shows that we can meet our design goal of $3 \times 10^{-7}M$ Am in the solvent leaving this section if we use six stages. We added two more stages to allow for process variations, for a total of eight Am strip stages for the base-case design.

b. Pu Strip and Scrub

The designs of the Pu strip and scrub sections are driven by the need to prevent the precipitation of Pu oxalate in the first strip stage. A model for $Pu(C_2O_4)_2$ solubility was developed

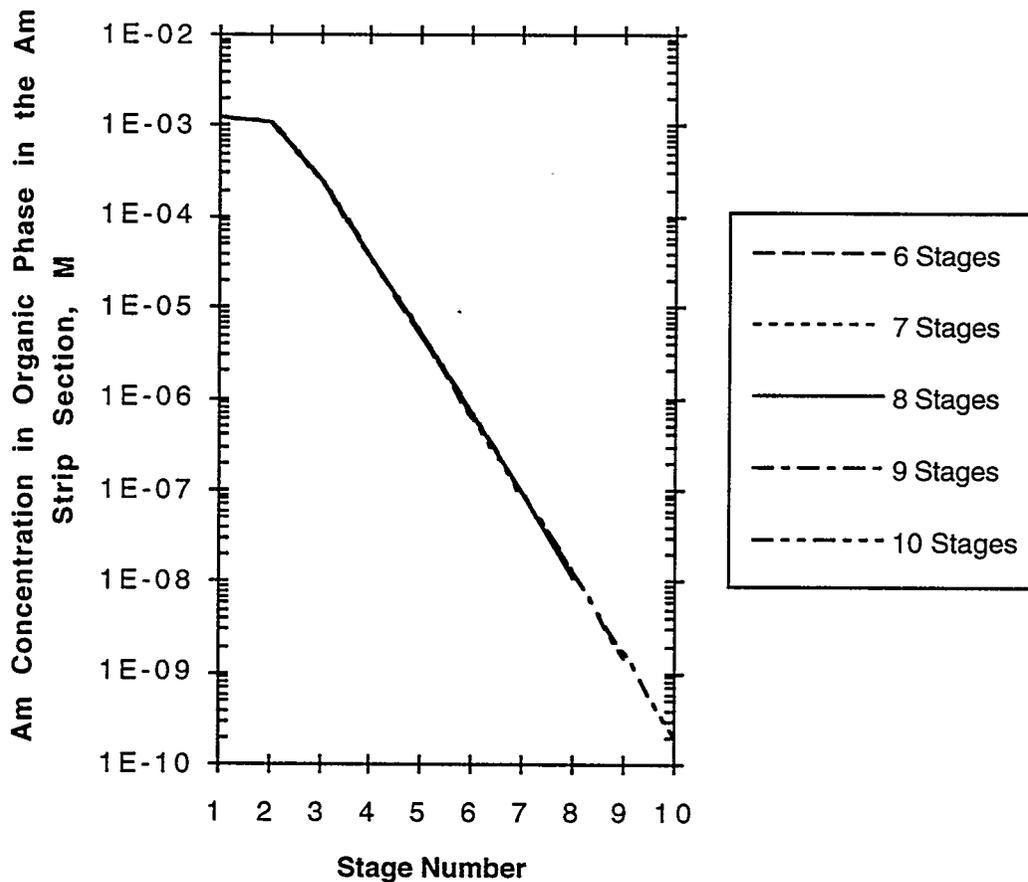


Fig. III-6. Effect of the Total Number of Am Strip Stages on the Am Concentration Profile in the Organic Phase of the Am Strip Stages

and compared with experimental data (Table III-3). The numbers generally agree within a factor of 2. For design purposes, we applied a factor of 0.2 to all the Pu solubility values from the Pu solubility model. These modified values are also listed in Table III-3. Since the maximum value for [Pu] in DF can be $1.3 \times 10^{-4} \text{M}$, the [Pu] in the first Pu strip stage can be as high as $1.73 \times 10^{-4} \text{M}$. From Table III-3, one can see that if $[\text{H}_2\text{C}_2\text{O}_4]$ is between 0.02 and 0.05M and if $[\text{HNO}_3]$ is between 0.002 and 0.005M, Pu solubility with the design factor included will be greater than $1.04 \times 10^{-4} \text{M}$. Thus, the precipitation of Pu oxalate should not occur within the centrifugal contactor used to carry out the TRUEX process.

The four Pu scrub stages that are a part of the overall Pu stripping operation get the Am and Pu concentrations down to less than 0.01 nCi/mL in the organic phase going to the solvent cleanup sections. This actually occurs in the first two Pu scrub stages. The last two Pu scrub stages provide a safety factor and also reduce the amount of $\text{H}_2\text{C}_2\text{O}_4$ and HNO_3 that are carried by the organic phase into the solvent cleanup sections. This reduces the rate at which the K_2CO_3 in the first carbonate wash is neutralized by these two acids. As noted above, if some Pu and Am are tied up in the solvent by solvent degradation products, the total Pu and Am concentrations in the organic phase going to the solvent cleanup sections could be greater than 0.01 nCi/mL.

Table III-3. Effect of Nitric Acid and Oxalic Acid Concentrations on Pu Oxalate Solubility in the Aqueous Phase

| [H ₂ C ₂ O ₄], <u>M</u> | [HNO ₃], <u>M</u> | Pu Solubility | | | |
|--|----------------------------------|------------------------------|-----------------------------|--|----------|
| | | Actual, ^a mg/L | Model, ^b mg/L | Model with Design Factor ^c | |
| | | | | mg/L | <u>M</u> |
| 0.01 | 1E-07 | | 1562 | 312 | 1.31E-03 |
| | 1E-05 | | 1562 | 312 | 1.31E-03 |
| | 0.001 | | 1416 | 283 | 1.18E-03 |
| | 0.002 | | 1293 | 259 | 1.08E-03 |
| | 0.005 | | 1017 | 203 | 8.5E-04 |
| | 0.01 | | 735 | 147 | 6.2E-04 |
| | 0.02 | | 455 | 91 | 3.8E-04 |
| | 0.05 | | 183 | 37 | 1.5E-04 |
| | 0.1 | | 75 | 15 | 6.3E-05 |
| | 0.2 | | 27 | 5 | 2.3E-05 |
| | 0.5 | | 7 | 1 | 5.9E-06 |
| 0.015 | 1.61 | 15 | 3 | 1 | 2.5E-06 |
| 0.02 | 1E-07 | | 1604 | 321 | 1.34E-03 |
| | 1E-05 | | 1604 | 321 | 1.34E-03 |
| | 0.001 | | 1516 | 303 | 1.27E-03 |
| | 0.002 | | 1439 | 288 | 1.20E-03 |
| | 0.005 | | 1240 | 248 | 1.04E-03 |
| | 0.01 | | 993 | 199 | 8.3E-04 |
| | 0.02 | | 688 | 138 | 5.8E-04 |
| | 0.05 | | 319 | 64 | 2.7E-04 |
| | 0.1 | | 186 | 37 | 1.6E-04 |
| | 0.2 | | 51 | 10 | 4.3E-05 |
| | 0.5 | | 12 | 2 | 1.0E-05 |
| 0.05 | 1E-07 | | 1633 | 327 | 1.37E-03 |
| | 1E-05 | | 1627 | 325 | 1.36E-03 |
| | 0.001 | | 1586 | 317 | 1.33E-03 |
| | 0.002 | | 1545 | 309 | 1.29E-03 |
| | 0.005 | | 1428 | 286 | 1.19E-03 |
| | 0.01 | | 1263 | 253 | 1.06E-03 |
| | 0.02 | | 1011 | 202 | 8.5E-04 |
| | 0.05 | | 580 | 116 | 4.9E-04 |
| | 0.1 | | 293 | 59 | 2.5E-04 |
| | 0.2 | | 116 | 23 | 9.7E-05 |
| | 0.5 | 26.6 | 27 | 5 | 2.3E-05 |
| | 1 | 13.3 | 9 | 2 | 7.5E-06 |
| | 2 | 9.7 | 4 | 1 | 3.3E-06 |
| 0.25 | 0.5 | 97 | 58 | 12 | 4.9E-05 |
| 0.25 | 0.75 | 58 | 40 | 8 | 3.3E-05 |
| 0.4 | 0.78 | 144 | 83 | 17 | 6.9E-05 |
| 0.6 | 0.5 | 361 | 233 | 47 | 1.9E-04 |

^aSee [WICK].^bBased on formation constants of the mono-, bis-, and tris-oxalato species from [KATZ].^cApply a design factor of 0.2 to all model values. This factor looks to be conservative; that is, the solubilities in these two columns should be lower than the actual solubility of Pu oxalate.

3. Solvent Cleanup Sections

The solvent cleanup sections consist of three carbonate wash sections and one acid rinse section. Each section is one stage, and the aqueous feed to each stage is recycled. The first carbonate wash uses 0.5M K_2CO_3 because it is more soluble than Na_2CO_3 and so can be used at higher concentrations. Thus, it can run longer before it is neutralized by any HNO_3 and $H_2C_2O_4$ in the organic phase entering to this stage. The carbonate washes clean up the solvent by removing any acidic degradation products as well as any Am or Pu that might be bound to these degradation products. These carbonate washes have been used with TRUEX flowsheets currently being tested at Argonne National Laboratory with good success for aqueous (DW) raffinate values of 0.1 nCi/mL for the TRU elements.

The acid rinse section removes any of the carbonate wash solution entrained in the solvent, thus preventing the carbonate from transferring back to the extraction section. When the solvent has not been rinsed, we have observed foaming in the interstage lines of the extraction section.

C. Sensitivity to Process Variations

In this section, we report the results of a sensitivity analysis applied to the base-case flowsheet by using the GTM. For a more detailed discussion of the bases and the techniques used in performing such analyses, see [REGALBUTO].

For the sensitivity analysis reported here, the process feeds are as shown in Fig. III-1 for the base case. The GTM was also set to include 0.5% other-phase carryover in each phase from each stage. Each parameter examined was varied by up to $\pm 40\%$ from its base-case value. A graph for each parameter shows its effect on one of the three variables of concern, the [Am] in DW, the [Pu] in EW, and the [Am] in FW. Also shown on each graph is the upper bound for the variable of concern, that is, the value above which the process would be out of specification with respect to the design criteria. Overall, the results show the process to be very robust with respect to changes in the feed flow rates or the feed concentrations.

1. Feed Flow Rates

The variations in feed flow rates, given in Figs. III-7 through III-18, show that the base-case flowsheet can tolerate reasonable variations in the process flows. In particular, Figs. III-8 and III-9 show how if the DX flow becomes too low (20% less than the design flow), we will exceed the upper bound for an acceptable [Pu] in EW; if the organic (DX) feed flow becomes too high (25% higher than the design flow), we will exceed the upper bound for an acceptable [Am] in FW. Figure III-18 shows how if the Am strip (EF) feed flow rate becomes too low (28% less than the design flow), we will exceed the upper bound for an acceptable [Am] in FW. If the EF flow becomes too high, the volume of Am strip (EW) product will become greater than it needs to be.

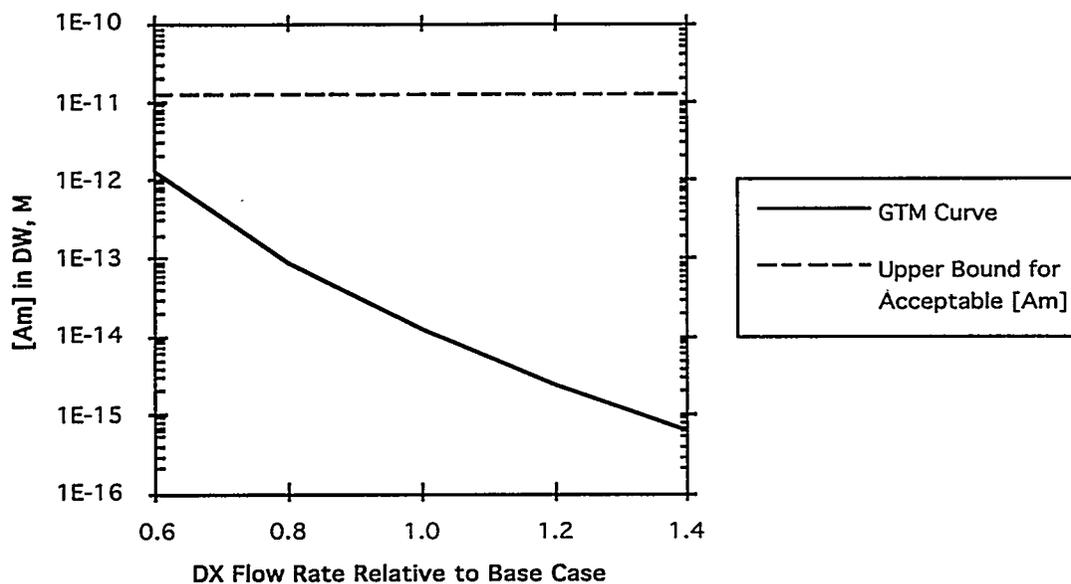


Fig. III-7. Effect of Flow Rate for DX Feed on Am Concentration in DW Raffinate. Base-case value of the flow rate for DX feed is 150 L/h.

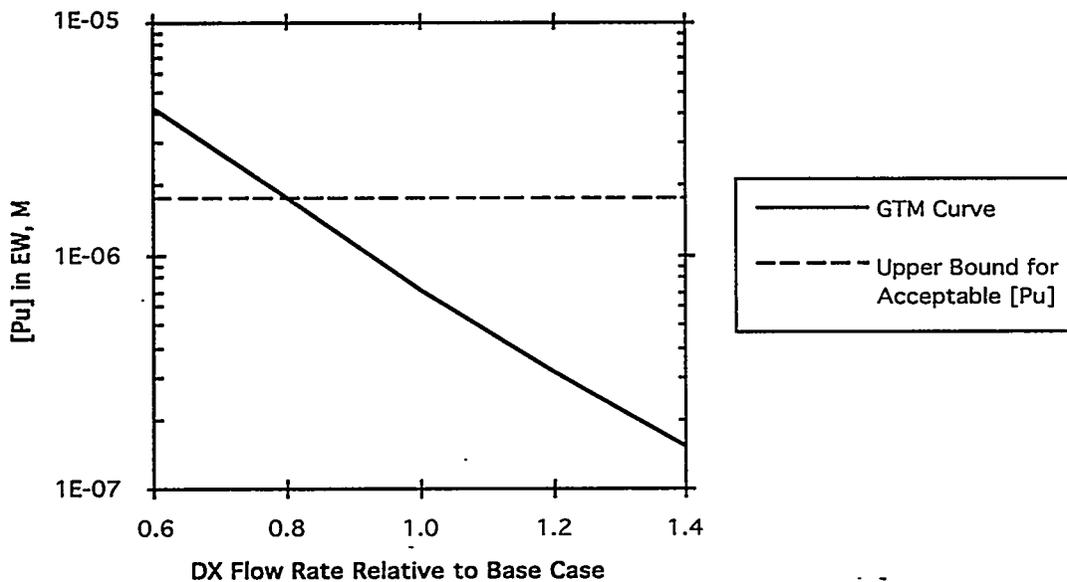


Fig. III-8. Effect of Flow Rate for DX Feed on Pu Concentration in EW Effluent. Base-case value of the flow rate for DX feed is 150 L/h.

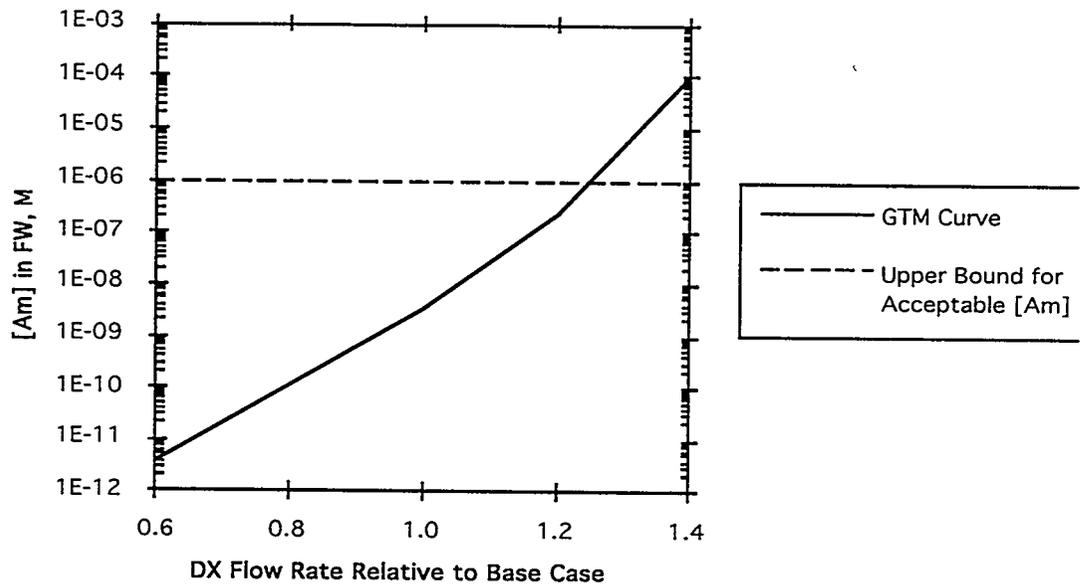


Fig. III-9. Effect of Flow Rate for DX Feed on Am Concentration in FW Effluent. Base-case value of the flow rate for DX feed is 150 L/h.

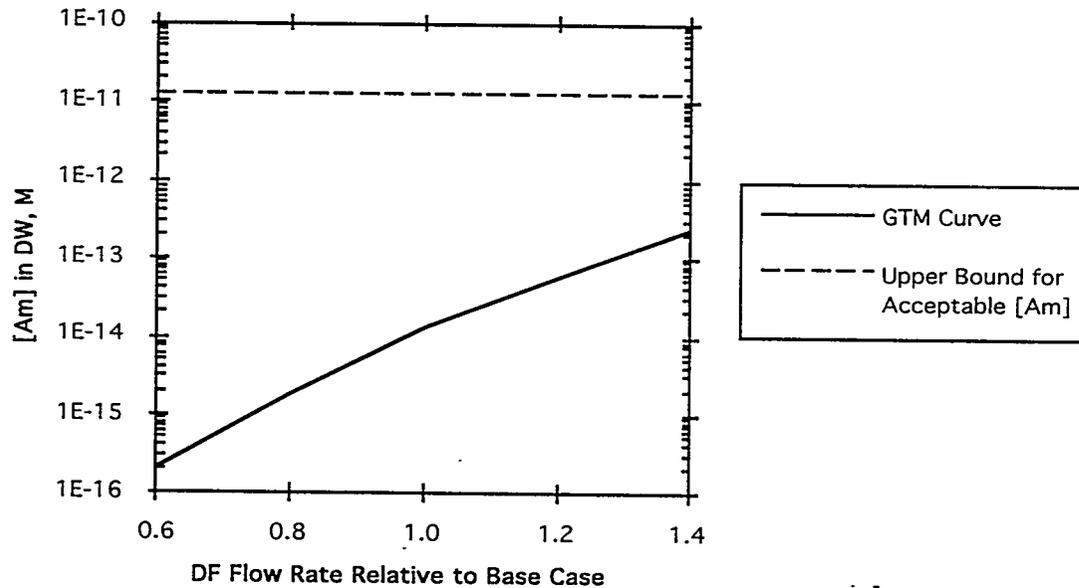


Fig. III-10. Effect of Flow Rate for DF Feed on Am Concentration in DW Raffinate. Base-case value of the flow rate for DF feed is 200 L/h.

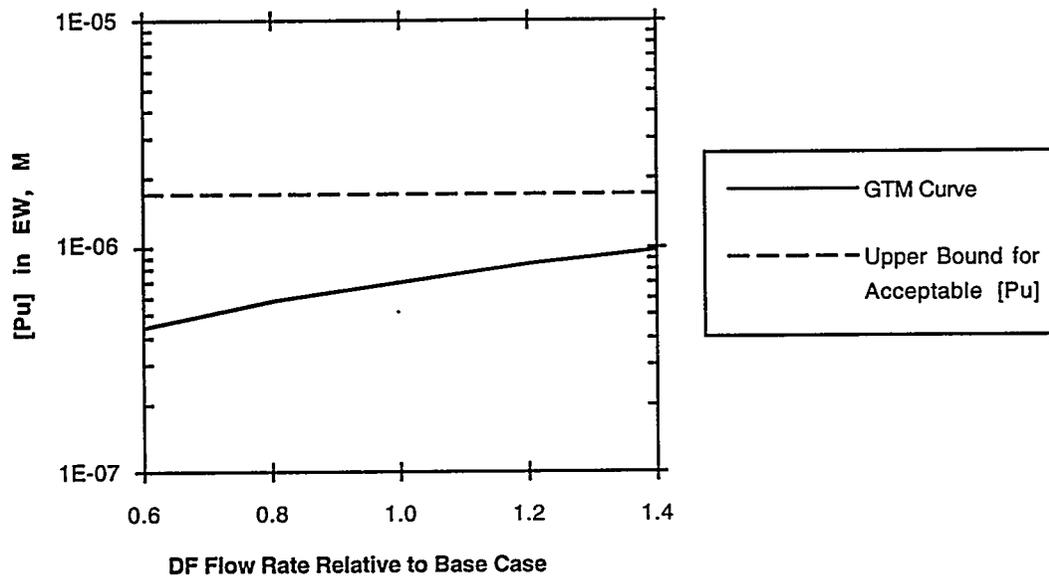


Fig. III-11. Effect of Flow Rate for DF Feed on Pu Concentration in EW Effluent. Base-case value of the flow rate for DF is 200 L/h.

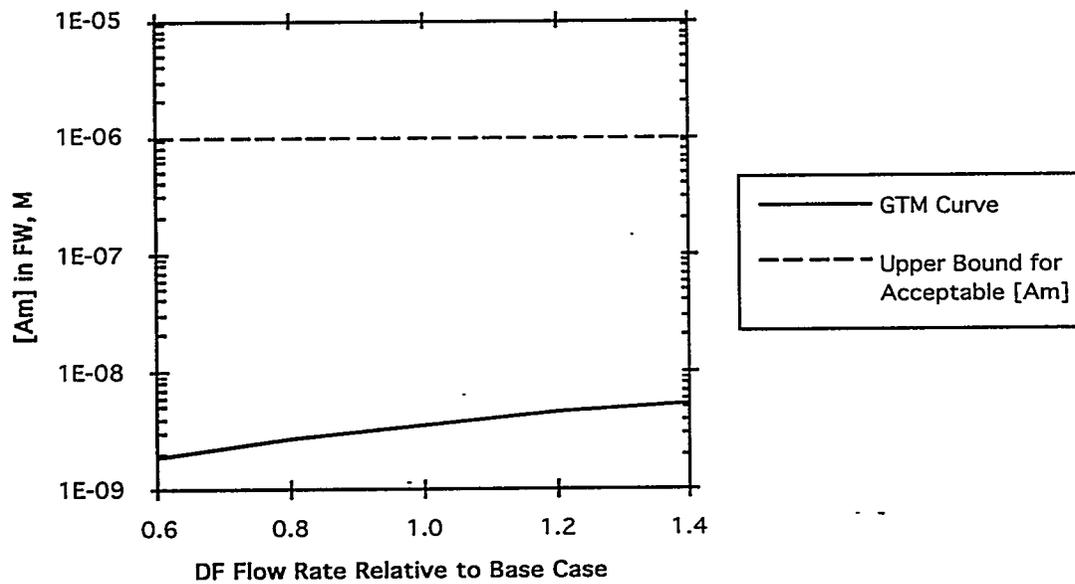


Fig. III-12. Effect of Flow Rate for DF Feed on Am Concentration in FW Effluent. Base-case value of the flow rate for DF feed is 200 L/h.

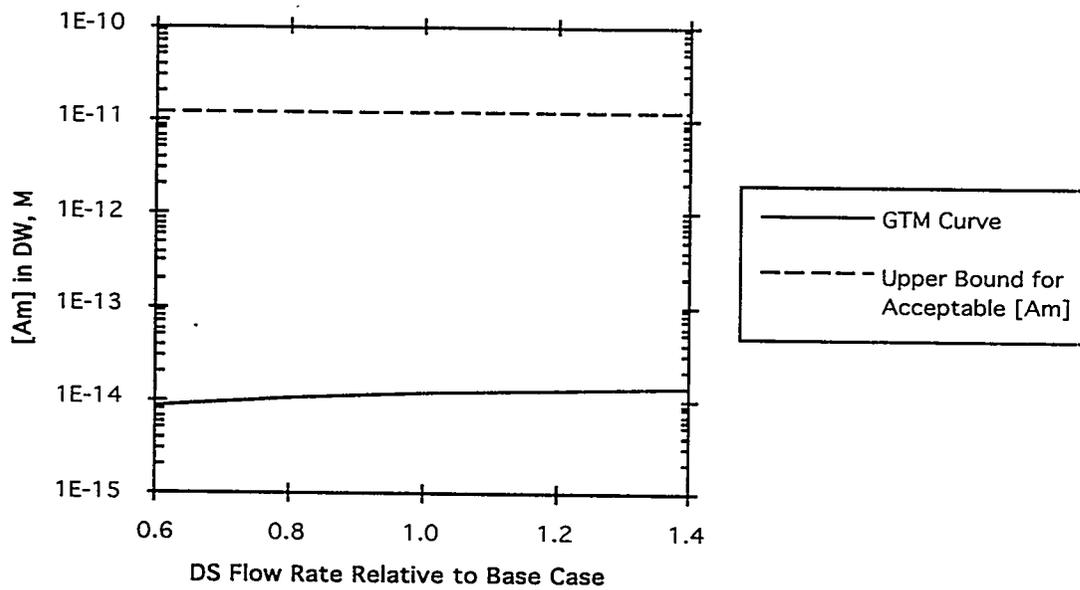


Fig. III-13. Effect of Flow Rate for DS Feed on Am Concentration in DW Raffinate. Base-case value of the flow rate for DS feed is 100 L/h.

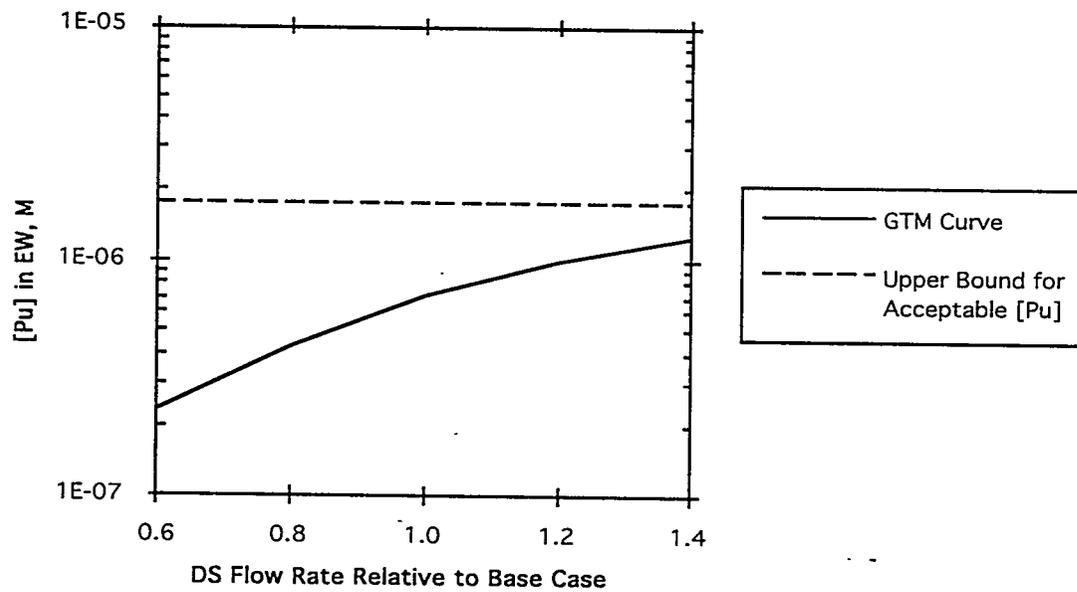


Fig. III-14. Effect of Flow Rate for DS Feed on Pu Concentration in EW Effluent. Base-case value of the flow rate for DS feed is 100 L/h.

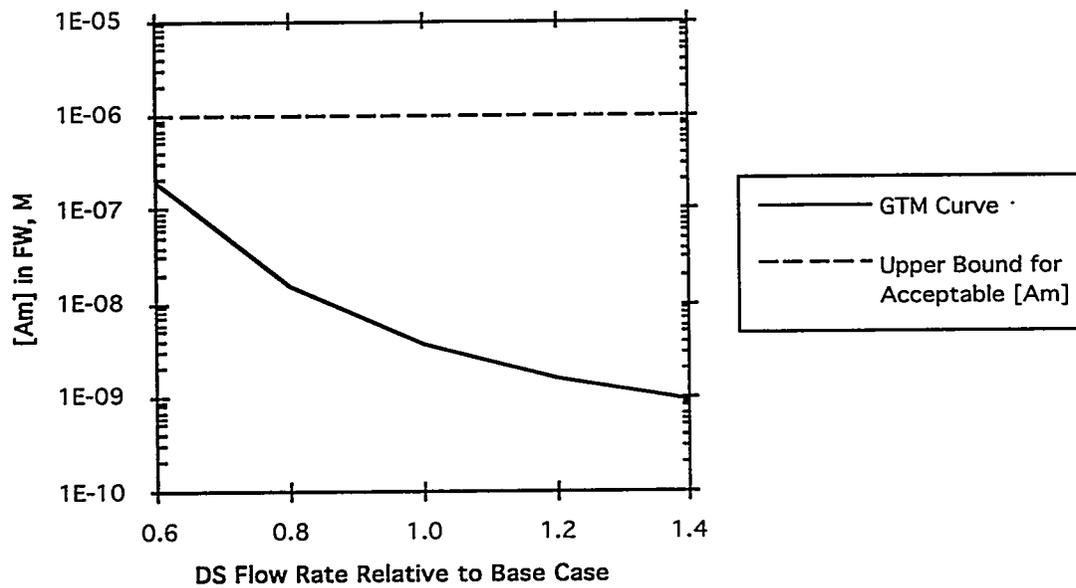


Fig. III-15. Effect of Flow Rate for DS Feed on Am Concentration in FW Effluent.
Base-case value of the flow rate for DS feed is 100 L/h.

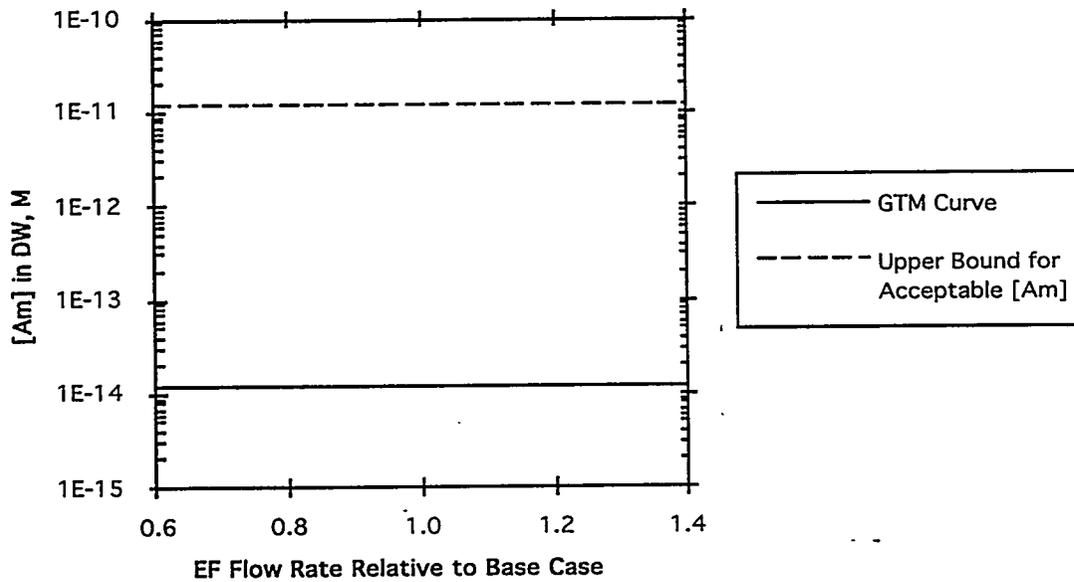


Fig. III-16. Effect of Flow Rate for EF Feed on Am Concentration in DW Raffinate.
Base-case value of the flow rate for EF feed is 150 L/h.

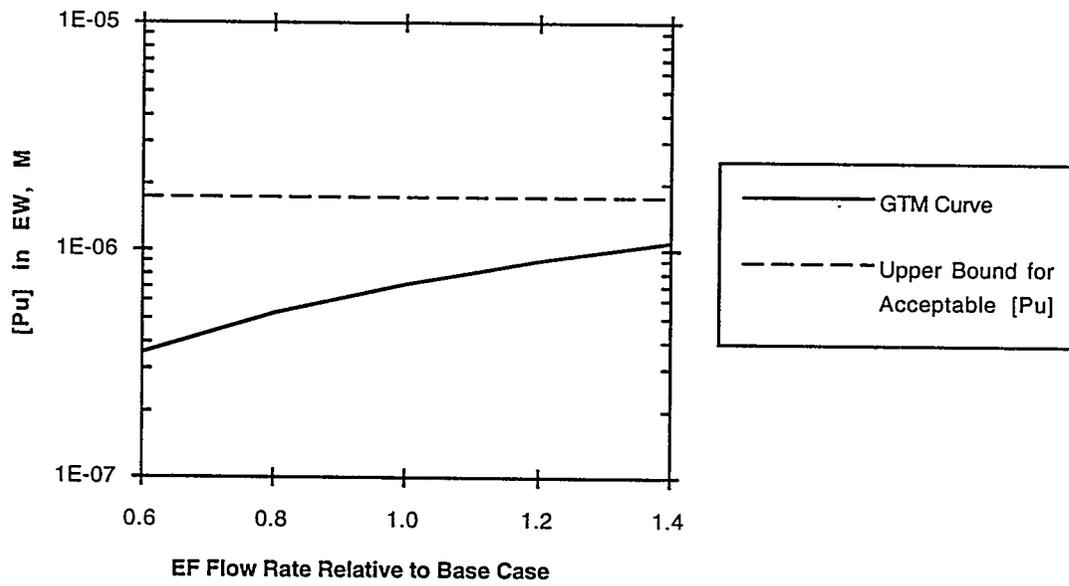


Fig. III-17. Effect of Flow Rate for EF Feed on Pu Concentration in EW Effluent. Base-case value of the flow rate for EF feed is 150 L/h.

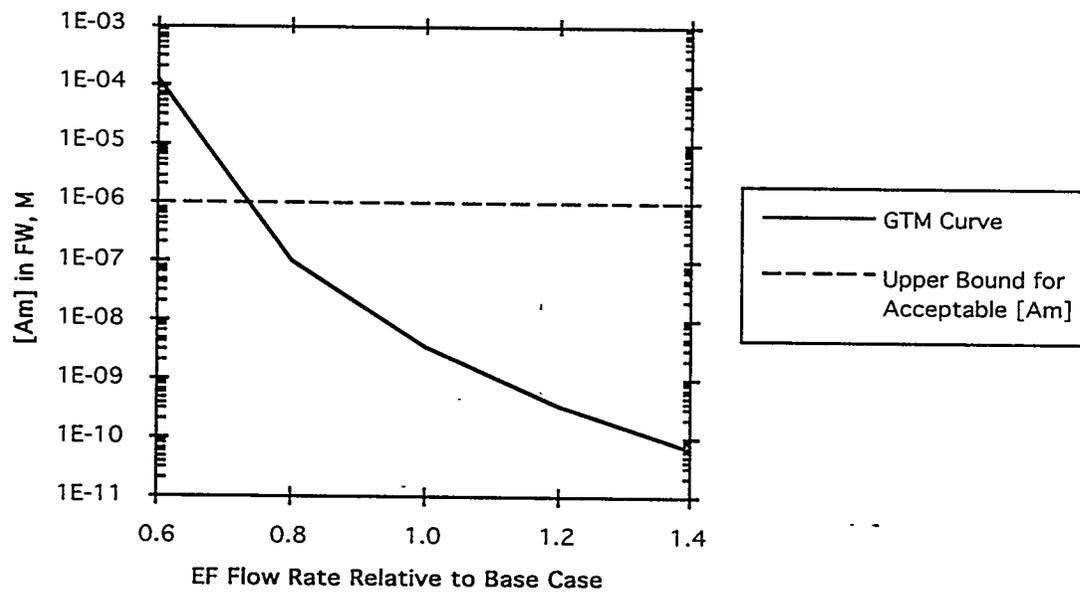


Fig. III-18. Effect of Flow Rate for EF Feed on Am Concentration in FW Effluent. Base-case value of the flow rate for EF feed is 150 L/h.

2. Feed Concentrations

The variations in feed concentrations, given in Figs. III-19 through III-24, show that the base-case flowsheet can tolerate reasonable variations in the component concentrations in the process feeds. In particular, Fig. III-20 shows how the base case process is somewhat sensitive to a low $[\text{HNO}_3]$ in the DS feed. Figure III-24 shows how the process is somewhat sensitive to a high $[\text{HNO}_3]$ in the EF feed.

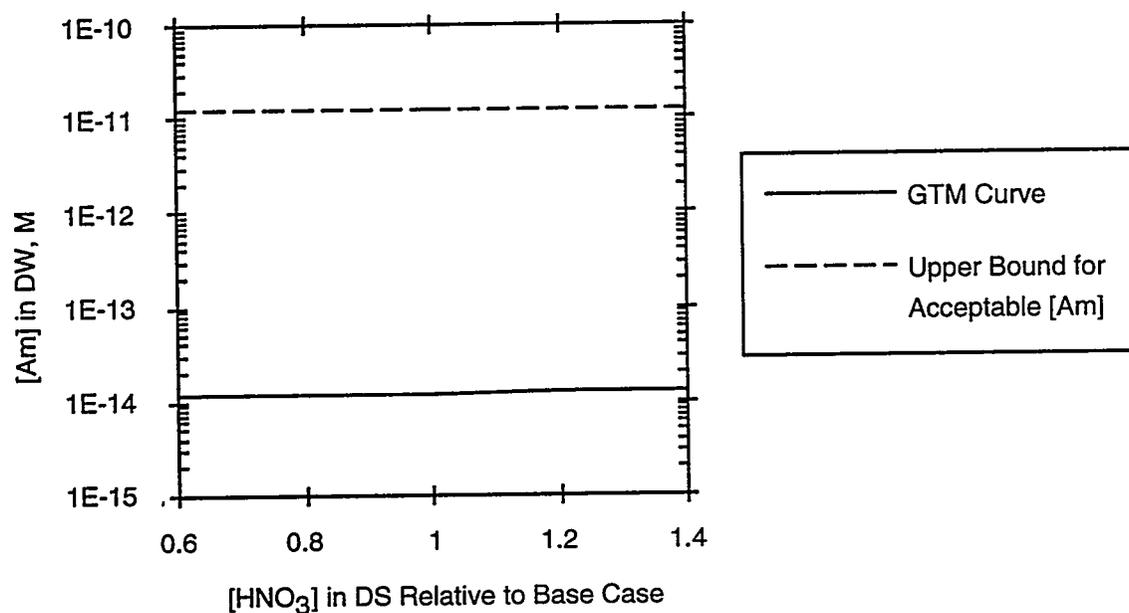


Fig. III-19. Effect of Nitric Acid Concentration in DS Feed on Am Concentration in DW Raffinate. Base-case value for nitric acid concentration in DS feed is 0.25M.

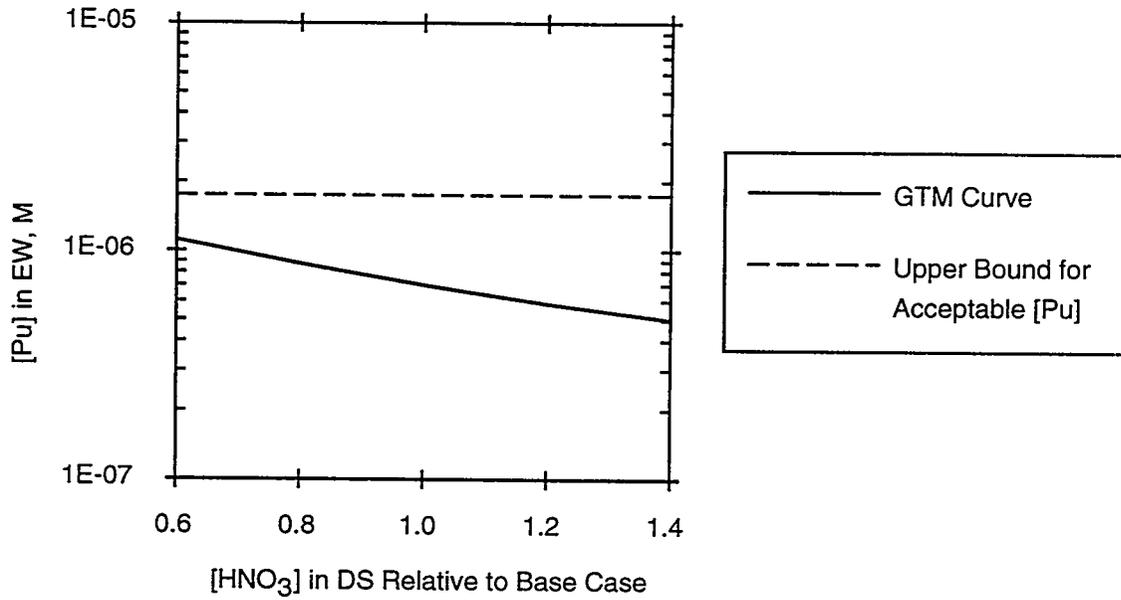


Fig. III-20. Effect of Nitric Acid Concentration in DS Feed on Pu Concentration in EW Effluent. Base-case value for nitric acid concentration in DS feed is 0.25M.

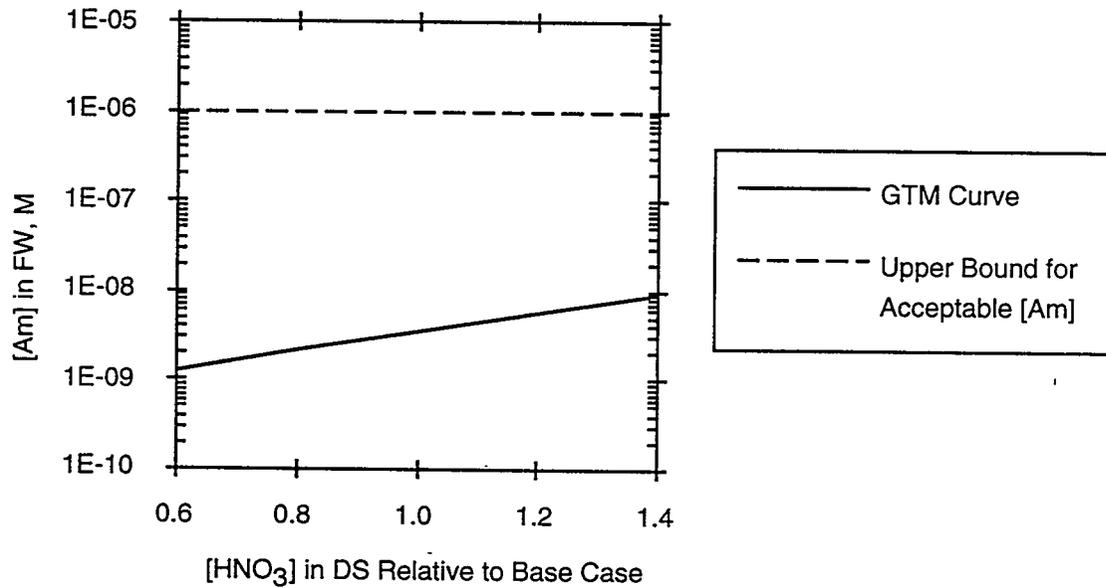


Fig. III-21. Effect of Nitric Acid Concentration in DS Feed on Am Concentration in FW Effluent. Base-case value for nitric acid concentration in DS feed is 0.25M.

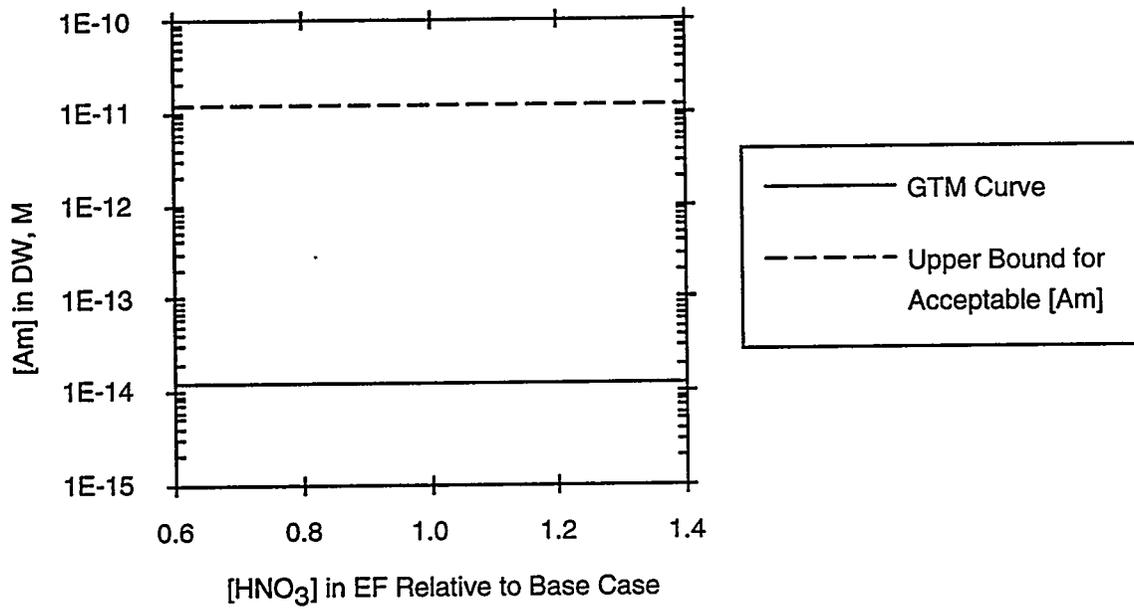


Fig. III-22. Effect of Nitric Acid Concentration in EF Feed on Am Concentration in DW Raffinate. Base-case value for nitric acid concentration in EF feed is 0.04M.

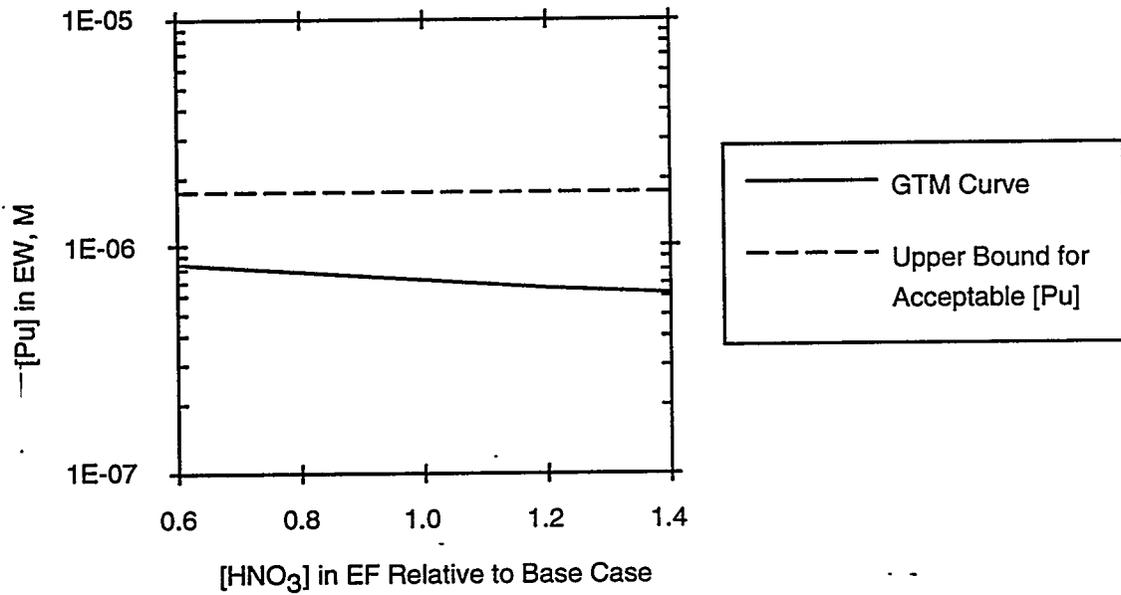


Fig. III-23. Effect of Nitric Acid Concentration in EF Feed on Pu Concentration in EW Effluent. Base-case value for nitric acid concentration in EF feed is 0.04M.

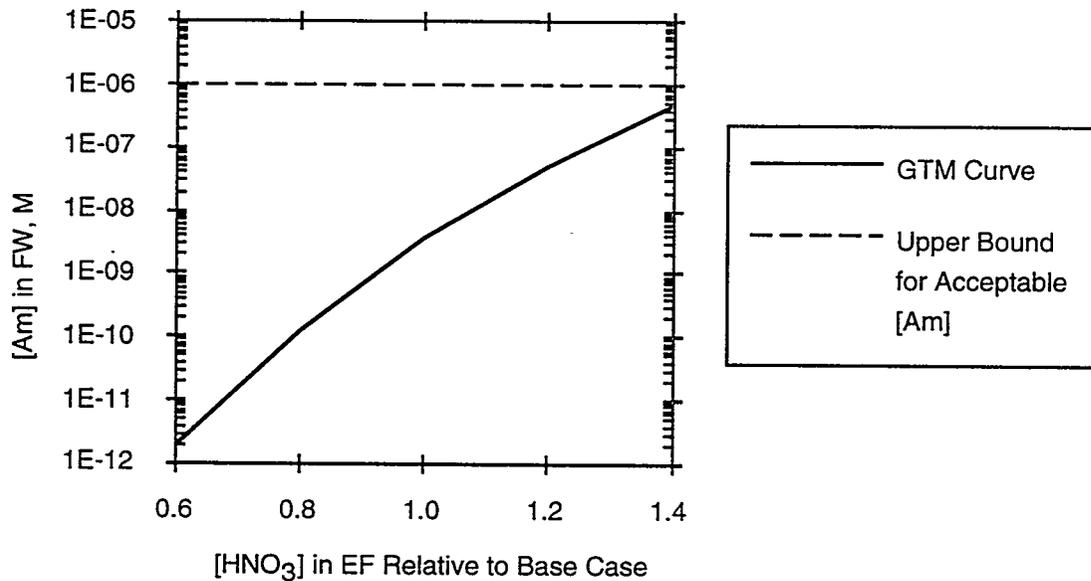


Fig. III-24. Effect of Nitric Acid Concentration in EF Feed on Am Concentration in FW Effluent. Base-case value for nitric acid concentration in EF feed is 0.04M.

3. Number of Stages

The variations in the number of stages, given in Figs. III-25 through III-36, show that the base-case flowsheet is reasonably robust and can tolerate the loss of at least one stage in each of the sections shown. However, this reduction is not recommended because the overall process becomes less tolerant of other process variations.

Figures III-29 and III-30 show how five stages are about optimum for the scrub section. If there are fewer scrub stages, the [Am] in FW will increase towards its upper bound for acceptable operation; if there are more scrub stages, the [Pu] in EW will increase towards its upper bound.

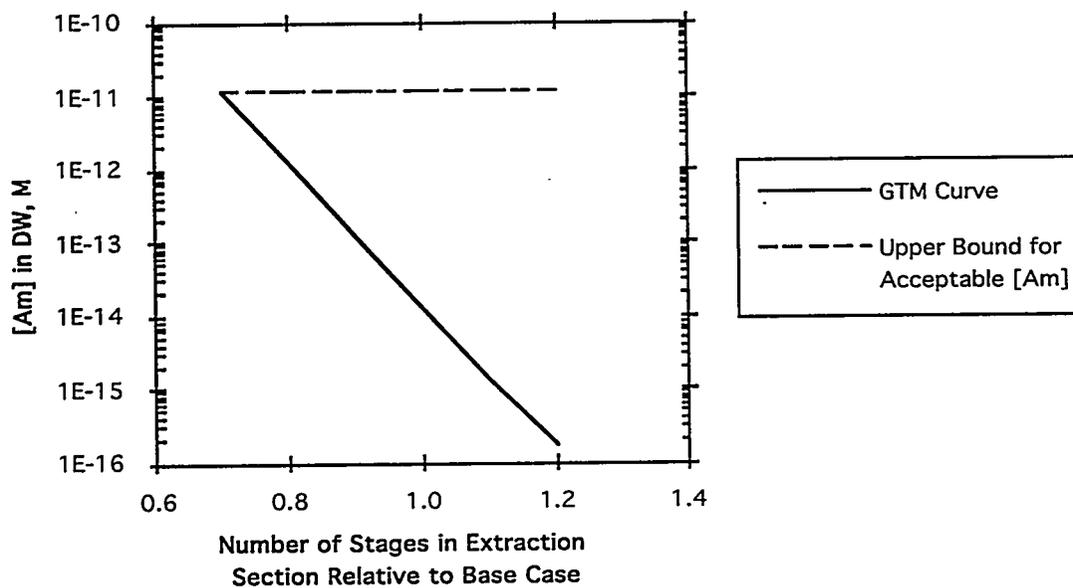


Fig. III-25. Effect of Number of Stages in Extraction Section on Am Concentration in DW Raffinate. Base-case value for number of stages in extraction section is 10 stages.

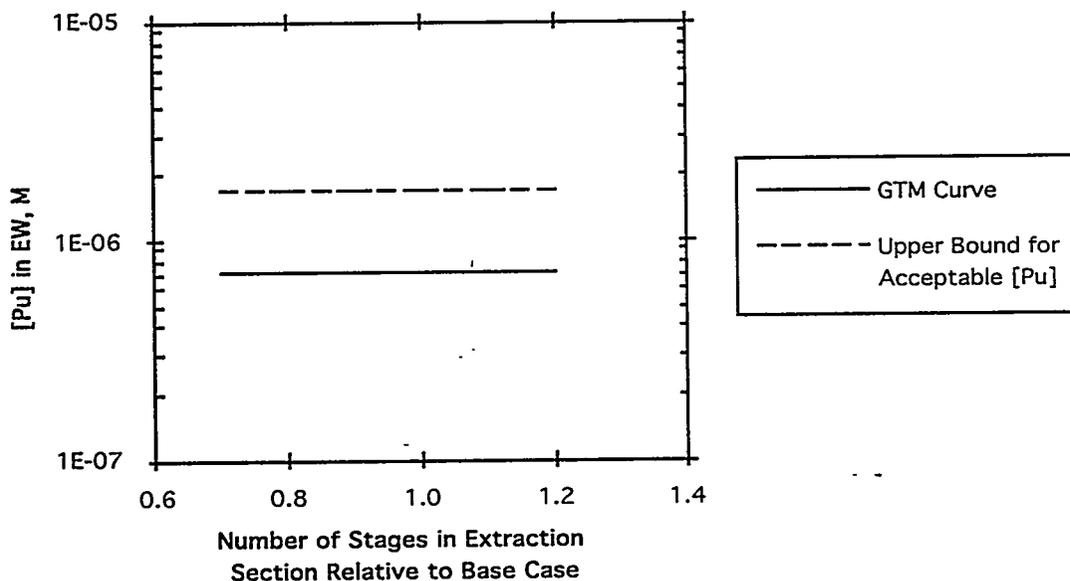


Fig. III-26. Effect of Number of Stages in Extraction Section on Pu Concentration in EW Effluent. Base-case value for number of stages in extraction section is 10 stages.

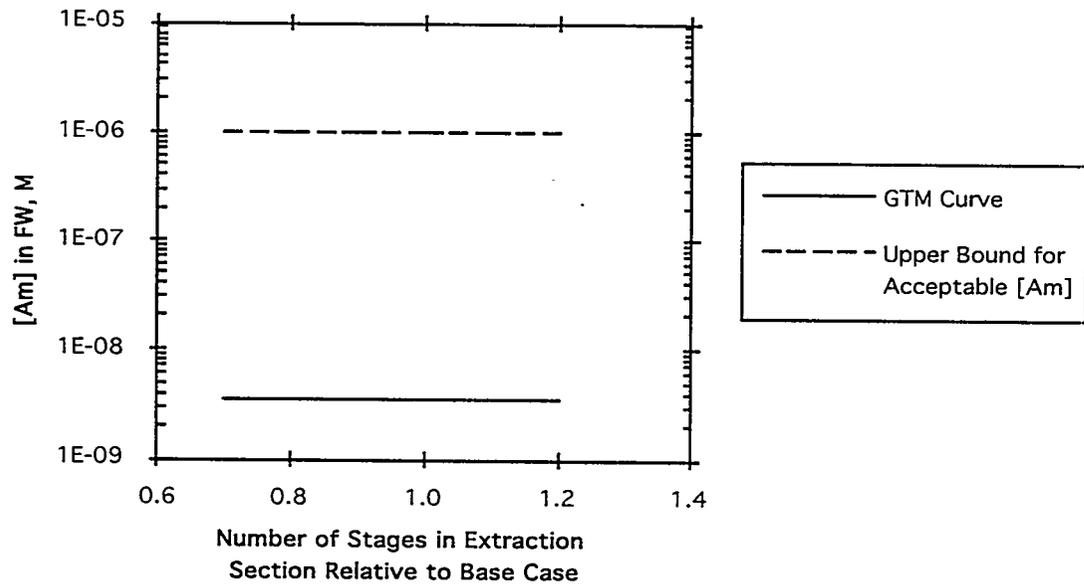


Fig. III-27. Effect of Number of Stages in Extraction Section on Am Concentration in FW Effluent. Base-case value for number of stages in extraction section is 10 stages.

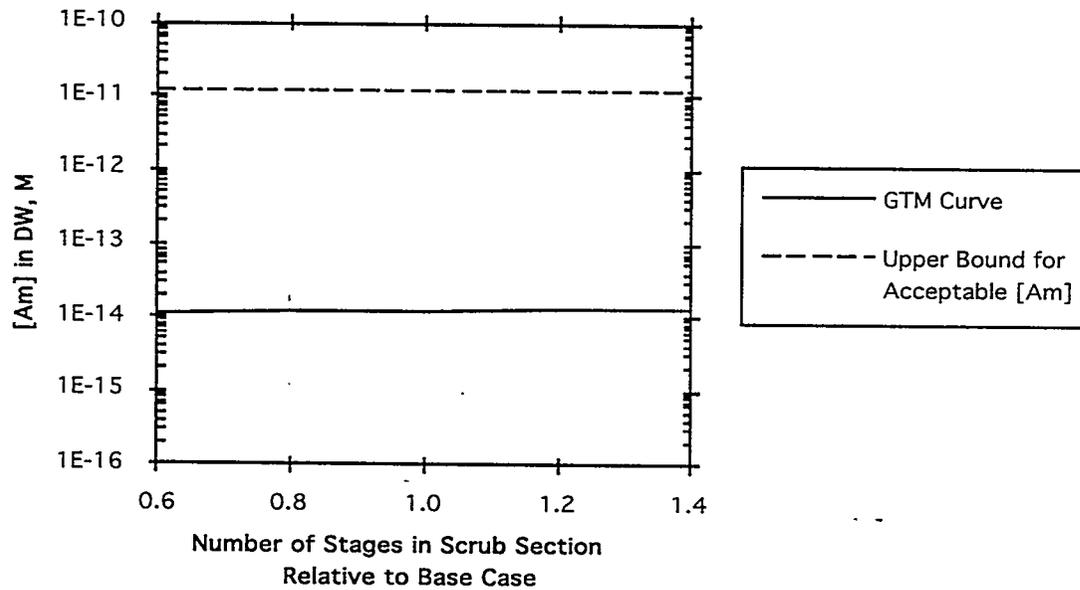


Fig. III-28. Effect of Number of Stages in Scrub Section on Am Concentration in DW Raffinate. Base-case value for number of stages in scrub section is five stages.

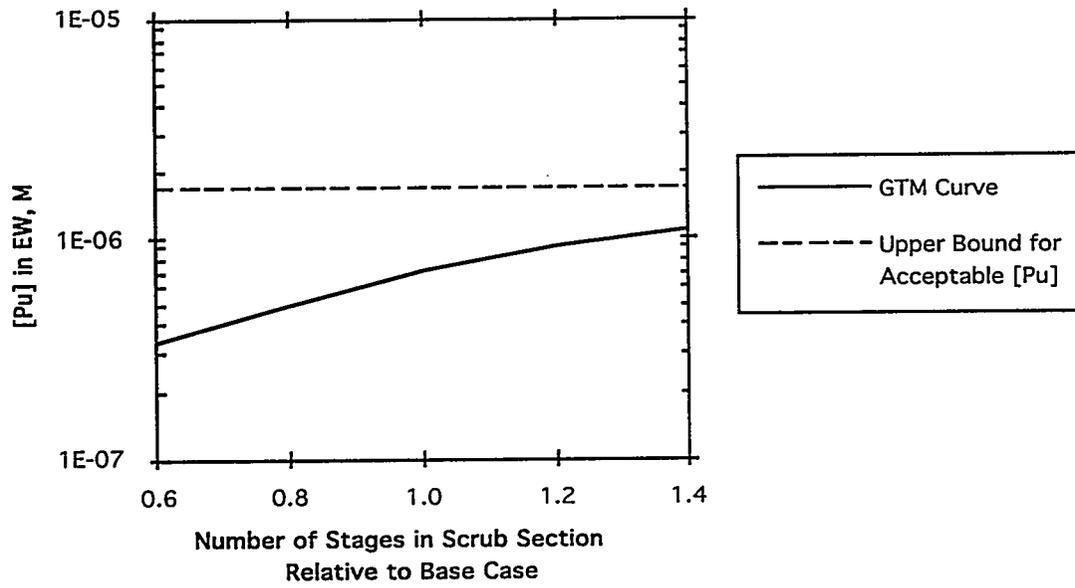


Fig. III-29. Effect of Number of Stages in Scrub Section on Pu Concentration in EW Effluent. Base-case value for number of stages in scrub section is five stages.

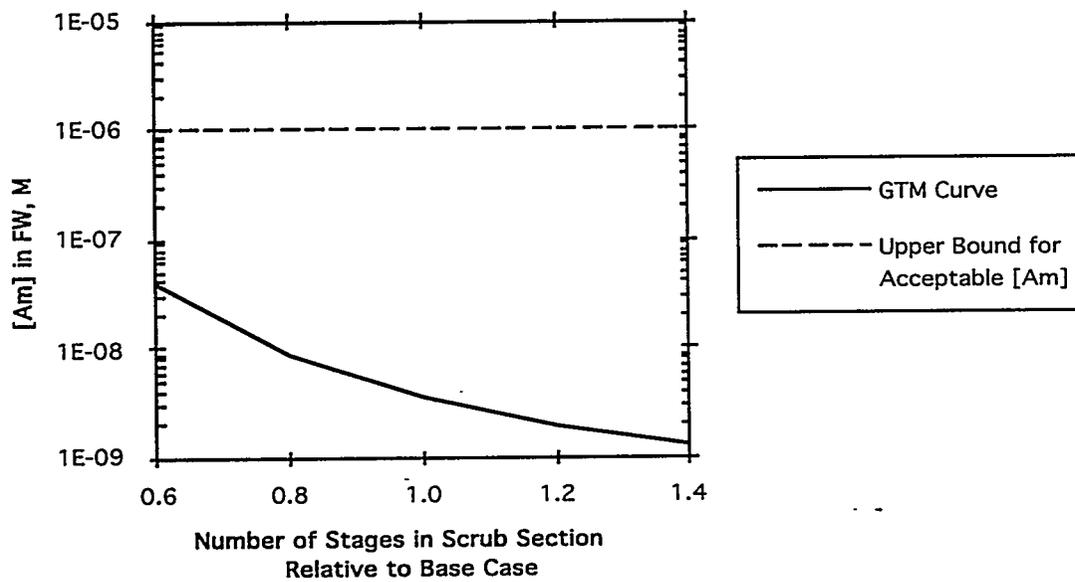


Fig. III-30. Effect of Number of Stages in Scrub Section on Am Concentration in FW Effluent. Base-case value for number of stages in scrub section is five stages.

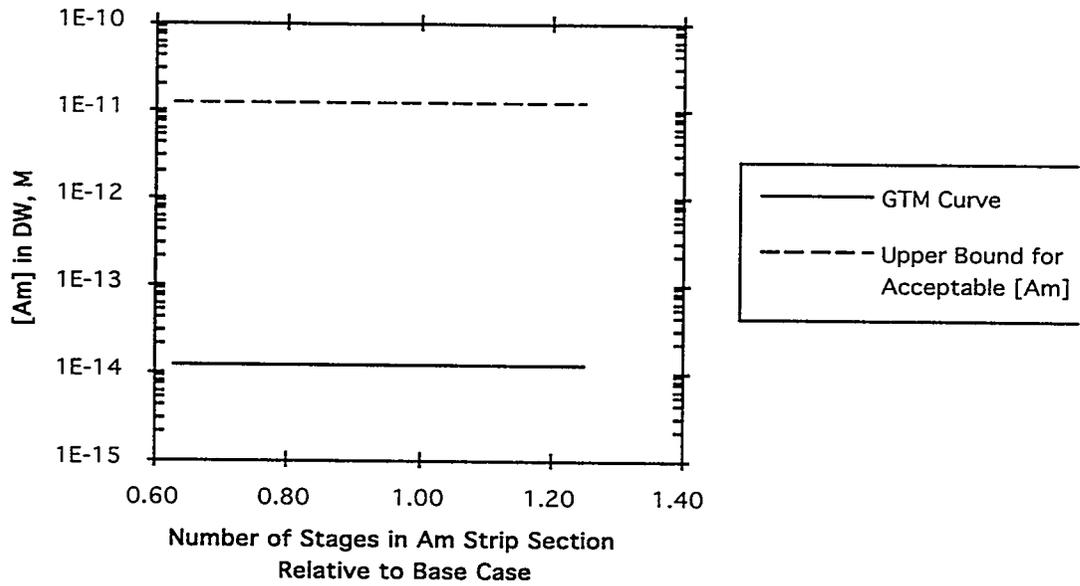


Fig. III-31. Effect of Number of Stages in Am Strip Section on Am Concentration in DW Raffinate. Base-case value for number of stages in Am strip section is eight stages.

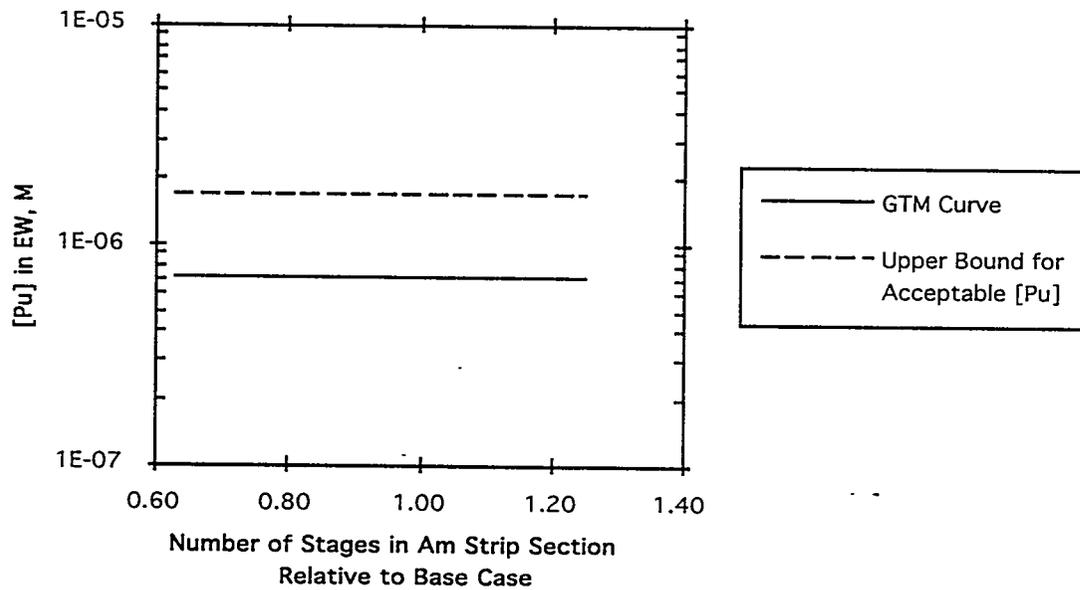


Fig. III-32. Effect of Number of Stages in Am Strip Section on Pu Concentration in EW Effluent. Base-case value for number of stages in Am strip section is eight stages.

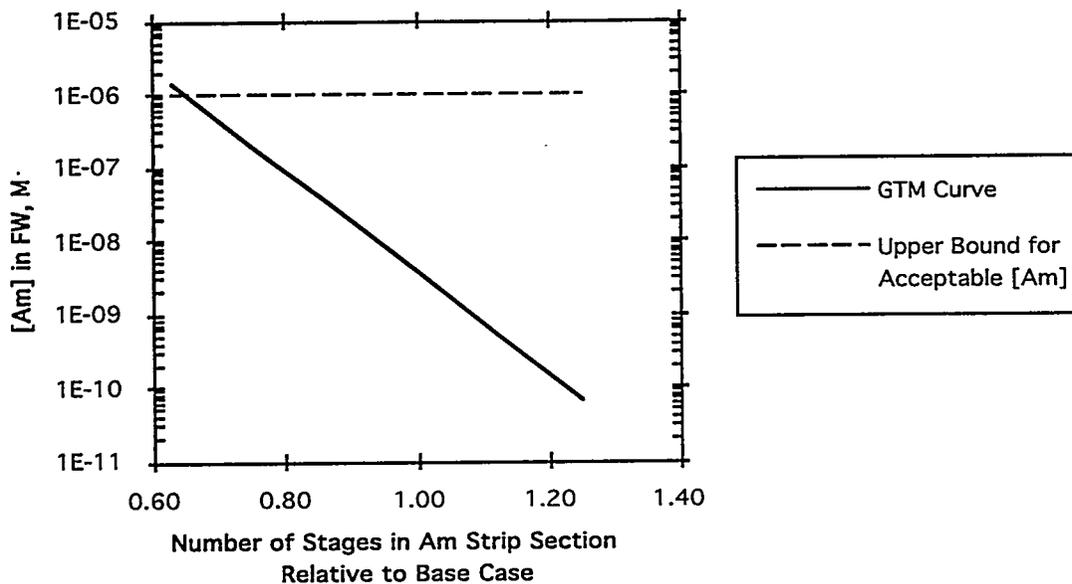


Fig. III-33. Effect of Number of Stages in Am Strip Section on Am Concentration in FW Effluent. Base-case value for number of stages in Am strip section is eight stages.

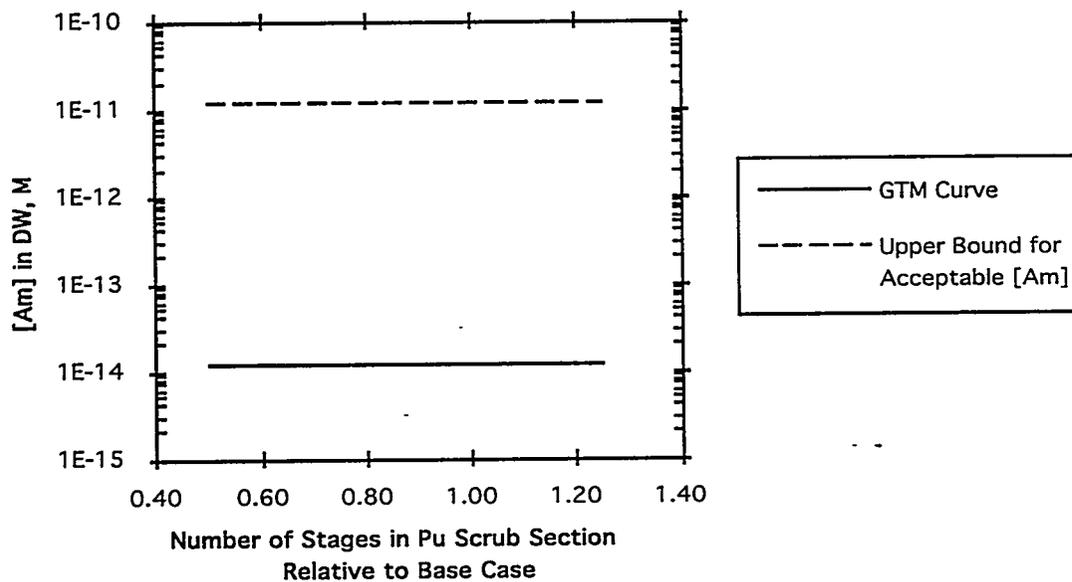


Fig. III-34. Effect of Number of Stages in Pu Scrub Section on Am Concentration in DW Raffinate. Base-case value for number of stages in Pu scrub section is four stages.

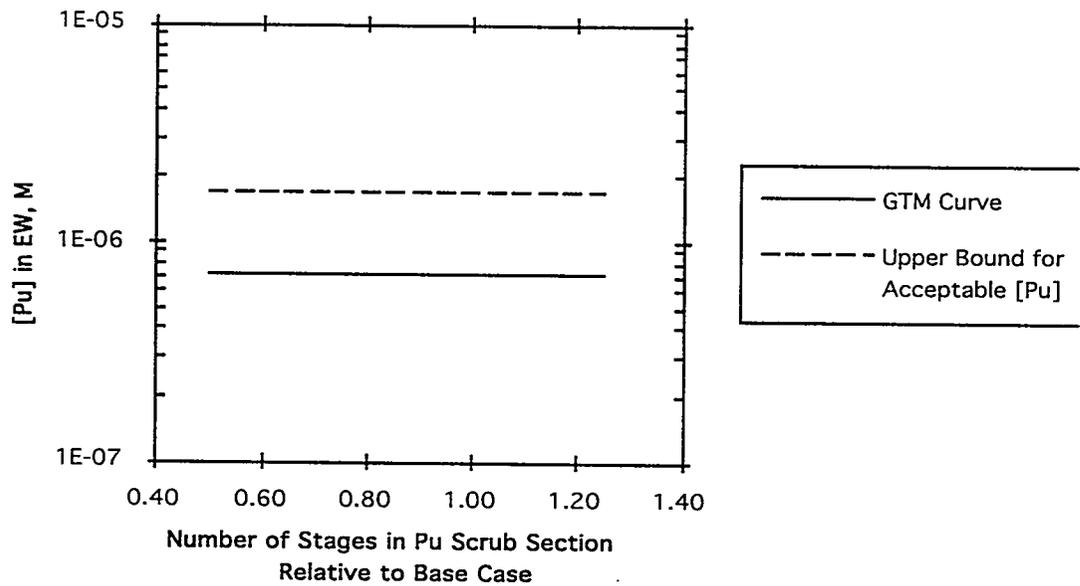


Fig. III-35. Effect of Number of Stages in Pu Scrub Section on Pu Concentration in EW Effluent. Base-case value for number of stages in Pu scrub section is four stages.

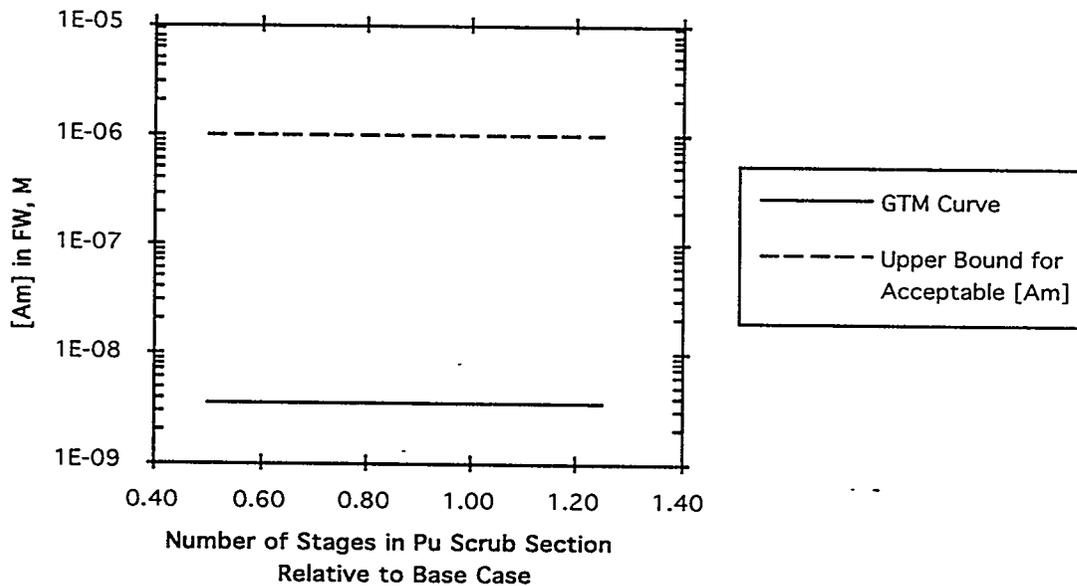


Fig. III-36. Effect of Number of Stages in Pu Scrub Section on Am Concentration in FW Effluent. Base-case value for number of stages in Pu scrub section is four stages.

IV. ALTERNATIVE TRUEX FLOWSHEET

In this section, we present an alternative TRUEX flowsheet that concentrates the Am in the Am strip (EW) effluent. This flowsheet is like the base case except that (1) it requires seven more stages and (2) the rate at which liquid is withdrawn from the Am strip effluent is greatly reduced. The rest of the aqueous phase exiting the Am strip section passes on to the scrub section, where it becomes the major scrub feed. Since the Am is pinched by the extraction section at one end and the strip section at the other end, the Am is concentrated in the EW effluent.

As with the base case, this alternative TRUEX flowsheet was developed by using the GTM.

A. Design Criteria

The design criteria for this alternative TRUEX flowsheet for RFP waste are essentially the same as those for the base case. The only difference is that the EW flow is now only 2% of the DF flow, so Am is concentrated in the EW effluent by a factor of 50.

B. Flowsheet Design

By using the design criteria for the alternative flowsheet, a second TRUEX process was developed for processing of TRU wastes at RFP. This flowsheet, shown in Fig. IV-1 and summarized in Table IV-1, achieves all of the design criteria using a 40-stage centrifugal contactor. Additional GTM reports and charts for the flowsheet are given in Appendix B. The designs for the various processing sections are the same as those for the base-case flowsheet, with three exceptions. First, two more extraction stages are required to hold back the higher concentrations of Am in the scrub section so that the same low level of Am is achieved in the DW raffinate. Second, a low-flow concentrated $\text{Al}(\text{NO}_3)_3$ stream is added to the aqueous feed going from the Am strip section to the scrub section so that the aqueous phase in the scrub section has an Al concentration sufficient to keep the F^- out of the Am strip. Third, the number of stages in the Am strip section is increased from 8 to 14 so that greater than 99.9% of the Am will be recovered there, even though the [Am] in the scrub section is higher and the stripping factor in the Am strip section is lower because the aqueous flow rate there has been reduced from 150 to 104 L/h.

Table IV-1. Summary of Feed and Effluent Concentrations for the Alternative Flowsheet Created by Using the GTM

| Section Name | Stage No. | Phase ^a | Flow Direction | Stream Identity ^b | Flow Rate, L/h ^c | Component Concentration, M | | | | | |
|--------------|-----------|--------------------|----------------|------------------------------|-----------------------------|----------------------------|------------------|------------------|-------------------------------|------------------|---|
| | | | | | | H ⁺ | Pu ⁴⁺ | Am ³⁺ | UO ₂ ²⁺ | Al ³⁺ | C ₂ O ₄ ²⁻ |
| Extraction | 1 | O | In | DX | 150 | 1E-15 | 1E-15 | 1E-15 | 1E-15 | 1E-15 | 1E-15 |
| Extraction | 12 | A | In | DF | 200 | 7.5 | 1.3E-04 | 2.1E-04 | 8.4E-06 | 0 | 0 |
| Scrub | 17 | A | In | DS | 3 | 0 | 0 | 0 | 0 | 1.5 | 0 |
| Am Strip | 31 | A | In | EF | 104 | 0.04 | 0 | 0 | 0 | 0 | 0 |
| Pu Strip | 32 | A | In | FF | 10 | 1.0 | 0 | 0 | 0 | 0 | 0.5 |
| Pu Scrub | 36 | A | In | FS | 140 | 9.0E-03 | 0 | 0 | 0 | 0 | 4.0E-03 |
| Extraction | 1 | A | Out | DW | 302.2 | 5.0 | <1E-16 | 1.63E-16 | <1E-16 | 0.0149 | 5.0E-16 |
| Am Strip | 18 | A | Out | EW | 4 | 0.24 | 8.1E-07 | 0.0105 | 1.49E-07 | 3.8E-04 | <1E-16 |
| Pu Strip | 32 | A | Out | FW | 150.8 | 0.0660 | 1.72E-04 | 1.36E-07 | 1.11E-05 | <1E-16 | 0.0314 |
| Pu Scrub | 36 | O | Out | FP ^d | 150 | 0.01119 | 3.0E-16 | <1E-16 | <1E-16 | <1E-16 | 5.46E-03 |

^aO = organic; A = aqueous.

^bOnly streams calculated by GTM are included here.

^cThese GTM flows differ from Fig. IV-1 flows in that they include the effect of 0.5% other-phase carryover in all interstage flows.

^dThis stream is not marked in Fig. IV-1 as it is neither a feed nor an effluent. It is the interstage organic-phase flow going from the plutonium scrub section to the first carbonate wash section.

C. Sensitivity to Process Variations

This alternative flowsheet has not been optimized, that is, developed to achieve the maximum robustness for a given number of stages. For this reason, no sensitivity analysis was applied to the alternative flowsheet design.

V. SOLVENT LOSSES AND REQUIRED VOLUME

Solvent losses can be accounted for by three primary mechanisms: (1) hydrolysis and radiolysis of the solvent components, (2) solubility of solvent components in the aqueous effluents, and (3) entrainment of the solvent in aqueous effluents. These three mechanisms are treated in separate sections below. In a fourth section, the solvent inventory required is discussed in terms of how solvent losses affect its makeup. Methods for recovery of solvent lost to aqueous raffinates are also discussed.

A. Radiolysis and Hydrolysis of the TRUEX Solvent

Radiolysis of the TRUEX solvent [SIMONZADEH] is caused by alpha and gamma decay of ^{241}Am , ^{239}Pu , and ^{240}Pu . For calculating the absorbed dose to the solvent, all plutonium was considered to be ^{239}Pu . Table V-1 shows the input data, the intermediate results, and the total dose to the TRUEX solvent when it is used to treat the RFP waste for 3600 h. The absorbed dose rate (0.0057 W/L) is a small fraction of that expected for treating high-level waste (0.01 to 10 W/L depending on how long it has been out of the reactor). Therefore, radiolysis of the solvent is of minor concern.

The GTM (option 9) was used to calculate the loss of CMPO that would occur due to a combination of radiolysis and hydrolysis if the base-case flowsheet were used at RFP. Table V-2 shows the predicted destruction rate of CMPO as a function of the ambient temperature of the processing facility. These results are plotted in Fig. V-1.

As well as calculating the rate at which CMPO is destroyed, the GTM estimates the rate at which the solvent quality is degraded. Figures V-2 and V-3 show how solvent damage by radiolysis and hydrolysis will affect the D value for Am as a function of dilute nitric acid concentration (0.01M and 0.05M), processing temperature, and number of cycles that the solvent passes through the process with no solvent-wash treatment. The increase in D_{Am} with the number of cycles at low HNO_3 concentrations is not good since Am becomes harder to remove from the solvent. These results can be used to estimate the number of cycles the solvent can be used without solvent washing or the fraction of the solvent stream that must be continually treated by solvent wash. At 25°C, treatment after every 100 cycles or treating 1% of the solvent stream continually should allow the solvent to strip Am. However, we recommend the continuous use of a solvent wash section as a precaution so that essentially all actinides are removed from the solvent before it is recycled.

Table V-1. Calculation of Dose Received by TRUEX Solvent after Processing RFP Nitric Acid Waste Stream for 3600 h

| TRU Element | Concn., M | Conv. factor, (nCi/mL)/M | Concn., nCi/mL | Concn., dps/L | Alpha Energy, eV | Power, (eV/s)/L | Absorbed Dose Rate, W/L | Dose, W•h/L ^a |
|-------------|-----------|--------------------------|----------------|---------------|------------------|-----------------|-------------------------|--------------------------|
| Pu | 1.3E-04 | 1.5E+07 | 1.95E+03 | 7.22E+07 | 5.16E+06 | 3.72E+14 | 5.96E-05 | 2.15E-01 |
| Am | 2.1E-04 | 8.3E+08 | 1.74E+05 | 6.45E+09 | 5.49E+06 | 3.54E+16 | 5.67E-03 | 2.04E+01 |
| Total | | | 1.76E+05 | 6.52E+09 | | 3.58E+16 | 5.73E-03 | 2.06E+01 |

^aTotal dose after 3600 h of exposure.

Table V-2. CMPO Loss by Hydrolysis and Radiolysis Calculated by Using the GTM

| Temperature, °C | Fraction of CMPO Lost per Cycle ^a | CMPO Loss ^b | |
|-----------------|--|------------------------|-------|
| | | kg/y | lb/y |
| 20 | 1.22E-06 | 0.024 | 0.052 |
| 25 | 2.15E-06 | 0.042 | 0.093 |
| 30 | 4.08E-06 | 0.080 | 0.176 |
| 35 | 8.01E-06 | 0.156 | 0.34 |
| 40 | 1.59E-05 | 0.31 | 0.68 |
| 45 | 3.12E-05 | 0.61 | 1.34 |
| 50 | 6.04E-05 | 1.18 | 2.6 |

^a Based on 0.17 h in the contactor per cycle.

^b The GTM was used to calculate the solvent damage due to hydrolysis and radiolysis. In these calculations, the dose to the solvent due to radioactive decay of Pu and Am was taken to be 0.0057 W/L. The TRUEX process is assumed to have 30 stages: 10 extraction stages with an average $[\text{HNO}_3]_{\text{aq}}$ of 6M, 10 scrub stages with an average $[\text{HNO}_3]_{\text{aq}}$ of 1.5M, and 10 strip stages with an average $[\text{HNO}_3]_{\text{aq}}$ of 0.03M. The solvent flow rate is assumed to be 150 L/h for 1600 h of operation each year.

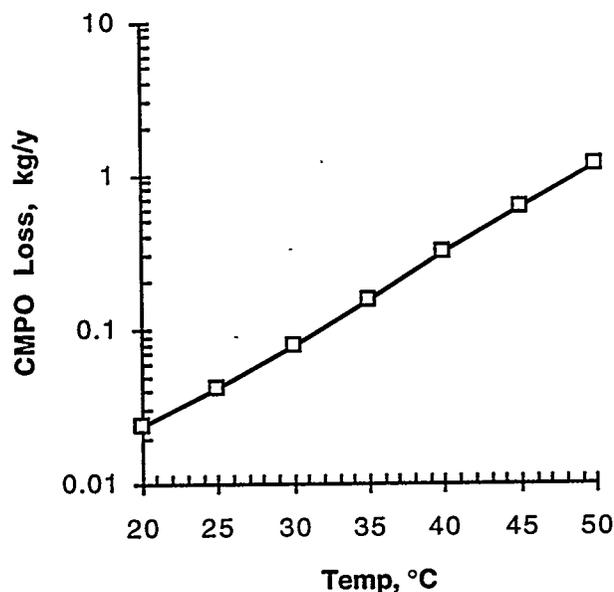


Fig. V-1. Calculated CMPO Loss due to Hydrolysis and Radiolysis as a Function of Temperature. See Table V-2 for assumptions.

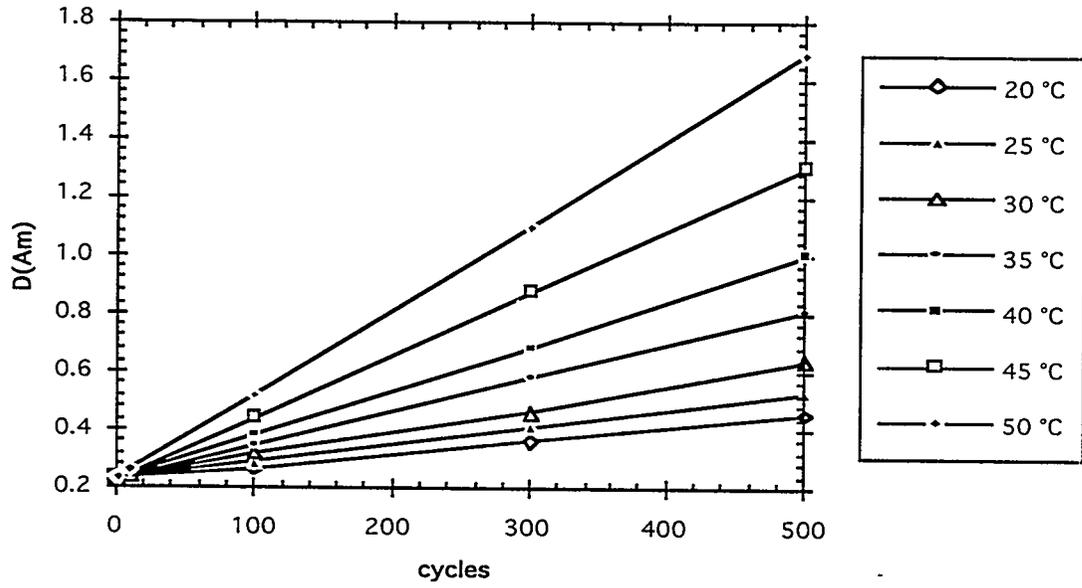


Fig. V-2. Effect of Process Temperature and Number of Cycles on the D Value for Am with No Solvent Cleanup When $[HNO_3] = 0.05M$

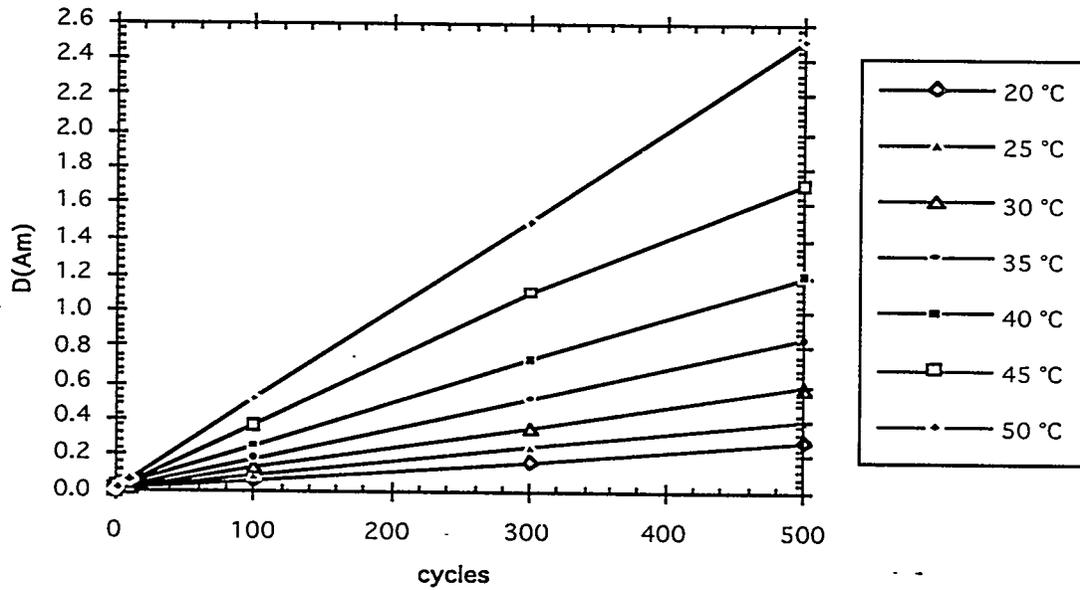


Fig. V-3. Effect of Process Temperature and Number of Cycles on the D Value for Am with No Solvent Cleanup When $[HNO_3] = 0.01M$

B. Solubility Losses

An earlier publication [VANDEGRIFT-1984A] addressed the loss of solvent components by their partitioning to the aqueous solutions during contact with the TRUEX solvent. That reference gives D values of 6.7×10^3 for CMPO and 1.6×10^3 for TBP in a solvent of 0.2M CMPO and 1.2M TBP in Norpar 12* and 1M HNO₃. In our calculations here, we have used D values of 7×10^3 for CMPO and 2×10^3 for TBP in a solution of 0.2M CMPO and 1.4M TBP in dodecane. The partitioning for dodecane** was estimated from data in Vandegrift [VANDEGRIFT-1984B]. The solubilities (in water) for hexane, heptane, and octane are 0.1 g/L, 0.03 g/L, and 0.01 g/L, respectively; the decrease suggests that the solubility of normal paraffinic hydrocarbons decreases by a factor of three each time the carbon length is increased by one. By extrapolating from octane to dodecane, we estimate that the solubility of dodecane is lower by a factor of $\sim 3^4$ (~ 100) than that of octane, giving dodecane an aqueous-phase solubility of 1×10^{-4} g/L (5×10^{-7} M). Table V-3 shows the expected partitioning losses of CMPO, TBP, and dodecane in the effluent streams for the base-case flowsheet. Over a year, 26 lb of CMPO, 394 lb of TBP, and 0.2 lb of dodecane will be lost to the aqueous effluents due to the solubility of the organics in these streams.

Table V-3. CMPO/TBP/Dodecane Loss in Base-Case Flowsheet Resulting from Solubility of Organics in the Aqueous Phase^a

| Section | No. of Stages | Feed Flow Rate, L/h ^f | Effluent Flow Rate, L/h | CMPO Loss ^{b,c} | | | TBP Loss ^{b,d} | | | Dodecane Loss ^{b,e} | | |
|--------------|---------------|----------------------------------|-------------------------|--------------------------|--------------|-----------|-------------------------|-------------|------------|------------------------------|----------------|------------|
| | | | | mol/h | kg/h | lb/y | mol/h | kg/h | lb/y | mol/h | kg/h | lb/y |
| Extraction | 10 | 200 | 300 | 0.009 | 0.004 | 12.91 | 0.21 | 0.06 | 196.86 | 0.00015 | 0.00003 | 0.09 |
| Scrub | 5 | 100 | 0 | 0 | | | | | | | | |
| Am Strip | 8 | 150 | 150 | 0.0045 | 0.002 | 6.45 | 0.105 | 0.03 | 98.43 | 7.5E-05 | 0.00001 | 0.04 |
| Pu Strip | 1 | 10 | 150 | 0.0045 | 0.002 | 6.45 | 0.105 | 0.03 | 98.43 | 7.5E-05 | 0.00001 | 0.04 |
| Pu Scrub | 4 | 140 | 0 | 0 | | | | | | | | |
| Carb. Wash 1 | 1 | 150 | recycle | | | 0.02 | | | 0.70 | | | 0.01 |
| Carb. Wash 2 | 1 | 150 | recycle | | | 0 | | | 0 | | | 0 |
| Carb. Wash 3 | 1 | 150 | recycle | | | 0 | | | 0 | | | 0 |
| Acid Rinse | 1 | 150 | recycle | | | 0 | | | 0 | | | 0 |
| TOTAL | 32 | | | 0.018 | 0.007 | 26 | 0.42 | 0.11 | 394 | 0.0003 | 0.00005 | 0.2 |

^a Assumptions are that $D_{\text{CMPO}} = 7 \times 10^3$ and $D_{\text{TBP}} = 2 \times 10^3$, on the basis of Vandegrift et al. [VANDEGRIFT-1984A], and that dodecane partitioning to aqueous phases is equal to the approximate solubility of dodecane in water, $\sim 1 \times 10^{-4}$ g/L or 5×10^{-6} M. Based on these assumptions, the aqueous phase will have [CMPO] = 3×10^{-5} M, [TBP] = 7×10^{-4} M, and [dodecane] = 5×10^{-7} M.

^b Based on processing time of 1600 h/y. Recycled streams such as the carbonate wash are assumed to have a holdup volume of 120 L. Based on the amount of acid (nitric and oxalic) in the solvent entering the first carbonate wash, the recycled carbonate wash solution would be half-neutralized in about one year, and should be changed at that time. Otherwise, the recycled carbonate wash feeds would need to be changed in the event of a process upset.

^c MW for CMPO is 407.6 g/mol.

^d MW for TBP is 266 g/mol.

^e MW for dodecane is 170 g/mol.

^f Organic flow rate is 150 L/h.

* A commercial normal aliphatic hydrocarbon mixture with an average carbon chain of 11.4.

** In this analysis, we use dodecane, one of the principal components of typical diluents (e.g., NPH and Norpar 12) to estimate diluent loss.

C. Entrainment Losses

When the centrifugal contactor is operating in its prescribed throughput range and the dispersion number for the contacting liquids is greater than or equal to the value for which the contactor was designed, the other-phase carryover (entrainment) can be limited to 0.04 volume %. That level of entrainment was used in calculating the entrainment losses presented in Table V-4. For CMPO (69 lb/y) and dodecane (313 lb/y), entrainment is a more serious factor than solubility in process losses; for TBP (315 lb/y), the solubility and entrainment losses are similar. These calculations show that entrainment is a significant factor in solvent loss and that process design criteria should include specifications on the extent of other-phase carryover.

Table V-4. CMPO/TBP/Dodecane Loss in Base-Case Flowsheet Resulting from Entrainment of Organics in the Aqueous Phase^a

| Section | No. of Stages | Feed Flow Rate, L/h ^b | Effluent Flow Rate, L/h | Solvent Loss, L/h ^c | CMPO Loss | | TBP Loss | | Dodecane Loss | |
|--------------|---------------|----------------------------------|-------------------------|--------------------------------|-----------|-------------------|----------|-------------------|---------------|-------------------|
| | | | | | kg/h | lb/y ^d | kg/h | lb/y ^d | kg/h | lb/y ^d |
| Extraction | 10 | 200 | 300 | 0.12 | 0.010 | 34 | 0.045 | 157 | 0.044 | 156 |
| Scrub | 5 | 100 | 0 | 0 | 0.000 | 0 | 0.000 | 0 | 0.000 | 0 |
| Am Strip | 8 | 150 | 150 | 0.06 | 0.005 | 17 | 0.022 | 79 | 0.022 | 78 |
| Pu Strip | 1 | 10 | 150 | 0.06 | 0.005 | 17 | 0.022 | 79 | 0.022 | 78 |
| Pu Scrub | 4 | 140 | 0 | 0 | 0.000 | 0 | 0.000 | 0 | 0.000 | 0 |
| Carb. Wash 1 | 1 | 150 | recycle | 0 | 0.000 | 0 | 0.000 | 0 | 0.000 | 0 |
| Carb. Wash 2 | 1 | 150 | recycle | 0 | 0.000 | 0 | 0.000 | 0 | 0.000 | 0 |
| Carb. Wash 3 | 1 | 150 | recycle | 0 | 0.000 | 0 | 0.000 | 0 | 0.000 | 0 |
| Acid Rinse | 1 | 150 | recycle | 0 | 0.000 | 0 | 0.000 | 0 | 0.000 | 0 |
| TOTAL | 32 | | | 0.24 | 0.020 | 69 | 0.089 | 315 | 0.089 | 313 |

^aThe TRUEX solvent is 0.2M CMPO and 1.4M TBP diluted by dodecane. This is equivalent to 81.4 g/L CMPO, 372 g/L TBP, and 370 g/L dodecane.

^bOrganic flow rate = 150 L/h.

^cBased on an aqueous entrainment of 0.04% in effluents. It is assumed that solvent entrained in recycled streams would be essentially zero, because the entrained solvent is returned to the process.

^dBased on processing time of 1600 h/y (200 production days per year).

D. Solvent Inventory and Makeup

Table V-5 summarizes the yearly losses of CMPO, TBP, and dodecane by hydrolysis/radiolysis, solubility, and entrainment. Table V-6 shows what these losses would mean for four possible solvent inventories. The minimum solvent inventory requirement would be twice the holdup in the contactor, lines, and pumps. For a flow rate for the TRU waste stream feed of 200 L/h, this holdup is estimated to be 50 L. A more conservative approach would be to have two separate tanks, each with two solvent holdup volumes. In that case, the backup supply would be available if a process upset caused loss, contamination, or chemical alteration of the solvent being used for processing. Increasing the solvent inventory to even larger volumes (4 hours or 8 hours) would reduce the rate at which the solvent composition changes with time and thereby lengthen the replacement interval.

Table V-5. CMPO/TBP/Dodecane Loss in Base-Case Flowsheet Resulting from Hydrolysis, Solubility, and Entrainment^a

| Reason for Loss | CMPO Loss | | TBP Loss | | Dodecane Loss | |
|------------------------------------|-----------|-----------|------------|------------|---------------|------------|
| | lb/y | kg/y | lb/y | kg/y | lb/y | kg/y |
| Hydrolysis/Radiolysis ^b | 0.09 | 0.04 | 1 | 0.5 | 0 | 0 |
| Solubility | 26 | 11.8 | 394 | 179 | 0.2 | 0.1 |
| Entrainment | 69 | 31 | 315 | 143 | 313 | 142 |
| TOTAL^c | 95 | 43 | 710 | 323 | 313 | 142 |

^aAssumes operation for 1600 h/y.

^bCMPO loss calculated by the GTM (option 9) for base-case flowsheet at 25°C. TBP loss is estimated to be about twice that of CMPO on a molar basis. Damage to dodecane would be nil.

^cThe loss of organic components would be divided among the various effluent streams.

Table V-6. Solvent Inventory and Replacement Rate

| Inventory Basis | Time to Recycle, h | Solvent Inventory ^a | | | | | | Solvent Loss Rate, L/h | Replacement Interval, h ^b |
|--|--------------------|--------------------------------|------|----|------|-----|-------------|------------------------|--------------------------------------|
| | | Liters | CMPO | | TBP | | Dodecane, L | | |
| | | | lb | kg | lb | kg | | | |
| Twice Holdup Volume | 0.67 | 100 | 18 | 8 | 126 | 57 | 53 | 0.24 | 417 |
| Two Tanks, Each with Twice Holdup Volume | 1.33 | 200 | 36 | 16 | 252 | 114 | 107 | 0.24 | 833 |
| 4 Hours | 4 | 600 | 107 | 49 | 755 | 342 | 320 | 0.24 | 2,500 |
| 8 Hours | 8 | 1200 | 215 | 98 | 1510 | 684 | 640 | 0.24 | 5,000 |

^aTRUEX solvent is 0.2M CMPO and 1.4M TBP in dodecane.

^bTime taken to deplete the entire solvent inventory if entrainment is 0.04 volume %. In practice, smaller quantities of solvent would be added at much shorter intervals.

E. Removal of Solvent Components from Aqueous Effluents

Recovery of the organic constituents of the solvent is an important concern for two reasons. First, and certainly more important, is the safety and operational concern of having organic constituents in an aqueous stream that is going to be concentrated by evaporation. This concern is discussed in Section VI. The second concern is the value of these constituents. It is likely that CMPO will cost close to \$1000/lb at the quantities needed at RFP. At that price, the total annual losses for CMPO, TBP, and dodecane shown in Table V-5 represent a significant cost, especially the CMPO. Table V-7 shows the concentrations of these constituents in the effluent streams. Because these concentrations are likely to be unacceptable in feeds to an evaporator (see Section VI.B), recovery of these organics, especially CMPO and TBP, should be investigated.

In an attractive approach, both entrained and soluble CMPO and TBP are removed and recovered by means of solvent extraction in centrifugal contactors with dodecane as the solvent. By using a four-stage unit in which dodecane and the aqueous effluent run countercurrently, CMPO and TBP could be effectively eliminated from the effluent and recovered in concentrated form for recycling.

Table V-7. Concentration of CMPO, TBP, and Dodecane in Aqueous Effluents for Base-Case Flowsheet

| Type of Concentration | CMPO, mg/L | TBP, mg/L | Dodecane, mg/L |
|-----------------------|------------|-----------|----------------|
| Soluble | 12 | 186 | 0.2 |
| Entrained | 33 | 149 | 148 |
| Total | 45 | 335 | 148 |

VI. SAFETY INFORMATION

Although TRUEX processing has never been run as a full-scale operation over extended periods of time, it has been studied at the laboratory and pilot-plant scale for almost eight years. Also, because the solvent is essentially a PUREX-process solvent (that is, TBP in a normal paraffinic hydrocarbon) and because with the exception of CMPO, the chemicals used in TRUEX processing (e.g., HNO_3 , HF, $\text{H}_2\text{C}_2\text{O}_4$) have been used in PUREX plants, the only unknown safety concern in opening and operating a new TRUEX facility would be CMPO. Material safety data sheets (MSDSs) for CMPO and the other chemicals can be obtained from their suppliers.

A. CMPO Safety Considerations

According to its primary commercial manufacturer, ATOCHEM North America, Philadelphia, Pennsylvania, CMPO can be stored in normal warehouse storage in a cool, dry area. Local exhaust is recommended for its use; mechanical (general) exhaust is required. In handling the material, goggles and butyl or nitrile gloves are recommended. Special precautions are a warning against eye contact ("may cause severe irritation to eyes") and, to a lesser extent, skin contact ("may cause mild irritation to skin upon contact"). The flash point for CMPO is considered to be high (above that of TBP— 146°C [295°F]) because of its low volatility and high molecular weight [VANDEGRIFT-1984A]. It should also be noted that compounds with structures similar to CMPO have been used as flame-retardant finishings for pile carpeting [HOECHST].

B. Safety Considerations for Feeds to the Evaporator for Nitric Acid Recycle

Heating concentrated nitric acid in the presence of organic compounds is always a concern because of the potential for violent oxidation/nitration reactions. Shoun and Thompson [SHOUN] cite a review paper by Burger [BURGER] as the authoritative source on this topic. Oxidation of TBP by nitric acid or nitric-acid/nitrous-acid mixtures does not occur at temperatures below 70°C ; TBP is oxidized at higher temperatures.

A recent publication shows that, except for one terrible accident, evaporation of PUREX effluents has been nearly accident-free [DURANT]. That one incident, a "red oil" explosion that occurred January 12, 1953, is brought up whenever evaporation of PUREX effluents is discussed. The incident occurred during evaporation of a uranyl nitrate solution to remove about 50% of the nitric acid. The incident was presumed to have resulted from the following series of events:

- Presence of about 80 lb of TBP in the 1800 gal of aqueous uranyl nitrate solution,
- Concentration of the solution to >78 wt % uranyl nitrate,
- A solution temperature $>130^\circ\text{C}$, and
- Buildup of a 50-100 psi (345-690 kPa) back pressure resulting from partial plugging of plates in the evaporator.

This incident and the investigation that followed led to the establishment of operating limits on organic concentrations and on temperature for evaporators at the Savannah River Site, Aiken, South Carolina. In an incident 22 years later, an explosion and fire occurred during calcining of uranyl nitrate, which was caused by the inadvertent addition of the uranyl-nitrate/TBP adduct to the denitration step [GRAY].

Shoun and Thompson [SHOUN] have summarized the findings of several investigators of "red oil" [COLVEN, EAKIN, NICHOLS, NOWAK, PUGH, SEGE, SHEFCIK, WAGNER, WILBOURN-1977A, -1977B] in the following statements:

- "Red oil" is a complex, dense (1.1-1.6 g/mL) mixture of organic compounds formed as a result of hydrolytic and other reactions (including nitration) between certain TBP-diluent mixtures and aqueous metal nitrate solutions.
- "Red oil" decomposes in the range of 130-150°C.
- The presence of organic-phase complexes of $\text{UO}_2(\text{NO}_3)_2$, $\text{Pu}(\text{NO}_3)_4$, or $\text{Th}(\text{NO}_3)_4$ is essential to its formation.
- Explosive decompositions and accompanying fires in several plant-scale evaporator incidents [COLVEN, DURANT, PUGH, SEGE, WAGNER] involved decomposition of $\text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP}$ rather than ignition of "red oil."
- The presence of "red oil" during evaporation or denitration of these metal-nitrate/TBP complexes complicates control of these operations because "red oil" decomposes at a lower temperature than the complexes and can trigger their decomposition. Control of these processes is vital to their safe operation.
- Current diluents such as NPH being used in fuel reprocessing are more resistant to the nitration and better suited for concentrator operations from a safety standpoint.
- Bench-scale experiments show that red oil is not formed when TBP/NPH is used as a solvent.

These findings lead to the conclusions that (1) under properly controlled conditions of feed composition and evaporator temperature, evaporation of the nonTRU and other effluent streams can be safely accomplished, and (2) removal of organics from this feed, especially CMPO and TBP, would increase evaporator operation and safety.

VII. INTERACTION OF TRUEX PROCESS WITH NITRIC ACID RECYCLE

In the implementation of the TRUEX process, the aqueous (DW) raffinate would be fed to an evaporator that concentrates the raffinate and recovers HNO_3 for recycle to the plant. This raffinate stream would contain all the elements that are in the aqueous (DF) feed except for the Am, Pu, and U. The concentration of these three elements would be very low (<0.01 nCi/mL). The concentrations of all the other elements would be somewhat less than their concentrations in the DF feed because of dilution by the scrub (DS) feed. The DW raffinate would also contain the aluminum that is added in the DS feed. None of the anions or cations in the DF feed are expected to be a problem in the evaporator if appropriate construction materials are used. In the past, entrained solvent has caused problems; however, as discussed in Section V.E, entrained CMPO and TBP can be removed and recovered by using a dodecane strip in centrifugal contactors. Without such a recovery system, the concentrations of the various organic constituents in the DW raffinate would have the concentrations shown in Section V.

VIII. FEED ANALYSIS REQUIREMENTS

For both the base-case and alternative TRUEX processes, the flowsheets are designed to handle the range of aqueous (DF) feed compositions given in Table III-1. Thus, no feed analyses should be required. If one feed analysis is to be done, it should be to contact a small quantity of the feed with a small quantity of TRUEX solvent to check that the TRU elements are extracted by the solvent in the expected amounts.

If the alternative TRUEX process is used, a radiation-counting device should be built into the process to monitor the [Am] in the Am (EW) product stream. The effluent pump that draws off the EW stream should be controlled by this monitor so that the [Am] in the EW stream is held essentially constant. In this way, even very low levels of Am in the DF feed would be concentrated to about 0.01M Am, and the total volume of the EW stream would be reduced accordingly.

IX. OTHER PROCESS CONSIDERATIONS

A. Ammonium Oxalate Strip

Recent work at Argonne National Laboratory has shown ammonium oxalate solutions to be an efficient and effective strip for removing Pu and Am from the TRUEX solvent. We began to investigate alternative stripping agents during our treatment of New Brunswick Laboratory Pu waste. Because the concentration of Pu in this waste is about 1 g/L, Pu will precipitate as it is stripped from the solvent by HF, H₂C₂O₄, or HEDPA. In our treatment of this waste, we used oxalic acid as the stripping agent, and, as predicted, Pu(C₂O₄)₂ precipitated in the contactor. On the basis of the extremely limited solubility of lanthanide oxalates, Am precipitation would be a much more severe problem.

1. Benefits of an Ammonium Oxalate Strip

Ammonium oxalate was chosen for two reasons. First, the oxalate-complexation equilibria for Pu(IV) predict increased solubility of Pu in oxalate solution at high oxalate ion concentrations. The tris-oxalato complex, Pu(C₂O₄)₃²⁻, will be the predominant species when the concentration of the oxalate ion is above 10⁻⁶M [KATZ], and the solubility of Pu in oxalate solution begins to increase appreciably as the oxalate ion concentration increases above 10⁻⁴M. Because of the complex acid equilibria of oxalic acid, it is impossible to prepare a solution of oxalic acid with an oxalate ion concentration above ~3 x 10⁻⁴M. However, using an oxalate salt provides an oxalate ion concentration limited only by the solubility of that salt. The second reason for using ammonium oxalate is its instability on heating to form ammonia and oxalic acid, both of which are volatile. After the Pu is stripped, the solution can be evaporated to dryness and calcined to prepare pure, salt-free PuO₂.

Experiments at Argonne using a 0.28M solution of ammonium oxalate have shown the solubility of Pu(IV) to be greater than or equal to 2 g/L (10⁻²M) and the Pu distribution ratios for the TRUEX solvent to be less than 10⁻⁴. The solubility of Am in that same solution was ~0.3 g/L (10⁻³M), and its distribution ratio was less than 10⁻³. By using this stripping agent, it should be possible to design a TRUEX flowsheet with only one strip for all TRU elements. Data must still be collected to find an upper limit to Pu solubility and to measure the solubility of UO₂²⁺ in ammonium oxalate.

Another very important research area is the safety aspects of using this reagent (and the ammonium bioxalate and nitrate formed by its reaction with nitric acid) in the solvent extraction process and the concentration and calcining steps. Safety concerns are discussed briefly below.

2. Safety Concerns

When ammonium oxalate is used, ammonium bioxalate and ammonium nitrate will be formed. This happens because the TRUEX solvent that contacts the strip solution will contain nitric acid, nitrate, uranyl nitrate, and americium nitrate. All four compounds will be stripped into the aqueous strip solution. Ammonium oxalate will react with the hydrogen ion to form ammonium bioxalate and with nitrate to form ammonium nitrate.

The major safety concern is the potential for explosion of ammonium nitrate. Ammonium nitrate volatilizes reversibly at moderate temperatures (300°C) to form gaseous NH₃ and HNO₃. At higher temperatures, it decomposes irreversibly, forming mostly N₂O. At still higher temperatures, its decomposition gives mostly N₂ and O₂. Because it can detonate when initiated by another high explosive, it is used in some bombs [COTTON]. Clearly, before ammonium oxalate can be considered for use in this process, a great deal of work must be done to test its safety in all operations of the plant.

B. Contactors Size

The TRUEX flowsheets shown in Figs. III-1 and IV-1 have process flow rates listed that could be handled easily in a 10-cm centrifugal contactor. Since a 10-cm contactor can handle up to 600 L/h (both phases), the flow rates on the two figures could be increased by up to 33%, and the process could still be carried out in a multistage 10-cm contactor. Since this contactor has a total diameter, including the stationary housing, of 12.5 cm (5 in.) in the mixing and separating zone and of 15 cm (6 in.) at the collector rings, it should be safe by design with respect to nuclear criticality.

If nuclear criticality is not an issue, larger contactors can be designed and built to handle higher throughputs. If it is an issue, additional process lines can be added to accommodate higher throughputs.

C. Filtration

As one approaches the low concentrations of TRU elements in the aqueous, nonTRU (DW) raffinate given in the two flowsheets discussed here, the presence of particles in the aqueous and organic phases becomes important. These particles, which could contain TRU elements, will not be extracted by the TRUEX process. As such, they could seriously limit our ability to reach a nonTRU (DW) raffinate with less than 0.01 nCi/mL of TRU elements. For this reason, it is recommended that the TRU (DF) feed, the recycled solvent (DX) feed, and the nonTRU (DW) raffinate go through appropriate filters. At Argonne, we have found that a sequence of a 5-micron filter followed by a 1-micron filter works well for achieving 0.1 nCi/mL of actinides in the raffinate. The filters used were Whatman disposable filter capsules with polypropylene membranes. They are available from the Cole-Parmer Instrument Company (Niles, Illinois). To achieve a lower goal, finer filters may be required. Westinghouse Hanford has some pilot plant experience with two other filters (cross-flow and sand filters).

X. FUTURE WORK

1. Depending on process throughputs and the need for equipment that is criticality-safe by geometry, it may be worthwhile to develop a high-throughput, criticality-safe contactor. Current criticality-safe contactor designs can handle throughputs up to 600 L/h. A high-throughput unit should be able to handle flows from two to eight times higher.
2. If a concentrated Am product stream is desirable and if a 40-stage contactor can be accommodated, the alternative (high Am) flowsheet should be optimized so that it is not sensitive to process changes.
3. Further work should be done to develop and test a correlation for $\text{Pu}(\text{C}_2\text{O}_4)_2$ solubility at the low concentrations of oxalic and nitric acid that will be present in the Pu strip section.
4. If the use of ammonium oxalate would be acceptable, it should be considered as an alternative to the use of oxalic acid in the Pu strip section. $\text{Pu}(\text{C}_2\text{O}_4)_2$ appears to be considerably more soluble when $(\text{NH}_4)_2\text{C}_2\text{O}_4$ is used to strip the Pu from the solvent. This study should also be extended to $\text{Am}_2(\text{C}_2\text{O}_4)_3$.
5. The use of HEDPA should be considered for stripping residual Am and Pu from the solvent before it goes to the solvent cleanup section.
6. The base-case TRUEX flowsheet should be tested on a laboratory scale in a 32-stage centrifugal contactor.
7. Techniques for reducing solvent entrainment in the aqueous (DW) raffinate and for recovering and concentrating residual solvent in the DW raffinate should be further developed and tested.

REFERENCES

BURGER

L. L. Burger, *The Chemistry of Tributyl Phosphate: A Review*, U.S. Atomic Energy Commission Report HW-40910, General Electric Co., Richland, WA (1955).

COLVEN

T. J. Colven, Jr., G. M. Nichols, and T. H. Siddall, *Interim Technical Report, TNX Evaporator Incident, January 12, 1953*, U.S. Atomic Energy Commission Report DP-25, E. I. du Pont de Nemours & Co., Aiken, SC (1953).

COTTON

F. A. Cotton and G. Wilkinson, "Ammonium Salts," *Advanced Inorganic Chemistry—A Comprehensive Text*, 3rd ed., Wiley, New York, 349 (1972).

DURANT

W. S. Durant, D. K. Craig, M. J. Vitacco, and J. A. McCormick, "Adverse Experiences with Nitric Acid at the Savannah River Site," Westinghouse Savannah River Company Report WSRC-TR-91-22, Rev. 1, Aiken, SC (1991).

EAKIN

D. E. Eakin, "Hazardous Properties of Purex Plant Chemicals," U.S. Atomic Energy Commission Report ARH-1404, Atlantic Richfield Hanford Co., Richland, WA (1969).

GRAY

L. W. Gray, "An Explosion and Fire During Conversion of Liquid Uranyl Nitrate to Solid Uranium Oxide," *Nuclear Safety*, 19, 91 (1978).

HOECHST

A. G. Hoechst, *Flame-Retardant Finishing of Pile Carpeting*, Ger. Offern. 2,825,818, January 11, 1979; Swiss Appl. 777,300, June 14, 1977 [Chemical Abstracts Reference 90:1390355].

KATZ

J. J. Katz, G. T. Seaborg, and L. R. Morss, *The Chemistry of Actinide Elements*, 2nd ed., Vol. 1, Chapman and Hall, New York, 811 (1986).

LEONARD

R. A. Leonard et al., "SASSE Development," in J. E. Battles et al., *Nuclear Technology Programs Semiannual Progress Report, April-September 1991*, Argonne National Laboratory Report ANL-93/21 (1993).

LIDE

D. R. Lide, ed., *CRC Handbook of Chemistry and Physics*, 71st ed., CRC Press, Boca Raton, FL (1990).

NICHOLS

G. S. Nichols, *Decomposition of the Tributylphosphate-nitrate Complexes*, U.S. Atomic Energy Commission Report DP-526, E. I. du Pont de Nemours & Co., Aiken, SC (1960).

NOWAK

Z. Nowak and M. Nowak, "Thermal Degradation of TBP-diluent Systems," *Radiochem. Radioanal. Lett.*, 38, 377, 1979.

PUGH

R. A. Pugh, *Notes Pertaining to Recuplex Product Evaporation*, U.S. Atomic Energy Commission Report HW-32100, General Electric Co., Richland, WA (1954).

REGALBUTO

M. C. Regalbuto, B. Misra, D. B. Chamberlain, R. A. Leonard, and G. F. Vandegrift, *The Monitoring and Control of TRUOX Processes, Volume One: The Use of Sensitivity Analysis to Determine Key Process Variables and Their Control Bound*, Argonne National Laboratory Report ANL-92/7 (1992).

SEGE

G. Sege, *Overconcentration in Initial Operation of Uranium Evaporator-231 Building*, U.S. Atomic Energy Commission Report HW-28690, General Electric Co., Richland, WA (1953).

SHEFCIK

J. J. Shefcik, *Safety Aspects of Purex Plant Concentration Operation*, U.S. Atomic Energy Commission Report HW-40556, General Electric Co., Richland, WA (1955).

SHOUN

R. R. Shoun and M. C. Thompson, "Chemical Properties and Reactions," in *Science and Technology of Tributyl Phosphate*, Vol. I, W. W. Schulz and J. D. Navratil, eds., CRC Press, Inc., Boca Raton, FL, 152-153 (1984).

SIMONZADEH

N. Simonzadeh, A. M. Crabtree, L. E. Trevorow, and G. F. Vandegrift, "Radiolysis and Hydrolysis of TRUOX-NPH Solvent," Argonne National Laboratory Report ANL-90/14 (1992).

WAGNER

R. M. Wagner, *Investigation of Explosive Characteristics of Purex Solvent Decomposition Products (Red Oil)*, U.S. Atomic Energy Commission Report HW-27492, General Electric Co., Richland, WA (1953).

WICK

O. J. Wick, "Plutonium Oxalates," *Plutonium Handbook: A Guide to the Technology*, Vol. I, Gordon and Breach, New York, 370 (1967).

WILBOURN-1977A

R. G. Wilbourn, *Safety Aspects of Solvent Nitration in HTGR Fuel Reprocessing*, Energy Research and Development Administration Report GA-A14372 (1977).

WILBOURN-1977B

R. G. Wilbourn, "Safety Aspects of Solvent Nitration in HTGR Fuel Reprocessing," *Trans. Am. Nucl. Soc.*, 27, 291 (1977).

VANDEGRIFT-1984A

G. F. Vandegrift et al., *Transuranic Decontamination of Nitric Acid Solutions by the TRUEX Solvent Extraction Process: Preliminary Development Studies*, Argonne National Laboratory Report ANL-84-45 (1984).

VANDEGRIFT-1984B

G. F. Vandegrift, "Diluents for TBP Extraction Systems," in *Science and Technology of Tributyl Phosphate*, Vol. I, W. W. Schulz and J. D. Navratil, eds., CRC Press, Inc., Boca Raton, FL, p. 72 (1984).

VANDEGRIFT-1993

G. F. Vandegrift, D. B. Chamberlain, C. Conner, J. M. Copple, J. A. Dow, L. Everson, J. C. Hutter, R. A. Leonard, L. Nuñez, M. C. Regalbuto, J. Sedlet, B. Srinivasan, S. Weber, and D. G. Wygmans, "Development and Demonstration of the TRUEX Solvent Extraction Process," Proceedings of the Waste Management Symposium, February 28-March 4, 1993, Tucson, AZ (in press).

APPENDIX A

GTM Reports for the Base-Case TRUEX Flowsheet

The concentration profile report and the chart for each component were generated by the GTM version 2.4 for the base-case flowsheet shown in Fig. III-1. These items are included here as Figs. A-1 through A-7. The full summary report generated by the GTM is included in the main body of this report as Table III-2.

| Component Name | Phase | Section No. | Stage-to-Stage Profile for each Component | | | | | | | | | | | | | | | | | | | |
|----------------|--------------|-------------|---|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | | | | | | | | | | |
| H | Aqueous | x, M | 5.023E+00 | 5.791E+00 | 5.882E+00 | 5.894E+00 | 5.895E+00 | | |
| | Organic | y, M | 1.538E+00 | 1.715E+00 | 1.737E+00 | 1.739E+00 | 1.740E+00 | |
| | D(effective) | | 3.061E-01 | 2.862E-01 | 2.953E-01 | 2.951E-01 | |
| A1 | Aqueous | x, M | 9.935E-03 | 9.940E-03 | |
| | Organic | y, M | 9.935E-06 | 9.940E-06 | |
| | D(effective) | | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | |
| UC2 | Aqueous | x, M | 3.365E-19 | 2.664E-19 | 2.594E-19 | 2.623E-19 | 6.215E-19 | 3.497E-17 | 3.318E-15 | 3.171E-13 | 3.031E-11 | 2.931E-09 | 2.823E+03 | 2.715E-07 | 2.607E-05 | 2.500E-03 | 2.393E-01 | 2.286E-01 | 2.179E-01 | 2.072E-01 | 1.965E-01 | 1.858E-01 |
| | Organic | y, M | 9.999E-16 | 9.998E-16 | 1.000E-15 | 1.015E-15 | 2.405E-15 | 1.353E-13 | 1.284E-11 | 1.227E-09 | 1.173E-07 | 1.121E-05 | 1.070E-03 | 1.019E-01 | 9.68E-01 | 9.27E-01 | 8.86E-01 | 8.45E-01 | 8.04E-01 | 7.63E-01 | 7.22E-01 | 6.81E-01 |
| | D(effective) | | 2.971E+03 | 3.753E+03 | 3.856E+03 | 3.868E+03 | 3.870E+03 |
| Am | Aqueous | x, M | 1.217E-14 | 1.478E-13 | 1.489E-12 | 1.470E-11 | 1.449E-10 | 1.428E-09 | 1.406E-08 | 1.385E-07 | 1.365E-06 | 1.347E-05 | 1.329E-04 | 1.311E-03 | 1.293E-02 | 1.275E-01 | 1.257E-01 | 1.239E-01 | 1.221E-01 | 1.203E-01 | 1.185E-01 | 1.167E-01 |
| | Organic | y, M | 3.018E-13 | 3.245E-12 | 3.225E-11 | 3.180E-10 | 3.133E-09 | 3.087E-08 | 3.041E-07 | 2.995E-06 | 2.951E-05 | 2.859E-04 | 2.767E-03 | 2.675E-02 | 2.583E-01 | 2.491E-01 | 2.400E-01 | 2.308E-01 | 2.216E-01 | 2.124E-01 | 2.032E-01 | 1.940E-01 |
| | D(effective) | | 2.481E+01 | 2.196E+01 | 2.166E+01 | 2.163E+01 | 2.162E+01 |
| Pu_4 | Aqueous | x, M | 2.577E-19 | 2.564E-19 | 2.569E-19 | 3.145E-19 | 5.818E-18 | 5.321E-16 | 5.085E-14 | 4.862E-12 | 4.650E-10 | 4.498E-08 | 4.346E-06 | 4.194E-04 | 4.042E-02 | 3.890E-01 | 3.738E-01 | 3.586E-01 | 3.434E-01 | 3.282E-01 | 3.130E-01 | 2.978E-01 |
| | Organic | y, M | 1.000E-15 | 1.000E-15 | 1.002E-15 | 1.227E-15 | 2.270E-14 | 2.076E-12 | 1.984E-10 | 1.897E-08 | 1.814E-06 | 1.734E-04 | 1.654E-02 | 1.574E-01 | 1.494E-01 | 1.414E-01 | 1.334E-01 | 1.254E-01 | 1.174E-01 | 1.094E-01 | 1.014E-01 | 9.34E-02 |
| | D(effective) | | 3.881E+03 | 3.900E+03 | 3.902E+03 |
| C2O4 | Aqueous | x, M | 5.013E-16 | 4.323E-18 | 2.139E-20 | 1.005E-22 | 4.692E-25 | 2.189E-27 | 1.021E-29 | 4.765E-32 | 2.223E-34 | 1.037E-36 | 4.681E-39 | 2.135E-41 | 9.801E-44 | 4.446E-47 | 2.001E-50 | 9.006E-53 | 4.002E-56 | 1.801E-59 | 8.001E-62 | 3.601E-65 |
| | Organic | y, M | 6.041E-18 | 2.064E-20 | 9.151E-23 | 4.242E-25 | 1.977E-27 | 9.223E-30 | 4.303E-32 | 2.007E-34 | 9.364E-37 | 4.368E-39 | 2.007E-41 | 9.364E-44 | 4.368E-47 | 2.007E-50 | 9.364E-53 | 4.368E-56 | 2.007E-59 | 9.364E-62 | 4.368E-65 | 2.007E-68 |
| | D(effective) | | 1.205E-02 | 4.775E-03 | 4.278E-03 | 4.221E-03 | 4.214E-03 | 4.213E-03 |

Fig. A-1. Concentration Profile Report from the GTM for the Base-Case TRUOX Process (1 of 3).

| Component Name | Phase | Section No. | | | | | | | | | | | | | | | | | | | |
|----------------|--------------|-------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 | 2 |
| Stage No. | | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 |
| H | Aqueous | 2.689E+00 | 1.503E+00 | 9.520E-01 | 6.285E-01 | 4.081E-01 | 1.835E-01 | 7.361E-02 | 4.500E-02 | 4.064E-02 | 4.008E-02 | 2.689E+00 | 1.503E+00 | 9.520E-01 | 6.285E-01 | 4.081E-01 | 1.835E-01 | 7.361E-02 | 4.500E-02 | 4.064E-02 | 4.008E-02 |
| | Organic | 9.663E-01 | 6.056E-01 | 3.929E-01 | 2.478E-01 | 1.439E-01 | 3.499E-02 | 7.045E-03 | 2.847E-03 | 2.316E-03 | 2.249E-03 | 9.663E-01 | 6.056E-01 | 3.929E-01 | 2.478E-01 | 1.439E-01 | 3.499E-02 | 7.045E-03 | 2.847E-03 | 2.316E-03 | 2.249E-03 |
| | D(effective) | 3.593E-01 | 4.031E-01 | 4.127E-01 | 3.943E-01 | 3.525E-01 | 1.907E-01 | 1.907E-01 | 9.571E-02 | 6.328E-02 | 5.700E-02 | 5.610E-02 | 3.593E-01 | 4.031E-01 | 4.127E-01 | 3.943E-01 | 3.525E-01 | 1.907E-01 | 1.907E-01 | 9.571E-02 | 6.328E-02 |
| Al | Aqueous | 2.982E-02 | 3.000E-02 | 3.000E-02 | 3.000E-02 | 3.000E-02 | 1.808E-04 | 1.095E-06 | 6.627E-09 | 4.013E-11 | 2.430E-13 | 2.982E-02 | 3.000E-02 | 3.000E-02 | 3.000E-02 | 3.000E-02 | 1.808E-04 | 1.095E-06 | 6.627E-09 | 4.013E-11 | 2.430E-13 |
| | Organic | 2.982E-05 | 3.000E-05 | 3.000E-05 | 3.000E-05 | 3.000E-05 | 1.808E-07 | 1.095E-09 | 6.627E-12 | 4.013E-14 | 2.430E-16 | 2.982E-05 | 3.000E-05 | 3.000E-05 | 3.000E-05 | 3.000E-05 | 1.808E-07 | 1.095E-09 | 6.627E-12 | 4.013E-14 | 2.430E-16 |
| | D(effective) | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| UO2 | Aqueous | 8.793E-09 | 1.502E-08 | 1.980E-08 | 2.438E-08 | 3.134E-08 | 1.260E-07 | 5.896E-07 | 1.467E-06 | 1.848E-06 | 1.905E-06 | 8.793E-09 | 1.502E-08 | 1.980E-08 | 2.438E-08 | 3.134E-08 | 1.260E-07 | 5.896E-07 | 1.467E-06 | 1.848E-06 | 1.905E-06 |
| | Organic | 1.121E-05 | 1.121E-05 | 1.122E-05 | 1.122E-05 | 1.120E-05 | 1.167E-05 | 1.254E-05 | 1.291E-05 | 1.297E-05 | 1.297E-05 | 1.121E-05 | 1.121E-05 | 1.122E-05 | 1.122E-05 | 1.120E-05 | 1.167E-05 | 1.254E-05 | 1.291E-05 | 1.297E-05 | 1.297E-05 |
| | D(effective) | 1.275E+03 | 7.467E+02 | 5.663E+02 | 4.603E+02 | 3.573E+02 | 9.263E+01 | 2.127E+01 | 8.804E+00 | 7.020E+00 | 6.809E+00 | 6.809E+00 | 1.275E+03 | 7.467E+02 | 5.663E+02 | 4.603E+02 | 3.573E+02 | 9.263E+01 | 2.127E+01 | 8.804E+00 | 7.020E+00 |
| Am | Aqueous | 8.945E-06 | 9.259E-06 | 1.098E-05 | 1.408E-05 | 1.879E-05 | 2.800E-04 | 8.490E-04 | 4.590E-04 | 8.569E-05 | 1.214E-05 | 8.945E-06 | 9.259E-06 | 1.098E-05 | 1.408E-05 | 1.879E-05 | 2.800E-04 | 8.490E-04 | 4.590E-04 | 8.569E-05 | 1.214E-05 |
| | Organic | 2.861E-04 | 2.873E-04 | 2.893E-04 | 2.924E-04 | 2.799E-04 | 8.457E-04 | 4.529E-04 | 8.302E-05 | 1.166E-05 | 1.590E-06 | 2.861E-04 | 2.873E-04 | 2.893E-04 | 2.924E-04 | 2.799E-04 | 8.457E-04 | 4.529E-04 | 8.302E-05 | 1.166E-05 | 1.590E-06 |
| | D(effective) | 3.199E+01 | 3.103E+01 | 2.636E+01 | 2.077E+01 | 1.490E+01 | 3.020E+00 | 5.334E-01 | 1.809E-01 | 1.360E-01 | 1.310E-01 | 1.310E-01 | 3.199E+01 | 3.103E+01 | 2.636E+01 | 2.077E+01 | 1.490E+01 | 3.020E+00 | 5.334E-01 | 1.809E-01 | 1.360E-01 |
| Pu_4 | Aqueous | 4.756E-08 | 5.486E-08 | 6.906E-08 | 9.501E-08 | 1.448E-07 | 7.071E-07 | 3.439E-06 | 8.504E-06 | 1.070E-05 | 1.104E-05 | 4.756E-08 | 5.486E-08 | 6.906E-08 | 9.501E-08 | 1.448E-07 | 7.071E-07 | 3.439E-06 | 8.504E-06 | 1.070E-05 | 1.104E-05 |
| | Organic | 1.734E-04 | 1.734E-04 | 1.734E-04 | 1.734E-04 | 1.733E-04 | 1.761E-04 | 1.811E-04 | 1.833E-04 | 1.836E-04 | 1.837E-04 | 1.734E-04 | 1.734E-04 | 1.734E-04 | 1.734E-04 | 1.733E-04 | 1.761E-04 | 1.811E-04 | 1.833E-04 | 1.836E-04 | 1.837E-04 |
| | D(effective) | 3.645E+03 | 3.160E+03 | 2.511E+03 | 1.825E+03 | 1.197E+03 | 2.490E+02 | 5.266E+01 | 2.155E+01 | 1.716E+01 | 1.664E+01 | 1.664E+01 | 3.645E+03 | 3.160E+03 | 2.511E+03 | 1.825E+03 | 1.197E+03 | 2.490E+02 | 5.266E+01 | 2.155E+01 | 1.716E+01 |
| C2O4 | Aqueous | 1.411E-38 | 2.907E-39 | 1.551E-39 | 1.030E-39 | 5.283E-40 | 1.972E-40 | 3.692E-40 | 1.408E-40 | 1.059E-41 | 5.450E-43 | 1.411E-38 | 2.907E-39 | 1.551E-39 | 1.030E-39 | 5.283E-40 | 1.972E-40 | 3.692E-40 | 1.408E-40 | 1.059E-41 | 5.450E-43 |
| | Organic | 2.062E-39 | 1.215E-39 | 8.752E-40 | 5.431E-40 | 1.946E-40 | 3.670E-40 | 1.383E-40 | 9.829E-42 | 4.892E-43 | 2.381E-44 | 2.062E-39 | 1.215E-39 | 8.752E-40 | 5.431E-40 | 1.946E-40 | 3.670E-40 | 1.383E-40 | 9.829E-42 | 4.892E-43 | 2.381E-44 |
| | D(effective) | 1.461E-01 | 4.180E-01 | 5.644E-01 | 5.271E-01 | 3.683E-01 | 1.861E+00 | 3.745E-01 | 6.883E-02 | 4.621E-02 | 4.369E-02 | 4.369E-02 | 1.461E-01 | 4.180E-01 | 5.644E-01 | 5.271E-01 | 3.683E-01 | 1.861E+00 | 3.745E-01 | 6.883E-02 | 4.621E-02 |

Fig. A-1. Concentration Profile Report from the GTM for the Base-Case TRUOX Process (2 of 3).

| Component Name | Phase | Section No. | | | | | | | |
|----------------|--------------|-------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | 3 | 3 | 3 | 4 | 5 | | | |
| | | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 |
| H | Aqueous | 4.001E-02 | 4.000E-02 | 4.000E-02 | 6.630E-02 | 6.072E-02 | 5.229E-02 | 4.023E-02 | 2.483E-02 |
| | Organic | 2.240E-03 | 2.239E-03 | 2.239E-03 | 5.908E-02 | 5.119E-02 | 3.989E-02 | 2.550E-02 | 1.087E-02 |
| | D(effective) | 5.598E-02 | 5.597E-02 | 5.598E-02 | 8.911E-01 | 8.430E-01 | 7.628E-01 | 6.339E-01 | 4.380E-01 |
| A1 | Aqueous | 1.471E-15 | 8.909E-18 | 5.363E-20 | 3.215E-22 | 2.074E-24 | 1.338E-26 | 8.630E-29 | 5.562E-31 |
| | Organic | 1.471E-18 | 8.909E-21 | 5.363E-23 | 3.215E-25 | 2.074E-27 | 1.338E-29 | 8.630E-32 | 5.562E-34 |
| | D(effective) | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| UO2 | Aqueous | 1.909E-06 | 1.874E-06 | 1.634E-06 | 1.102E-05 | 7.977E-08 | 5.146E-10 | 3.320E-12 | 2.139E-14 |
| | Organic | 1.294E-05 | 1.269E-05 | 1.107E-05 | 1.913E-08 | 7.977E-11 | 5.146E-13 | 3.320E-15 | 2.139E-17 |
| | D(effective) | 6.777E+00 | 6.773E+00 | 6.772E+00 | 1.737E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| Am | Aqueous | 1.654E-06 | 2.212E-07 | 2.645E-08 | 3.557E-09 | 2.295E-11 | 1.480E-13 | 9.549E-16 | 6.153E-18 |
| | Organic | 2.155E-07 | 2.979E-08 | 3.442E-09 | 3.557E-12 | 2.295E-14 | 1.480E-16 | 9.549E-19 | 6.153E-21 |
| | D(effective) | 1.302E-01 | 1.301E-01 | 1.301E-01 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| Pu_4 | Aqueous | 1.109E-05 | 1.105E-05 | 1.043E-05 | 1.718E-04 | 1.108E-06 | 7.148E-09 | 4.611E-11 | 2.971E-13 |
| | Organic | 1.836E-04 | 1.829E-04 | 1.725E-04 | 1.718E-07 | 1.109E-09 | 7.148E-12 | 4.611E-14 | 2.971E-16 |
| | D(effective) | 1.656E+01 | 1.655E+01 | 1.655E+01 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| C2O4 | Aqueous | 2.667E-44 | 1.293E-45 | 5.995E-47 | 3.151E-02 | 2.976E-02 | 2.561E-02 | 1.956E-02 | 1.188E-02 |
| | Organic | 1.156E-45 | 5.621E-47 | 2.758E-48 | 2.941E-02 | 2.552E-02 | 1.985E-02 | 1.267E-02 | 5.395E-03 |
| | D(effective) | 4.335E-02 | 4.346E-02 | 4.597E-02 | 9.333E-01 | 8.575E-01 | 7.752E-01 | 6.479E-01 | 4.542E-01 |

Fig. A-1. Concentration Profile Report from the GTM for the Base-Case TRUJEX Process (3 of 3).

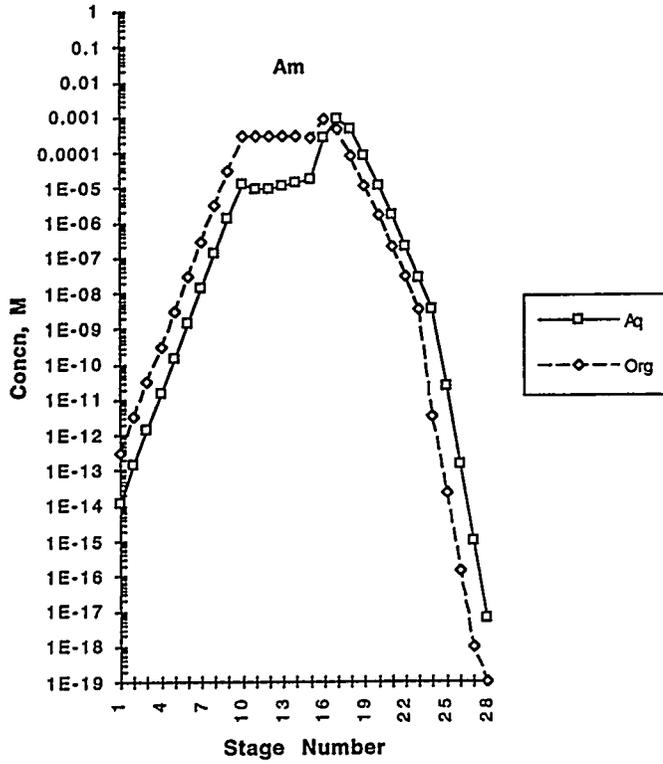
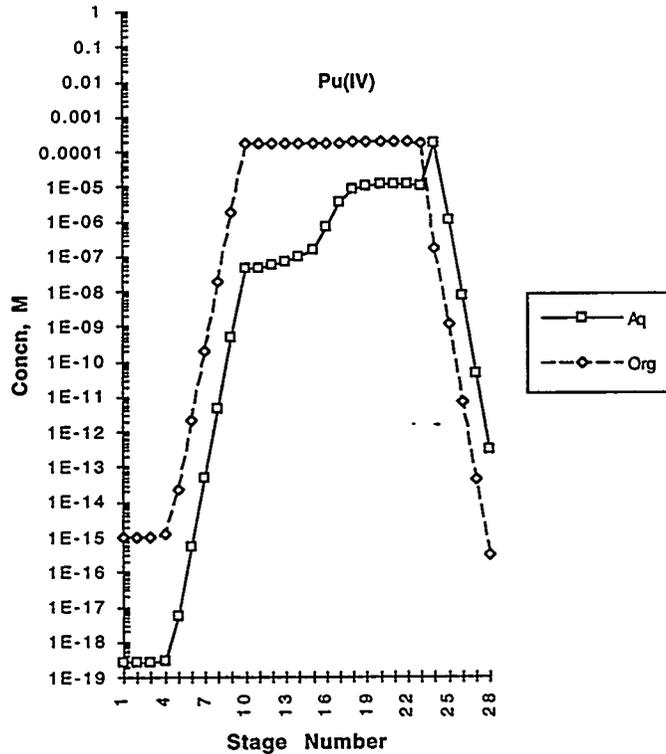


Fig. A-2.

Americium Concentration Profile from the GTM for the Base-Case TRUEX Process

Fig. A-3.

Plutonium Concentration Profile from the GTM for the Base-Case TRUEX Process



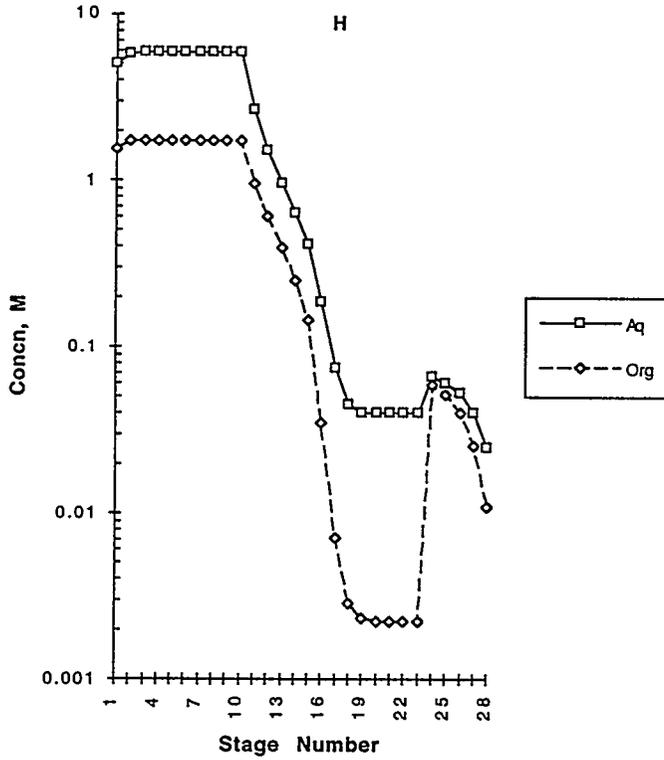
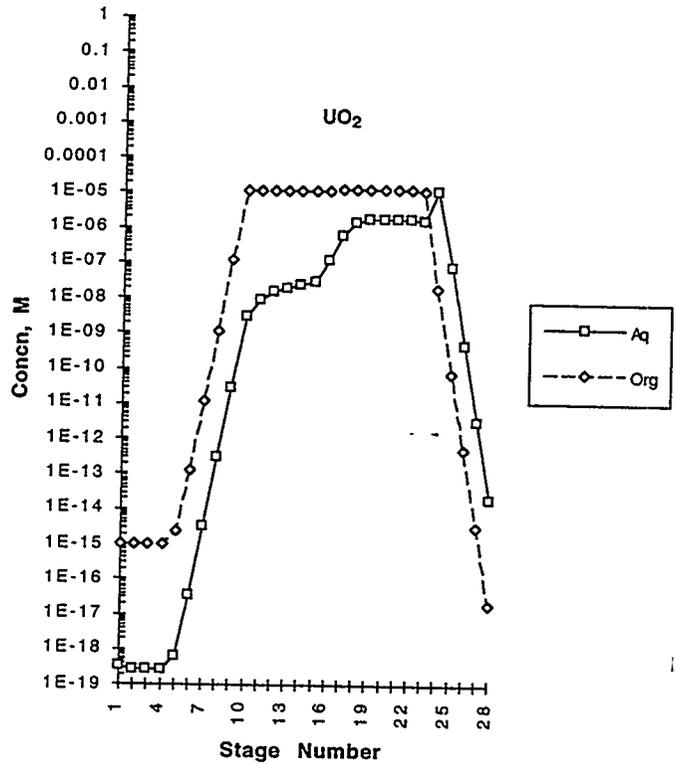


Fig. A-4.

Hydrogen Concentration Profile from the GTM for the Base-Case TRUEX Process

Fig. A-5.

Uranyl Concentration Profile from the GTM for the Base-Case TRUEX Process



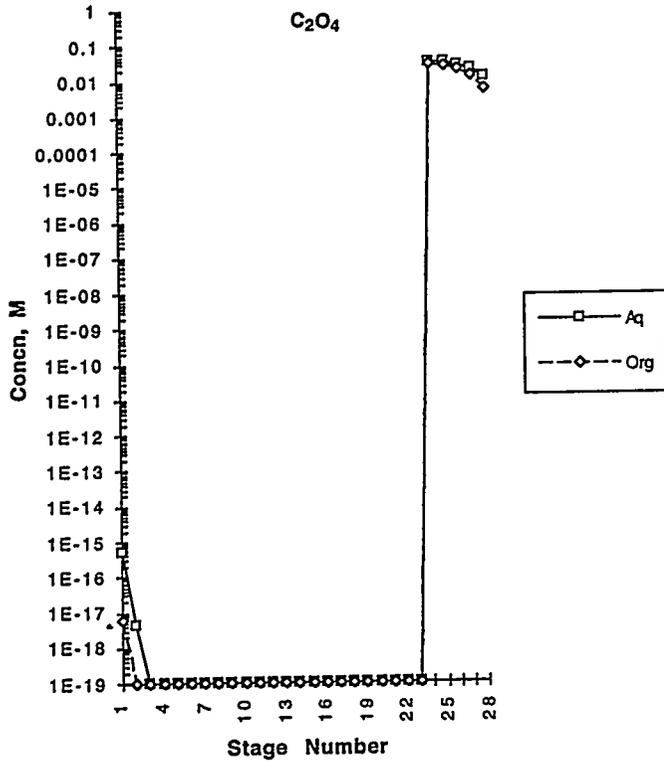
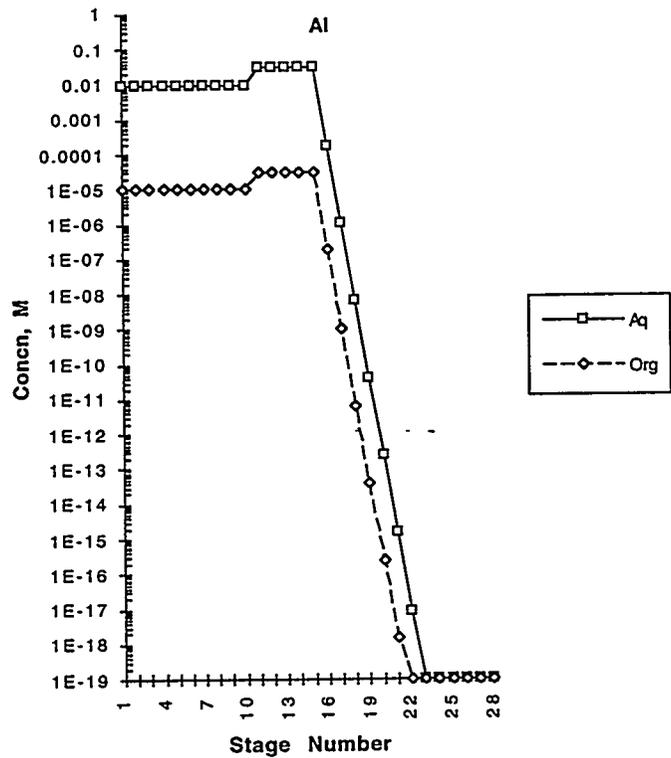


Fig. A-6.

Oxalate Concentration Profile from the GTM for the Base-Case TRUEX Process

Fig. A-7.

Aluminium Concentration Profile from the GTM for the Base-Case TRUEX Process



APPENDIX B

GTM Reports for the Alternative TRUEX Flowsheet

The concentration profile report and the chart for each component were generated by the GTM for the alternative flowsheet shown in Fig. IV-1. These items are included here as Figs. B-1 through B-7. The full summary report generated by the GTM version 2.4 is included in the main body of this report as Table IV-1.

| Component Name | Phase | Stage-to-Stage Profile for each Component | | | | | | | | | | | | |
|----------------|--------------|---|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | Section No. Stage No. | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
| H | Aqueous | x, M | 4.972E+00 | 5.729E+00 | 5.817E+00 | 5.829E+00 |
| | Organic | y, M | 1.528E+00 | 1.703E+00 | 1.724E+00 | 1.727E+00 |
| | D(effective) | | 3.074E-01 | 2.973E-01 | 2.984E-01 | 2.983E-01 | 2.983E-01 | 2.982E-01 |
| | | | 1.489E-02 |
| A1 | Aqueous | x, M | 1.489E-02 |
| | Organic | y, M | 1.489E-05 |
| | D(effective) | | 1.000E-03 |
| | | | 3.401E-19 | 2.699E-19 | 2.621E-19 | 2.620E-19 | 2.620E-19 | 2.660E-19 |
| UC2 | Aqueous | x, M | 9.999E-16 | 9.998E-16 |
| | Organic | y, M | 2.940E+03 | 3.704E+03 | 3.803E+03 | 3.815E+03 | 3.817E+03 |
| | D(effective) | | 1.626E-16 | 1.534E-15 | 1.506E-14 | 1.485E-13 | 1.484E-12 | 1.444E-11 | 1.424E-10 | 1.405E-09 | 1.385E-08 | 1.366E-07 | 1.347E-06 | 1.331E-05 |
| | | | 4.075E-15 | 3.409E-14 | 3.302E-13 | 3.250E-12 | 3.205E-11 | 3.160E-10 | 3.117E-09 | 3.074E-08 | 3.031E-07 | 2.990E-06 | 2.948E-05 | 2.860E-04 |
| Am | Aqueous | x, M | 2.508E+01 | 2.222E+01 | 2.193E+01 | 2.189E+01 |
| | Organic | y, M | 2.578E-19 | 2.565E-19 | 2.564E-19 | 2.564E-19 | 2.570E-19 |
| | D(effective) | | 1.000E-15 | 1.000E-15 | 1.000E-15 | 1.000E-15 | 1.003E-15 | 1.241E-15 | 2.380E-14 | 2.159E-12 | 2.044E-10 | 1.935E-08 | 1.832E-06 | 1.734E-04 |
| | | | 3.880E+03 | 3.899E+03 | 3.901E+03 |
| Pu_4 | Aqueous | x, M | 4.963E-16 | 4.365E-18 | 2.200E-20 | 1.053E-22 | 6.005E-25 | 2.378E-27 | 1.130E-29 | 5.366E-32 | 2.540E-34 | 1.211E-36 | 5.753E-39 | 2.733E-41 |
| | Organic | y, M | 6.236E-18 | 2.207E-20 | 9.992E-23 | 4.719E-25 | 2.240E-27 | 1.064E-29 | 5.055E-32 | 2.401E-34 | 1.141E-36 | 6.419E-39 | 2.674E-41 | 1.223E-43 |
| | D(effective) | | 1.257E-02 | 6.055E-03 | 4.541E-03 | 4.463E-03 | 4.476E-03 | 4.475E-03 |
| | | | 1.626E-16 | 1.534E-15 | 1.506E-14 | 1.485E-13 | 1.484E-12 | 1.444E-11 | 1.424E-10 | 1.405E-09 | 1.385E-08 | 1.366E-07 | 1.347E-06 | 1.331E-05 |
| C204 | Aqueous | x, M | 4.963E-16 | 4.365E-18 | 2.200E-20 | 1.053E-22 | 6.005E-25 | 2.378E-27 | 1.130E-29 | 5.366E-32 | 2.540E-34 | 1.211E-36 | 5.753E-39 | 2.733E-41 |
| | Organic | y, M | 6.236E-18 | 2.207E-20 | 9.992E-23 | 4.719E-25 | 2.240E-27 | 1.064E-29 | 5.055E-32 | 2.401E-34 | 1.141E-36 | 6.419E-39 | 2.674E-41 | 1.223E-43 |
| | D(effective) | | 1.257E-02 | 6.055E-03 | 4.541E-03 | 4.463E-03 | 4.476E-03 | 4.475E-03 |
| | | | 1.626E-16 | 1.534E-15 | 1.506E-14 | 1.485E-13 | 1.484E-12 | 1.444E-11 | 1.424E-10 | 1.405E-09 | 1.385E-08 | 1.366E-07 | 1.347E-06 | 1.331E-05 |

Fig. B-1. Concentration Profile Report from the GTM for the Alternative TRUJEX Process (1 of 3)

| Component Name | Phase | Section No. | | | | | | | | | | | |
|----------------|--------------|-------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | 25 | 26 | 27 | 28 | 29 | 30 | 31 | 32 | 33 | 34 | 35 | 36 |
| H | Aqueous | 5.773E-02 | 4.701E-02 | 4.170E-02 | 4.035E-02 | 4.007E-02 | 4.001E-02 | 4.000E-02 | 6.598E-02 | 6.027E-02 | 5.208E-02 | 4.049E-02 | 2.527E-02 |
| | Organic | 7.000E-03 | 3.378E-03 | 2.469E-03 | 2.284E-03 | 2.247E-03 | 2.240E-03 | 2.239E-03 | 6.888E-02 | 6.131E-02 | 4.046E-02 | 2.823E-02 | 1.119E-02 |
| | D(effective) | 1.213E-01 | 7.184E-02 | 5.922E-02 | 5.660E-02 | 5.600E-02 | 5.599E-02 | 5.597E-02 | 8.938E-01 | 8.514E-01 | 7.768E-01 | 6.478E-01 | 4.429E-01 |
| Al | Aqueous | 1.479E-18 | 1.284E-20 | 1.120E-22 | 9.766E-25 | 8.516E-27 | 7.426E-29 | 6.420E-31 | 3.849E-33 | 2.493E-35 | 1.602E-37 | 1.033E-39 | 6.658E-42 |
| | Organic | 1.473E-21 | 1.284E-23 | 1.120E-25 | 9.766E-28 | 8.516E-30 | 7.426E-32 | 6.420E-34 | 3.849E-36 | 2.493E-38 | 1.602E-40 | 1.033E-42 | 6.658E-45 |
| | D(effective) | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| LiCl | Aqueous | 7.887E-07 | 1.053E-08 | 1.621E-06 | 1.800E-06 | 1.836E-06 | 1.819E-06 | 1.652E-06 | 1.114E-05 | 8.076E-08 | 5.210E-10 | 3.361E-12 | 2.166E-14 |
| | Organic | 1.192E-05 | 1.244E-05 | 1.244E-05 | 1.246E-05 | 1.245E-05 | 1.233E-05 | 1.119E-05 | 1.945E-08 | 8.076E-11 | 5.210E-13 | 3.361E-15 | 2.166E-17 |
| | D(effective) | 1.512E+01 | 1.169E+01 | 7.672E+00 | 6.924E+00 | 6.781E+00 | 6.778E+00 | 6.773E+00 | 1.748E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| Am | Aqueous | 3.917E-03 | 1.909E-03 | 7.177E-04 | 1.628E-04 | 3.246E-05 | 6.152E-06 | 1.009E-06 | 1.355E-07 | 8.742E-10 | 5.640E-12 | 3.638E-14 | 2.344E-16 |
| | Organic | 1.301E-03 | 4.868E-04 | 1.091E-04 | 2.176E-05 | 4.226E-06 | 8.016E-07 | 1.311E-07 | 1.355E-10 | 8.742E-13 | 5.640E-15 | 3.638E-17 | 2.344E-19 |
| | D(effective) | 3.321E-01 | 2.551E-01 | 1.520E-01 | 1.337E-01 | 1.302E-01 | 1.303E-01 | 1.301E-01 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| Pu_4 | Aqueous | 4.821E-06 | 6.293E-06 | 9.632E-06 | 1.068E-05 | 1.091E-05 | 1.089E-05 | 1.047E-05 | 1.724E-04 | 1.112E-06 | 7.176E-09 | 4.629E-11 | 2.983E-13 |
| | Organic | 1.777E-04 | 1.800E-04 | 1.807E-04 | 1.808E-04 | 1.808E-04 | 1.805E-04 | 1.793E-04 | 1.724E-07 | 1.112E-08 | 7.176E-12 | 4.629E-14 | 2.983E-16 |
| | D(effective) | 3.685E+01 | 2.860E+01 | 1.878E+01 | 1.692E+01 | 1.657E+01 | 1.657E+01 | 1.655E+01 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 | 1.000E-03 |
| C2O4 | Aqueous | 1.402E-48 | 1.042E-49 | 7.456E-51 | 5.280E-52 | 3.742E-53 | 2.639E-54 | 1.745E-55 | 3.144E-02 | 2.971E-02 | 2.568E-02 | 1.973E-02 | 1.202E-02 |
| | Organic | 6.600E-50 | 4.629E-51 | 3.275E-52 | 2.322E-53 | 1.646E-54 | 1.162E-55 | 7.992E-57 | 2.944E-02 | 2.665E-02 | 2.008E-02 | 1.288E-02 | 5.464E-03 |
| | D(effective) | 4.635E-02 | 4.440E-02 | 4.392E-02 | 4.397E-02 | 4.395E-02 | 4.402E-02 | 4.578E-02 | 9.362E-01 | 8.633E-01 | 7.820E-01 | 6.527E-01 | 4.545E-01 |

Fig. B-1. Concentration Profile Report from the GTM for the Alternative TRUEX Process (3 of 3)

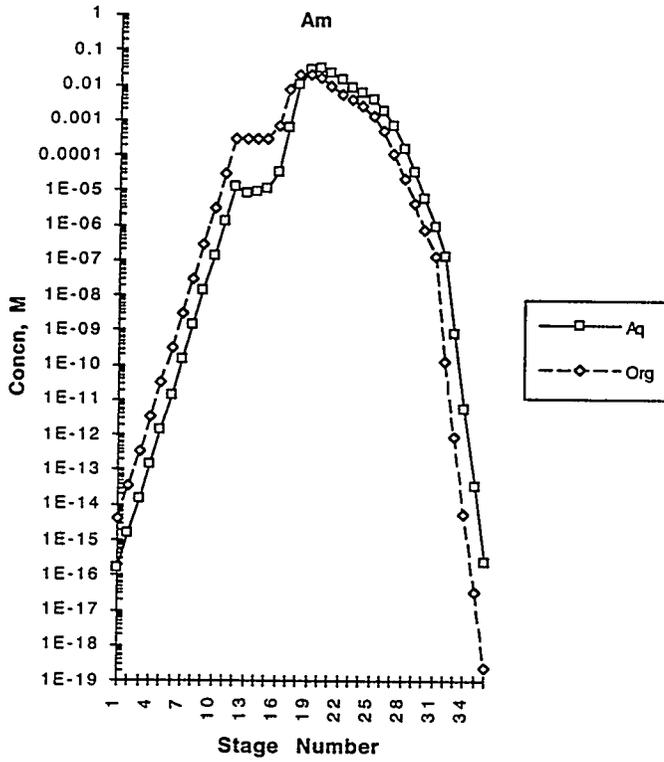
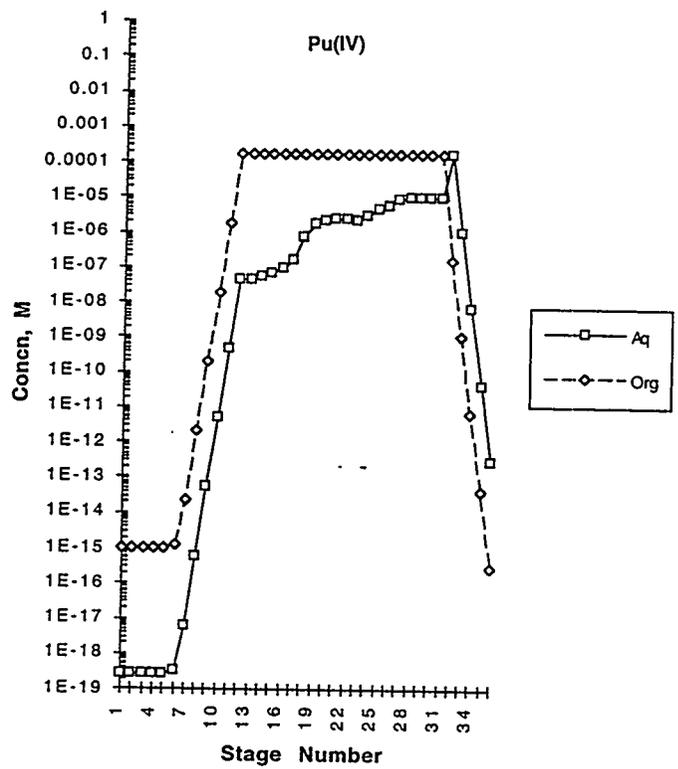


Fig. B-2.

Americium Concentration Profile from the GTM for the Alternative TRUEX Process

Fig. B-3.

Plutonium Concentration Profile from the GTM for the Alternative TRUEX Process



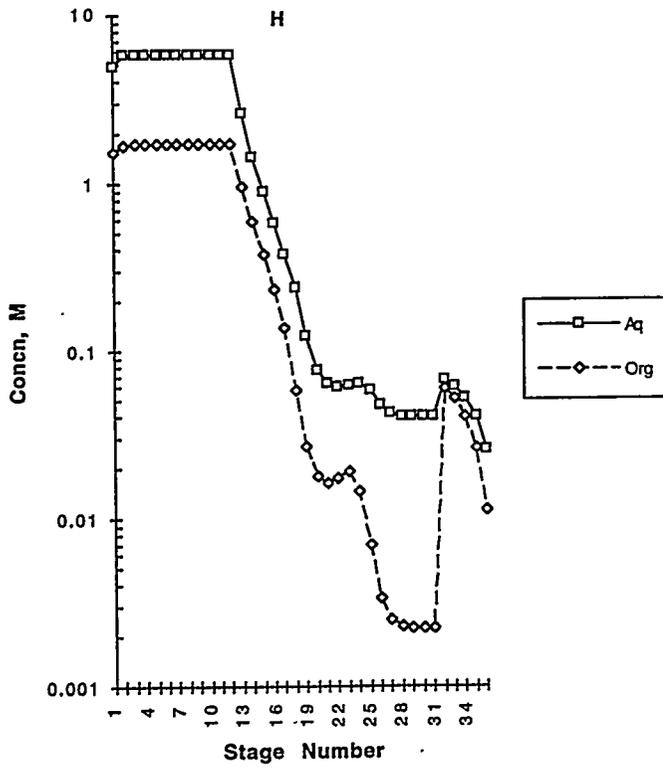
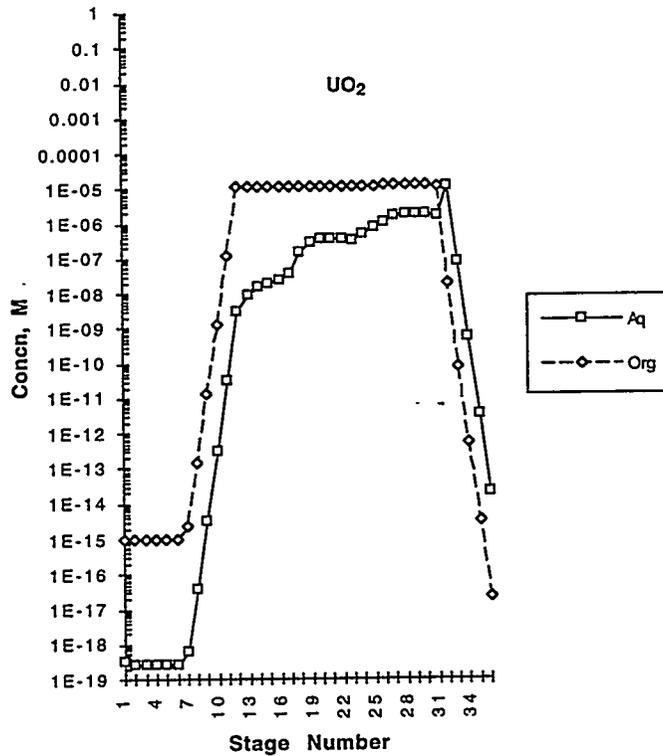


Fig. B-4.

Hydrogen Concentration Profile from the GTM for the Alternative TRUOX Process

Fig. B-5.

Uranyl Concentration Profile from the GTM for the Alternative TRUOX Process



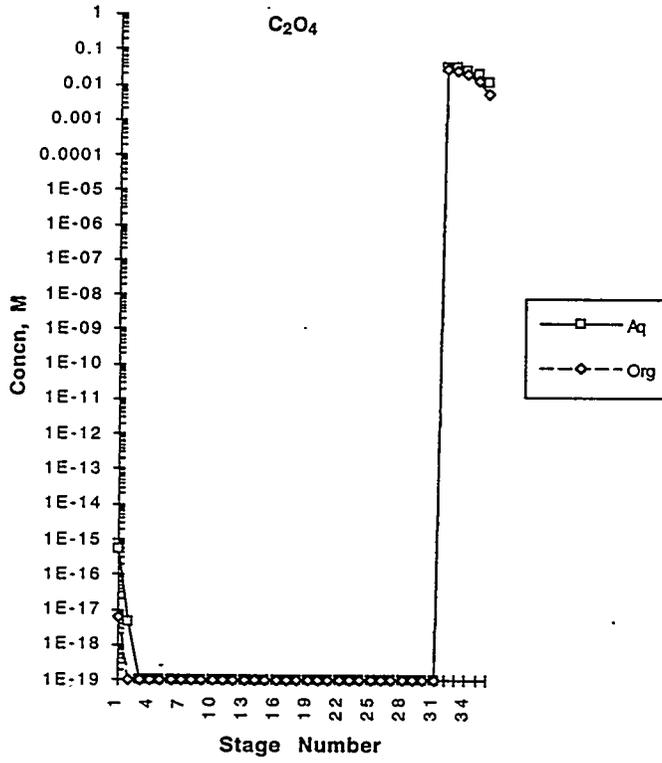
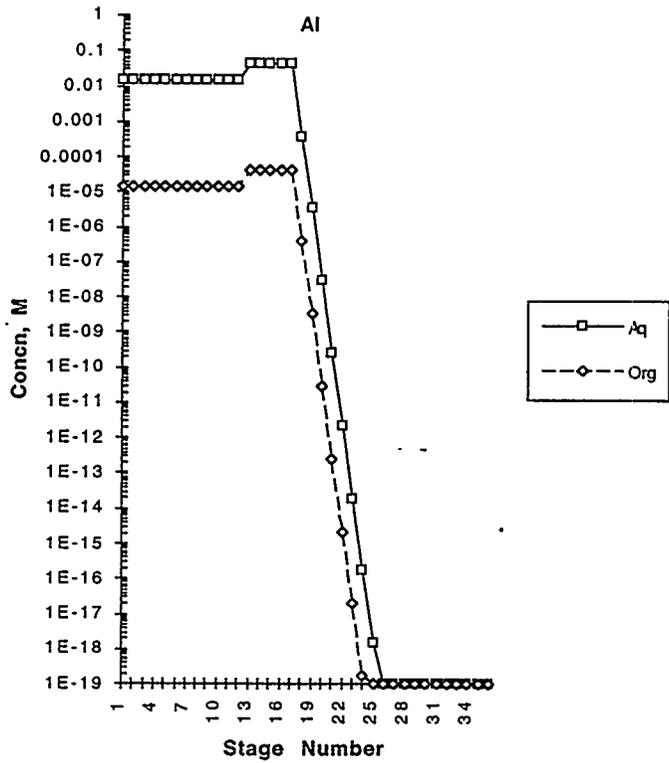


Fig. B-6.

Oxalate Concentration Profile from the GTM for the Alternative TRUEX Process

Fig. B-7.

Aluminum Concentration Profile from the GTM for the Alternative TRUEX Process



Distribution for ANL-94/33Internal:

| | | |
|-------------------|--------------------|-----------------------|
| J. M. Andrew | D. Dong | J. Sedlet |
| B. D. Babcock | J. E. Harmon | W. B. Seefeldt |
| J. E. Battles | E. P. Horwitz | S. A. Slater |
| B. A. Buchholz | J. C. Hutter | B. Srinivasan |
| D. B. Chamberlain | R. J. Jaskot | M. J. Steindler |
| L. Chen | J. J. Laidler | G. F. Vandegrift (10) |
| M. K. Clemens | R. A. Leonard (10) | R. D. Wolson |
| C. J. Conner | C. J. Mertz | D. G. Wygmans |
| J. M. Copple | L. Nunez | TIS Files |
| H. Diamond | M. C. Regalbuto | |

External:

DOE-OSTI (2)

ANL-E Library (2)

ANL-W Library

Manager, Chicago Operations Office, DOE

A. Bindokas, DOE-CH

J. Haugen, DOE-CH

A. L. Taboas, DOE-CH/AAO

Chemical Technology Division Review Committee Members:

E. R. Beaver, Monsanto Company, St. Louis, MO

D. L. Douglas, Consultant, Bloomington, MN

R. K. Genung, Oak Ridge National Laboratory, Oak Ridge, TN

J. G. Kay, Drexel University, Philadelphia, PA

G. R. St. Pierre, Ohio State University, Columbus, OH

J. Stringer, Electric Power Research Institute, Palo Alto, CA

J. B. Wagner, Arizona State University, Tempe, AZ

M. G. Adamson, Lawrence Livermore National Laboratory, Livermore, CA

J. Allison, USDOE, Office of Waste Operations, Washington, DC

S. Barker, Westinghouse Hanford Company, Richland, WA

C. L. Bendixsen, Lockheed Idaho Technology Company, Idaho Falls, ID

D. Benker, Oak Ridge National Laboratory, Oak Ridge, TN

S. E. Berk, USDOE, Office of Fusion Energy, Germantown, MD

W. Bourcier, Lawrence Livermore National Laboratory, Livermore, CA

D. F. Bowersox, Los Alamos National Laboratory, Los Alamos, NM

L. Brewer, University of California, Berkeley, CA

K. A. Chacey, USDOE, Office of Waste Management, Germantown, MD

M. O. Cloninger, Mac Technical Services, Inc., Richland, WA

M. S. Coops, Lawrence Livermore National Laboratory, Livermore, CA

M. Dinehart, Los Alamos National Laboratory, Los Alamos, NM

M. C. Dionisio, USDOE, Office of Technology Development, Washington, DC
M. Duff, USDOE, Office of Waste Operations, Germantown, MD
R. E. Erickson, USDOE, Office of Waste Operations, Germantown, MD
G. Escobar, Rocky Flats Plant, Golden, CO
R. C. Ewing, University of New Mexico, Albuquerque, NM
R. Fish, B&W Fuel Company, Las Vegas, NV
C. W. Frank, USDOE, Office of Technology Development, Washington, DC
T. Fryberger, USDOE, Office of Environmental Management, Germantown, MD
S. M. Gibson, USDOE, Office of Environmental Management, Germantown, MD
R. Gilchrist, Westinghouse Hanford Company, Richland, WA
R. Goheen, Pacific Northwest Laboratory, Richland, WA
R. Herbst, Los Alamos National Laboratory, Germantown, MD
S. Herbst, Lockheed Idaho Technology Company, Idaho Falls, ID
T. M. Hohl, Westinghouse Hanford Company, Richland, WA
J. O. Honeyman, Westinghouse Hanford Operations, Richland, WA
C. Interrante, U. S. Nuclear Regulatory Commission, Washington, DC
G. Jansen, Westinghouse Hanford Company, Richland, WA
C. Jantzen, Westinghouse Savannah River Company, Aiken, SC
E. Jones, Pacific Northwest Laboratory, Richland, WA
R. T. Jubin, Oak Ridge National Laboratory, Oak Ridge, TN
R. Kirkbride, Lockheed Idaho Technology Company, Richland, WA
D. A. Knecht, Lockheed Idaho Technology Company, Idaho Falls, ID
B. Knutson, Westinghouse Hanford Company, Richland, WA
M. Levenson, Menlo Park, CA
S. C. T. Lien, USDOE, Office of Technology Development, Germantown, MD
G. J. Lumetta, Pacific Northwest Laboratory, Richland, WA
W. Lutze, University of New Mexico, Albuquerque, NM
C. P. McGinnis, Oak Ridge National Laboratory, Oak Ridge, TN
R. McKee, Pacific Northwest Laboratory, Richland, WA
W. H. McVey, USDOE, Division of LMFBR Fuel Cycle Projects, Germantown, MD
R. Morissette, Science Applications International Corp., Las Vegas, NV
A. C. Muscatello, Rocky Flats Plant, Golden, CO
P. K. Nair, Southwest Research Institute, San Antonio, TX
J. L. Nelson, Westinghouse Hanford Co., Richland, WA
M. O'Hara, Martin Marietta Energy Systems, Oak Ridge, TN
D. R. Olander, University of California, Berkeley, CA
A. L. Olson, Lockheed Idaho Technology Company, Idaho Falls, ID
R. Orth, Pacific Northwest Laboratory, Richland, WA
M. Palmer, Los Alamos National Laboratory, Los Alamos, NM
R. Palmer, West Valley Nuclear Services, West Valley, NY
G. Pfennigworth, Martin Marietta Energy Systems, Oak Ridge, TN
W. G. Ramsey, Westinghouse Savannah River Company, Aiken, SC
G. Reddick, Westinghouse Hanford Company, Richland, WA
C. G. Russomanno, USDOE, Civilian Radioactive Waste Management, Washington, DC
P. S. Schaus, Westinghouse Hanford Company, Richland, WA
H. Shaw, Lawrence Livermore National Laboratory, Livermore, CA

D. Siemer, Lockheed Idaho Technology Company, Idaho Falls, ID
E. Slaathug, Westinghouse Hanford Company, Richland, WA
B. Spencer, Oak Ridge National Laboratory, Oak Ridge, TN
J. Sproull, Westinghouse Savannah River Company, Aiken, SC
D. Stahl, M&O/B&W Fuel Company, Las Vegas, NV
R. B. Stout, Lawrence Livermore National Laboratory, Livermore, CA
J. L. Swanson, Pacific Northwest Laboratory, Richland, WA
D. W. Tedder, Georgia Institute of Technology, Atlanta, GA
M. Thompson, Westinghouse Savannah River Company, Aiken, SC
T. A. Todd, Lockheed Idaho Technology Co., Idaho Falls, ID
USDOE, Office of Basic Energy Sciences, Washington, DC
R. M. Wham, Oak Ridge National Laboratory, Oak Ridge, TN
S. Yarbro, Los Alamos National Laboratory, Los Alamos, NM