RK Sharp

BNWL-204 PT2

AEC
RESEARCH
and
DEVELOPMENT
REPORT

AQUEOUS DECLADDING AND DISSOLUTION OF
PLUTONIUM RECYCLE TEST REACTOR FUELS,
PART 2: $\text{PuO}_2\text{-UO}_2$ FUELS

W. W. SCHULZ

NOVEMBER, 1966

BATTELLE-NORTHWEST

BATTELLE MEMORIAL INSTITUTE / PACIFIC NORTHWEST LABORATORY

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

BNWL-204 PT2
UC-10, Chemical Separations
Processes for Plutonium
and Uranium

AQUEOUS DECLADDING AND DISSOLUTION OF PLUTONIUM
RECYCLE TEST REACTOR FUELS. PART 2: PuO_2 - UO_2 FUELS

By

W. W. Schulz

Chemical Development Section
Chemistry Department

Laboratory work done by:

D. G. Bouse
L. C. Neil
W. W. Schulz

November 1966

FIRST UNRESTRICTED
DISTRIBUTION MADE DEC 8 '66

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

Printed in USA. Price \$1.00. Available from the
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards
U. S. Department of Commerce
Springfield, Virginia

TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	1
SUMMARY AND CONCLUSIONS	1
FUEL DESCRIPTION	1
DISSOLUTION CHEMISTRY AND LITERATURE REVIEW	2
EXPERIMENTAL	3
Fuel Samples	3
Dissolution Equipment	3
Decladding and Dissolution Procedures	4
PuO ₂ Powder	4
PuO ₂ -UO ₂ Fuels	4
Decladding Process	4
Dissolution Schemes	5
Special Dissolution Schemes	6
Analyses	6
RESULTS	7
Zirflex Process Decladding of PuO ₂ -UO ₂ Fuels	7
Dissolution of PuO ₂ Powder in HNO ₃ -NH ₄ F Solutions	8
Dissolution of PRTR PuO ₂ -UO ₂ Fuels in HNO ₃ -NH ₄ F Solutions	10
Dissolution of PRTR PuO ₂ -UO ₂ Fuels in HNO ₃	13
CHEMICAL FLOWSHEETS	14
REDOX PLANT DISSOLUTION EXPERIENCE	14
ACKNOWLEDGEMENT	16
REFERENCES	16

AQUEOUS DECLADDING AND DISSOLUTION OF
PLUTONIUM RECYCLE TEST REACTOR FUELS
PART 2: $\text{PuO}_2\text{-UO}_2$ FUELS

INTRODUCTION

This is the second of a two-part report describing laboratory studies of the decladding and dissolution of various Plutonium Recycle Test Reactor (PRTR) fuels. These studies were performed to develop satisfactory aqueous headend procedures for use in Hanford Redox and/or Purex plant reprocessing of PRTR fuels. Studies with $\text{PuO}_2\text{-UO}_2$ fuels are discussed in this part; studies with plutonium-aluminum alloy fuels are discussed in Part 1.

SUMMARY AND CONCLUSIONS

Laboratory studies of the decladding and dissolution of PRTR $\text{UO}_2\text{-0.5}$ to 1.0 wt% PuO_2 fuels were performed with both unirradiated and irradiated material. These studies culminated in two satisfactory procedures for dissolution of the $\text{PuO}_2\text{-UO}_2$ core. One procedure employs a boiling $\text{HNO}_3\text{-NH}_4\text{F}$ solution as the solvent. It is suitable for use in a dissolver constructed of a material resistant to the highly corrosive boiling acid-fluoride solution; the service dissolver recently proposed of the Hanford Purex Plant is an example of such a dissolver. The second dissolution technique is suitable for at least limited use in stainless steel

equipment; the technique employs a $\text{HNO}_3\text{-NH}_4\text{F}$ solution at 60 to 80 °C to dissolve the $\text{PuO}_2\text{-UO}_2$ core. This latter procedure was used in the Hanford Redox plant to dissolve three charges (29 assemblies) of PRTR $\text{UO}_2\text{-0.5}$ wt% PuO_2 fuel.

Plutonium dioxide in unirradiated PRTR $\text{PuO}_2\text{-UO}_2$ oxide mixture fuels was only sparsely (0.6 to 6.0%) soluble in HNO_3 solutions not containing fluoride. Irradiation of such fuels to 1000 to 2000 MWd/ton U apparently resulted in significant in-reactor conversion of oxide mixtures to solid solutions as testified by the much greater (60 to 75%) solubility of the plutonium in HNO_3 . The degree of conversion to solid solution increases with exposure level reflecting, probably, the effects of a longer time at elevated temperature. A single datum indicates essentially complete conversion to a solid solution occurs during sufficient reactor irradiation (~ 5000 MWd/ton U).

FUEL DESCRIPTION

The basic oxide mixture ($\text{PuO}_2\text{-UO}_2$) PRTR fuel element is a 19 rod cluster, which is 3.25 in. in diameter and 8 ft long. Each rod is individually clad in zircaloy (0.035 in. wall thickness); the diameter of the clad rod is nomin-

ally 0.57 in. The fuel composition is UO_2 -0.48 or 1.0 wt% PuO_2 *. Each element contains approximately 117 lb of UO_2 and 29 lb of zircaloy including end fittings.

Two versions of this basic fuel element have been fabricated and irradiated. Mark 1-L elements are vibrationally compacted while Mark 1-M elements are swage compacted. Each of these compaction techniques can be applied to either (1) physical mixtures of ceramic grade UO_2 and PuO_2 or (2) sized particles of PuO_2 dispersed in UO_2 as formed by a high energy rate, pneumatic-impaction. Elements prepared from physical mixtures of UO_2 and PuO_2 are prepared by an incremental loading technique and are called "incremental loaded" in this report; elements loaded with pneumatically impacted fuel are termed "Nupac." Detailed dimensions and fabrication techniques for Mark 1-L and 1-M elements are given in references (1) and (2). Specifications for nuclear grade UO_2 , PuO_2 , and oxide mixtures for use in preparing PRTR PuO_2 - UO_2 fuels have also been listed.^(3,4,5)

PuO_2 - UO_2 reactor fuels can be prepared most simply by physically mixing ceramic grade PuO_2 and UO_2 in the desired proportions and then compacting to desired densities. PRTR PuO_2 - UO_2 fuels are prepared this way; Wilson⁽⁶⁾ refers to fuels prepared in this manner as an "oxide mixture." Also, PuO_2 - UO_2 fuels can be prepared by coprecipitation

of uranium(VI) and plutonium(IV) ions in ammoniacal solutions, ignition of the resulting precipitate, and hydrogen reduction of the dried precipitate. The resulting product, termed a "mixed oxide" by Wilson, is a single phase which is a solid solution of UO_2 and PuO_2 . A distinguishing feature of these two types of PuO_2 - UO_2 fuels is that the mixed oxide is readily soluble in HNO_3 alone. Addition of fluoride ion to the HNO_3 is required to dissolve the oxide mixture. Prolonged sintering of oxide mixtures at high temperatures in a hydrogen atmosphere converts them to mixed oxides.^(7,8)

The aluminum alloy 6061 T canisters used to ship PRTR 19-rod cluster elements to the Hanford Redox plant are described in Part 1. It is expected that similar equipment will be used to transport PuO_2 - UO_2 elements to the Hanford Purex plant.

DISSOLUTION CHEMISTRY AND LITERATURE REVIEW

The whole subject of dissolution of PuO_2 in mineral acids, as well as by fusion and halogenation techniques, has been reviewed recently by Gelman.⁽⁹⁾ The brief review given here is confined to dissolution in mineral acids.

Rainey and Uriarte at Oak Ridge National Laboratory (ORNL) have recently completed a comprehensive study of the initial instantaneous dissolution rates of unirradiated, commercially prepared UO_2 , PuO_2 , and PuO_2 - UO_2 pellets in boiling HNO_3 alone and with additives.⁽⁷⁾ Dissolution of PuO_2 powders in HNO_3 solutions containing added fluoride has also been studied by European workers.⁽¹⁰⁾

* Recently PuO_2 - UO_2 fuel elements containing 2 wt% PuO_2 have been fabricated for use in the PRTR. It is expected technology developed for 0.5 and 1 wt% fuels will apply also to the 2 wt% material.

In general, UO_2 dissolves readily in either HNO_3 , HNO_3 -HF, or HNO_3 - $\text{Al}(\text{NO}_3)_3$ -HF solutions. Plutonium dioxide, however, is virtually insoluble either in boiling, concentrated HNO_3 alone or in HNO_3 containing additives such as O_3 , H_2O_2 , $\text{Na}_2\text{Cr}_2\text{O}_7$, or HCl . Boiling 9 M HBr dissolves refractory PuO_2 ⁽¹¹⁾, but this solvent is not satisfactory for use in stainless steel equipment. Wilson found cerium (IV) ions catalyzed HNO_3 dissolution of PuO_2 .⁽⁶⁾ Subsequent work at ORNL has confirmed the existence of the catalytic effect but indicated it to be too small to be of practical value in plant aqueous reprocessing schemes for PuO_2 - UO_2 fuels.

The dissolution rate of PuO_2 in HNO_3 solutions is greatly increased by addition of fluoride. All investigators presently regard such solvents as the most suitable for headend reprocessing of PuO_2 - UO_2 fuels and they were so used in this work.

It is well known that HNO_3 -fluoride solutions, especially when heated to boiling temperature, corrode stainless steel equipment. Some pertinent corrosion data are listed in a recent paper by Maness.⁽¹²⁾ To decrease corrosion in such systems, one may add $\text{Al}(\text{NO}_3)_3$ to complex the fluoride ion. This practice cannot be followed in dissolving PuO_2 . ORNL workers found⁽⁷⁾, as we did, that the effect of fluoride ion in the dissolution of PuO_2 is practically nullified by addition of three moles of $\text{Al}(\text{NO}_3)_3$ per mole of fluoride.

EXPERIMENTAL

Fuel Samples

Initial dissolution studies were performed with PuO_2 powder of the type

used in fabricating PRTR PuO_2 - UO_2 fuel rods. This material was prepared by calcining plutonium(IV) oxalate for 3 to 4 hr at 950 °C. The resulting PuO_2 particles were about 17 μ in diam and had a density about 95% TD.

The bulk of the dissolution studies were performed with 1/2 to 1 in. long pieces cut from PRTR PuO_2 - UO_2 fuel rods; these are described in Table I. Except for the ends, zircaloy cladding was intact. In all cases the fuel rods had been autoclaved. In some experiments zircaloy cladding was removed chemically before dissolution of the PuO_2 - UO_2 core. In other instances, however, the cladding was removed mechanically and the chemical decladding step was omitted.

Finally, a few experiments were performed with sintered UO_2 -0.05 wt% PuO_2 pellets. These pellets (1/2 in. high and 1/2 in. diam) were pressed from a mixture of ceramic grade UO_2 and PuO_2 with an organic binder. The pellets were then sintered in hydrogen at about 1600 °C for 12 to 14 hr.

Dissolution Equipment

Fuel specimens were declad and/or dissolved in cylindrical dissolver pots (6 in. high and 4 in. diam) constructed of either 304-L stainless steel or Teflon. Each dissolver was provided with a stainless steel condenser and a press-fit Teflon top. The lower end of the condenser was press-fit into the Teflon top so both pieces of equipment could be removed as a unit to provide quick access to the pot interior. A 1 in. diam hole was bored in the Teflon top to accommodate, as desired, sample transfer pipets or a thermometer. This sample port

TABLE 1. PRTR $\text{PuO}_2\text{-UO}_2$ Fuel Samples Used in Dissolution Studies

Fuel No.	Irradiation, MWd/ton U	Nominal Initial Percent PuO_2	Fabrication Procedure ^(c)
1 ^(a)	None	0.48	Incremental Loaded-Swaged
5187 ^(b)	1972	0.48	Incremental Loaded-Vibrational Packed
5168 ^(b)	5000	0.48	Incremental Loaded-Swaged
2 ^(a)	None	0.48	Incremental Loaded-Vibrational Packed
3 ^(a)	None	1.00	Nupac-Swaged
5213 ^(b)	1068	1.00	Nupac-Swaged
4 ^(a)	None	1.00	Nupac-Vibrational Packed
5 ^(a)	None	0.05	Sintered Pellets

(a) For identification purpose only.

(b) Actual PRTR fuel element assembly number.

(c) See discussion under Fuel Description

was closed with a Teflon plug when not in use. Pots were heated by either a heating mantle or a hot plate.

Teflon end-caps were press-fitted to the ends of the zircaloy-clad fuel pieces in many decladding experiments. These caps prevented premature axial attack of the $\text{PuO}_2\text{-UO}_2$ core by the decladding solution.

Decladding and Dissolution Procedures PuO_2 Powder

The solvent of the desired composition and volume was added to the Teflon dissolver vessel. This solution was brought to and maintained at the desired temperature ($\pm 1^\circ\text{C}$) by manual regulation (Variac) of the voltage supplied to the heater. A weighed quantity of PuO_2 was then added, and dissolution was allowed to proceed for periods up to 24 hr. Small (0.2 to 0.5 ml) samples of the dissolver solution were taken periodically.

In all cases the amount of PuO_2 used was such that the final dissolver solution would contain 1.8 to 2.0 g/liter of plutonium if all the PuO_2 were dissolved.

$\text{PuO}_2\text{-UO}_2$ Fuels

Decladding Process

Zircaloy cladding was either mechanically removed or chemically removed by the Zirflex process.^(13,14) The procedure for the Zirflex process follows below. A weighed fuel section of measured length was charged to the dissolver. (In some instances Teflon end caps were attached). The desired volume of boiling 2.0 to 5.7M NH_4F -0.17 to 0.55M NH_4NO_3 solution was added, and the solution was boiled 2 to 5 hr. Initial decladding solution compositions and volumes were selected to yield either 0.10, 0.25, or 0.60M zirconium solutions, as may be seen in Table II. When dissolving to 0.6M zirconium,

TABLE II. Conversion of UO_2 to UF_4 During Zircaloy Decladding Step

Conditions: Zircaloy-clad PuO_2 - UO_2 fuels declad according to conditions listed. After removal of decladding solution core material was dissolved in boiling $6M$ HNO_3 - $0.1M$ $Al(NO_3)_3$ solution; the fluoride concentration of this solution was determined.

Fuel No.	End caps used (a)	Decladding conditions				mmoles UF_4 formed per gram of PuO_2 - UO_2 charged	Percent UO_2 converted to UF_4	Estimated core dissolver Sol'n fluoride concentration (d), M	Loss to decladding waste	
		NH_4F M	NH_4NO_3 M	Time (c) hr	Terminal Zr, M				Percent U	Percent Pu
1	No	2.0	0.17	2.0	0.13	0.68	18.3	0.31	-	0.089
1	Yes	2.0	0.17	2.0	0.11	0.088	2.3	0.040	-	0.20
1	Yes	2.0	0.17	2.0	0.26	0.093	2.5	0.042	-	0.32
1	Yes	2.0	0.17	5.0	0.12	0.41	11.0	0.19	-	0.077
3	No	2.0	0.17	5.0	0.12	0.57	15.5	0.26	-	-
3	Yes	2.0	0.17	5.0	0.25	0.34	8.9	0.16	-	0.41
3	Yes	2.0	0.17	5.0	0.25	0.24	6.4	0.11	-	-
5187	Yes	2.0	0.17	5.0	0.11	0.050	1.3	0.022	-	-
5213	Yes	2.0	0.17	5.0	0.11	0.042	1.7	0.028	-	-
1	No	4.0	0.34	2.0	0.24	3.8	~100	1.7	-	-
1	Yes	4.0	0.34	2.0	0.25	0.77	-	0.35	0.91	-
3	No	4.0	0.34	2.0	0.23	3.4	91.	1.5	-	-
3	Yes	4.0	0.34	2.0	0.21	0.53	14.	0.24	0.74	0.42
5187	No	4.0	0.34	2.0	0.22	1.7	46.	0.78	-	-
3	Yes	5.7	0.55	4.0	0.64	0.50	14.	0.76	0.38	-
5213	Yes	5.7	0.55	2.0	0.57	0.053	1.4	0.14	1.3	0.34
5168	Yes	5.7	0.55	3.0	0.50	0.12	3.3	0.17	0.24	0.10

(a) Numbers correspond to those used in Table I.

(b) Exposed core covered by Teflon end caps, see page 4.

(c) Time at boiling temperature.

(d) Calculated on basis that declad core is dissolved to yield either a $0.42M$ $UO_2(NO_3)_2$ solution or (last 3 experiments listed) a $1.4M$ $UO_2(NO_3)_2$ solution.

water was added periodically to make up for that lost by evaporation; otherwise, the decladding proceeded without interruption.

Upon completion of the decladding period, decladding wastes which were about 0.1 or $0.25M$ in zirconium were cooled 2 hr to about $25^\circ C$ and then removed from the dissolver pot. Decladding wastes which were about $0.6M$ zirconium were cooled to $50^\circ C$, diluted with water to about $0.38M$ zirconium to prevent precipitation of $(NH_4)_2ZrF_6$, and removed. Special pains were taken in every case to remove all the decladding solution and none of the PuO_2 - UO_2 core material. The volume and plutonium and uranium concentration of the decladding wastes were determined. Finally, if present, end caps were removed.

Dissolution Schemes

Three different core dissolution schemes were tested with declad PuO_2 - UO_2 fuels. These schemes are:

Scheme 1 - Dissolve 2 to 4 hr at $80^\circ C$ in $11.1M$ HNO_3 - $0.1M$ NH_4F solution to yield $0.42M$ $UO_2(NO_3)_2$ - $10M$ HNO_3 solution.

Scheme 2 - Step 1: Dissolve 2 to 4 hr in boiling $5.12M$ HNO_3 - 0.0 or $0.25M$ NH_4F - $0.75M$ $Al(NO_3)_3$ solution to yield a $0.42M$ $UO_2(NO_3)_2$ - $4M$ HNO_3 solution.

Step 2: Treat residue 4 hr at $80^\circ C$ with a $10M$ HNO_3 - $0.1M$ NH_4F solution.

Scheme 3 - Dissolve 3 to 7 hr in boiling $8.7M$ HNO_3 - $0.15M$ NH_4F solution to yield a $1.4M$ $UO_2(NO_3)_2$ - $5.0M$ HNO_3 solution.

Mechanically-declad PuO_2 - UO_2 fuels were also dissolved according to one

of the above schemes by the experimental technique used for PuO_2 powder when appropriate modifications were made. To dissolve chemically-declad fuels, the solvent was heated to the desired temperature and added to the dissolver vessel containing the PuO_2 - UO_2 fuel. In all cases dissolution was studied as a function of time; zero time was taken as the time when fuel and solvent were brought into contact. In each case the final dissolver solution was allowed to cool to 25 °C and was carefully removed from the dissolver vessel so as not to disturb any solids which might be present; then the volume and composition were determined. For material balance, residues, if any, were treated with 100 to 200 ml of boiling 11.1M HNO_3 -0.1M NH_4F for an hour.

Special Dissolution Schemes

Special dissolution schemes were employed to estimate (a) the amount of UF_4 formed during chemical decladding and left as a residue along with unreacted PuO_2 and UO_2 and (b) the fraction of plutonium in various PuO_2 - UO_2 fuels which was soluble in HNO_3 only. For the former purpose, chemically declad PuO_2 - UO_2 fuels were treated with sufficient boiling 6M HNO_3 -0.1M $\text{Al}(\text{NO}_3)_3$ solution to dissolve all the UO_2 and any UF_4 which might be present. The resulting solution was analyzed for fluoride content. Prior to dissolution of PuO_2 - UO_2 fuels in HNO_3 only, the dissolvers were scrupulously cleaned to ensure the absence of fluoride ion. Unirradiated fuels were mechanically declad; irradiated fuels were charged to the dissolver with Zircaloy cladding intact.

In all cases the core was exposed 2 hr to boiling 11.1M HNO_3 to yield a 10M HNO_3 -0.42M $\text{UO}_2(\text{NO}_3)_2$ solution. This dissolver solution was carefully removed and its plutonium content determined. Residual PuO_2 was then dissolved by treatment at 80 °C for 4 hr with a large volume of 12.7M HNO_3 -0.1M NH_4F .

Analyses

The amount of plutonium in dissolver solutions obtained from unirradiated fuels was determined by direct alpha counting of aliquots. Sequential triisooctylamine extractions⁽¹⁵⁾ were used to determine plutonium amounts in decladding solutions and also in dissolver solutions obtained from irradiated fuels. Alpha pulse height analyses were performed whenever required.

Uranium amounts in decladding and dissolver solutions were determined either by X-ray fluorescence techniques⁽¹⁶⁾ or by a spectrophotometric procedure.⁽¹⁷⁾ The distillation - volumetric procedure described by Kolthoff and Sandell⁽¹⁸⁾ was used for analysis of fluoride ion in various dissolver solutions. The amount of zirconium in dissolver solutions obtained in Redox plant dissolutions of irradiated PRTR PuO_2 - UO_2 fuels was determined by the trioctylphosphine oxide extraction procedure of Young and White.⁽¹⁹⁾

Plutonium and uranium isotopic contents in certain of the dissolver solutions produced from the irradiated fuels were established by mass spectrometric analysis. Samples for mass spectrometric analysis were purified by ion exchange resin techniques. Aliquots of these purified samples were

also used to establish factors for converting plutonium dis/min/liter values to g/liter values.

RESULTS

Zirflex Process Decladding of $\text{PuO}_2\text{-UO}_2$ Fuels

A wide range of Zirflex process conditions proved suitable for dissolving zircaloy cladding from irradiated, as well as unirradiated, $\text{PuO}_2\text{-UO}_2$ fuels (Table II). In particular, the decladding procedure proposed and subsequently used in the Hanford Redox plant multipurpose dissolver (Figure 1) and the procedure proposed (Figure 2) for use in a Purex plant service dissolver dissolved zircaloy cladding very efficiently.

When not fitted with Teflon end caps, unirradiated fuel pieces crumbled to mud during the decladding step.

Under these conditions, considerable conversion of the UO_2 core to UF_4 (and/or NH_4UF_5 ?) also occurred as evidenced by the green color of the declad residue and by analytical data for fluoride concentrations (Table II). French workers also noted⁽²⁰⁾ extensive attack of the UO_2 powder by Zirflex decladding solutions. Physical and chemical attack of the core material decreased appreciably when the exposed ends of the fuel pieces were covered with Teflon end caps and only zircaloy cladding was exposed to solution initially. This latter condition

FIGURE 1. Zirflex Process Decladding
Flowsheet for Use in Hanford Redox
Plant with PRTR $\text{PuO}_2\text{-UO}_2$ Fuels

FIGURE 2. Proposed Zirflex Process
Deccladding Flowsheet for Use in Service
Dissolver

corresponds to that which prevails in actual plant decladding operations.

Preferred dissolution procedures, to be discussed later, call for dissolution of the declad cores in HNO_3 - NH_4F solutions. These procedures require careful control of the fluoride concentration to ensure adequately low corrosion of the dissolver vessel. Any UF_4 formed in the decladding step will contribute to the fluoride concentration of dissolver solution. It is highly important, therefore, to remove zircaloy cladding under conditions which result in minimum production of UF_4 . Data in Table II indicate the recommended decladding flowsheets (Figures 1 and 2) provide such conditions, especially for irradiated fuel.

Losses of plutonium to the decladding solution were 1% or less and

were as low as 0.1% in some cases. Uranium losses were generally higher but were also less than 1% except in one case.

Dissolution of PuO_2 Powder in HNO_3 - NH_4F Solutions

Data in Tables III and IV summarize dissolution experience with PuO_2 powder. These results guided selection of dissolution flowsheet conditions tested in subsequent experiments with PuO_2 - UO_2 fuels.

The need to minimize corrosion of the stainless steel dissolver vessel was an overriding factor in selection of dissolution conditions for use in the Hanford Redox plant. Maness' data are pertinent in this connection⁽¹²⁾. Corrosivity of HNO_3 -fluoride solutions to 304-L stainless steel

TABLE III. Dissolution of PuO_2 in $\text{HNO}_3\text{-NH}_4\text{F}$ Solution at 60 and 80 °C

Conditions: 0.2 g PuO_2 dissolved in 100 ml of solutions of composition indicated. See text, page 4.

NH_4F	80 °C, 15.5M HNO_3			60 °C, 15.5M HNO_3			80 °C, 12.7M HNO_3			80 °C, 10M HNO_3			80 °C, 10M HNO_3 , $\text{UO}_2(\text{NO}_3)_2$ Present			
	0.1M	0.05M	0.025M	0.1M	0.05M	0.025M	0.1M	0.05M	0.025M	0.1M	0.05M	0.025M	0.1M(a)	0.1M(b)	0.1M(c)	0.1M(d)
Time Min	Run 1	Run 2	Run 3	Run 4	Run 5	Run 6	Run 7	Run 8	Run 9	Run 10	Run 11	Run 12	Run 13	Run 14	Run 15	Run 16
5	7.6	17.	10.	11.	13.	8.2	15.	12.	4.5	13.	8.5	4.5	14.	14.	11.	11.
10	13.	28.	27.	20.	18.	15.	30.	21.	9.0	24.	13.	8.2	26.	27.	20.	20.
15	22.	42.	35.	25.	28.	22.	45.	33.	20.	33.	19.	11.	44.	39.	28.	31.
20	59.	53.	39.	33.	39.	27.	59.	57.	26.	45.	26.	15.	55.	48.	36.	41.
25	70.	61.	45.	41.	47.	35.	69.	-	32.	53.	31.	20.	63.	56.	43.	52.
30	81.	63.	52.	45.	59.	44.	75.	64.	37.	60.	38.	24.	69.	64.	52.	60.
60	89.	82.	70.	86.	87.	70.	92.	91.	60.	85.	65.	41.	88.	87.	85.	87.
90	100.	100.	81.	96.	96.	82.	100.	100.	73.	93.	79.	53.	96.	96.	95.	94.
120			87.	100.	100.	90.			80.	97.	86.	61.	100.	100.	100.	98.
180			95.			96.			88.	100.	97.	74.				100.
300			100.			100.			96.		100.	85.				
420									100.			92.				

(a) 0.1M $\text{UO}_2(\text{NO}_3)_2$ (c) 0.6M $\text{UO}_2(\text{NO}_3)_2$
 (b) 0.4M $\text{UO}_2(\text{NO}_3)_2$ (d) 0.8M $\text{UO}_2(\text{NO}_3)_2$

9

TABLE IV. Dissolution of PuO_2 in Boiling $\text{HNO}_3\text{-NH}_4\text{F}$ Solutions

Conditions: 0.2 g PuO_2 dissolved in boiling solutions of composition indicated. See text page 4.

Time hrs	Percent Dissolved														
	0.1M NH_4F					0.15M NH_4F (a)			0.025M NH_4F						
	2.5M HNO_3 (a)	4.0M HNO_3 (a)	4.0M HNO_3 (b)	6.0M HNO_3 (a)	8.0M HNO_3 (a)	3.0M HNO_3	4.0M HNO_3	5.0M HNO_3	2.5M HNO_3 (a)	4.0M HNO_3 (a)	4.0M HNO_3 (c)	10M HNO_3 (c)	10M HNO_3 (d)	10M HNO_3 (e)	10M HNO_3 (f)
0.5	8.2	48.	-	78.	-	28.	71.	82.	56.	93.	-	-	-	-	-
1.0	14.	65.	55.	91.	100.	38.	76.	89.	74.	98.	100.	100.	57.	2.9	1.6
2.0	23.	77.	72.	98.		54.	81.	95.	82.	100.			68.	4.5	1.8
3.0	31.	84.	80.	100.		69.	86.	99.	86.				73.	6.3	2.3
4.0	38.	90.	84.			81.	91.	100.	89.				77.	7.5	2.5
5.0	45.	92.	86.			88.	94.		92.				80.	8.5	2.6
6.0	48.	94.				89.	97.		93.				82.	9.3	3.3
7.0	51.					90.	100.		94.				-	-	-
20.0	-								-				-	26.	-
22.0	70.								98.				-	-	-
24.0													86.	30.	9.2

(a) Solutions were also 1.4M $\text{UO}_2(\text{NO}_3)_2$ (d) Solutions were also 0.25M $\text{Al}(\text{NO}_3)_3$
 (b) Solutions were also 0.05M $\text{Al}(\text{NO}_3)_3$ (e) Solutions were also 0.50M $\text{Al}(\text{NO}_3)_3$
 (c) Solutions were also 0.125M $\text{Al}(\text{NO}_3)_3$ (f) Solutions were also 0.75M $\text{Al}(\text{NO}_3)_3$

BNWL-204 PT2

varies directly with HNO_3 and fluoride concentrations and is much lower at 60 to 80 °C than at boiling temperature. For example, in boiling 10M HNO_3 -0.1M NH_4F solution, 304-L stainless steel corrodes at a rate greater than 0.21 mil/hr while at 80 °C the rate is only about 0.058 mil/hr, and at 60 °C only about 0.020 mil/hr.

Data in Table III show PuO_2 powder dissolved completely in about 3 hr in 10M HNO_3 -0.1M NH_4F at 80 °C. This dissolution time was not significantly altered when the solvent contained up to 0.8M $\text{UO}_2(\text{NO}_3)_2$.

A dissolver constructed of a material (e.g., HAPO 20* or Ni-O-Nel) resistant to boiling HNO_3 -fluoride solutions is desirable for repeated reprocessing of PuO_2 - UO_2 fuels. Such a service dissolver has been proposed for installation in the Hanford Purex plant.⁽²²⁾ Even in such a dissolver it is desirable

to restrict the fluoride concentration to 0.25M or less.⁽¹²⁾ Data in Table IV demonstrate PuO_2 powder dissolves rapidly in boiling 4 to 6M HNO_3 -0.1 to 0.25M NH_4F -1.4M $\text{UO}_2(\text{NO}_3)_2$ solutions.

Data in Table IV substantiate the statement made earlier; namely, the dissolution rate of PuO_2 in boiling HNO_3 - NH_4F - $\text{Al}(\text{NO}_3)_3$ solution is markedly reduced when the $\text{Al}(\text{NO}_3)_3$ concentration is equal to or greater than the fluoride concentration.

Dissolution of PRTR PuO_2 - UO_2 Fuels in HNO_3 - NH_4F Solutions

Three different schemes for dissolving PRTR PuO_2 - UO_2 fuels were demonstrated. Two of these (Tables V and VI) appear suitable for at least limited use in stainless steel equipment (Redox plant dissolver). The third (Table VII and Figure 3) employs a boiling HNO_3 - NH_4F solution for use in a vessel suitably resistant to such a solution.

The dissolution scheme (Table V and Figure 4) believed most suitable

* HAPO 20 is the designation of an experimental alloy (see reference 21) of the approximate composition 50 wt% Ni-25 wt% Cr-6 wt% Mo-16 wt% Fe-1 wt% Ti-1 wt% Cu-0.6 wt% Mn-0.4 wt% Si.

TABLE V. Dissolution of UO_2 - PuO_2 Fuels by Scheme 1

Dissolution Time, min	Fuel No (a)	Percent Dissolved											
		1(b)		5187(c,e)		2(b)		3(b)		3(c)		5213(c,f)	
		Pu	U	Pu	U	Pu	U	Pu	U	Pu	U	Pu	U
5		82.	81.	53.	35.	90.	29.	61.	60.	44.	-	55.	-
10		82.	100.	81.	57.	94.	38.	-	80.	53.	-	71.	-
15		-	-	93.	68.	95.	57.	66.	99.	61.	-	-	-
30		88.	-	98.	95.	96.	-	75.	-	75.	-	73.	-
60		95.	-	-	100.	100.	75.	85.	100.	88.	-	84.	-
120		100.	-	99.	-	-	-	96.	-	95.	-	93.	-
180		-	-	-	-	-	-	-	-	-	-	-	-
240		-	-	-	-	-	-	100.	-	100.	-	-	-
300		-	-	-	-	-	-	-	-	-	-	-	-
360		-	-	-	-	100.	-	-	-	-	-	-	100.

(a) Numbers correspond to those used in Table I.

(b) Mechanically deoxid.

(c) Chemically deoxid-5 hours boiling 2.0M NH_4F -0.17M NH_4NO_3 to 0.10M Zirconium.

(d) Initial dissolvent contained no NH_4F . Final dissolver solution was 0.18M fluoride.

(e) Final dissolver solution was 0.102M fluoride.

(f) Final dissolver solution was 0.118M fluoride.

TABLE VI. Dissolution of UO_2 - PuO_2 Fuels by Scheme 2

Dissolution Time Min.	Fuel No. (a)	Percent Dissolved													
		1(b)		5187(c,d)		2(b)		3(b)		3(c,d)		5213(c,d)		4(b)	
		Pu	U	Pu	U	Pu	U	Pu	U	Pu	U	Pu	U	Pu	U
Step 1															
120		4.3	-	56.	100.	-	-	2.6	94.	7.5	84.	55.	100.	2.2	-
240		3.7	-	52.	100.	-	-	1.8	99.	-	-	55.	100.	3.2	-
Step 2 (e)															
5		-	-	71.	-	-	-	-	-	-	-	39.	-	-	-
15		-	-	83.	-	-	-	-	-	-	-	59.	-	-	-
30		95.	-	95.	-	90.	-	70.	-	67.	-	71.	-	89.	-
60		99.	-	100.	-	99.	-	84.	-	83.	-	86.	-	100.	-
90		99.	-	-	-	100.	-	97.	-	91.	-	-	-	-	-
120		100.	-	-	-	-	-	98.	-	97.	-	85.	-	-	-
150		-	-	-	-	-	-	100.	-	-	-	-	-	-	-
180		-	-	-	-	-	-	-	-	100.	-	-	-	-	-
240		-	-	-	-	-	-	-	-	-	-	100.	-	-	-

(a) Numbers correspond to those used in Table I

(b) Mechanically de-clad

(c) Chemically de-clad 2 hr boiling 4.0M NH_4F -0.55M NH_4NO_3 to 0.10M zirconium

(d) Step 1 dissolvent contained no fluoride

(e) Percentages refer to amounts of plutonium dissolved of that left undissolved from Step 1

TABLE VII. Dissolution of UO_2 - PuO_2 Fuels by Scheme 3.

Dissolution Time, Min	Fuel No. (a) =	Percent Dissolved									
		1(b)		4(b)		5168(c,d)		5213(c,d)		5213(c,e)	
		Pu	U	Pu	U	Pu	U	Pu	U	Pu	U
30		94.	100	100	100	100	100	97	100	74.	100
60		96.	-	-	-	-	-	100	-	88.	-
120		100.	-	-	-	-	-	-	-	-	-
300		-	-	-	-	-	-	-	-	90.	-
420		-	-	-	-	-	-	-	-	95.	-

(a) Numbers correspond to those used in Table I.

(b) Mechanically de-clad.

(c) Chemically de-clad-2 hr boiling 5.7M NH_4F -0.55M NH_4NO_3 to 0.60M zirconium.

(d) Dissolver rinsed thoroughly between de-cladding and dissolution steps.

(e) Dissolver not rinsed after de-cladding step and fluoride not added to dissolvent.

for use in stainless steel equipment is a single-step process in which both PuO_2 and UO_2 are dissolved at 80 °C in a 11M HNO_3 -0.1M fluoride solution. This scheme assumes only minimal formation of UF_4 during the de-cladding step; this assumption appears well justified for irradiated fuels (see the data in Tables II and V).

The alternate scheme shown in Table VI is a two-step dissolution process. In the first step, all of the UO_2 and UF_4 which might be present from

the de-cladding step, and that fraction of plutonium soluble in HNO_3 (see page 13) are dissolved in a boiling HNO_3 -Al(NO_3)₃ solution. Residual PuO_2 is then dissolved at 80 °C in a 10M HNO_3 -0.1M NH_4F solution. This scheme is designed to guard against excessive corrosion of the dissolver vessel which might occur because of the formation of large amounts of UF_4 during the de-cladding step. (In some of the experiments listed in Table VI, the dissolvent contained 0.25M NH_4F to

FIGURE 3. Proposed Core Dissolution Flowsheet for Use in Service Dissolver

(a) It is estimated UF₄ formed during de-cladding will contribute 0.025 moles fluoride per liter of dissolvent.

FIGURE 4. Core Dissolution Flowsheet for Use in Hanford Redox Plant with PRTR PuO₂-UO₂ Fuels

simulate a concentration of fluoride ion which would result from dissolution of a large amount of UF_4). The major disadvantage to this scheme is the requirement of a precise separation of liquid and solid phases between the two dissolution steps. (In Redox plant equipment where a centrifuge to retain PuO_2 fines is not available, this disadvantage was considered overwhelming.)

Several points about the data listed in Tables V and VI merit comment. Firstly, PuO_2 in swaged, Nupac fuel dissolved more slowly at 80 °C than did PuO_2 in either swaged or vibrationally packed incremental-loaded fuel. Also, UO_2 in vibrationally packed fuels dissolved more slowly than did UO_2 in swaged fuel. Finally, fluoride concentrations (Table V) in dissolver solutions obtained from chemically declad fuels were in excellent agreement with those expected from the results in Table II.

In two of the experiments with irradiated fuel listed in Table VII the dissolver vessel was rinsed thoroughly with water and HNO_3 after the decladding step. The declad core was then dissolved completely in 30 to 60 min. in a dissolvent containing 0.15M NH_4F . In the third experiment the dissolver was not rinsed, and NH_4F was not added to the core dissolvent. The PuO_2 did not completely dissolve even after 7 hr boiling in this latter experiment. Apparently, in this experiment, slightly less UF_4 formed in the decladding step than found in the corresponding experiments in Table II.

Dissolution of PRTR PuO_2 - UO_2 Fuels in HNO_3

Unirradiated, sintered (12 to 14 hr in H_2 at 1600 °C) UO_2 -0.45 wt% PuO_2 pellets were almost completely soluble in HNO_3 (Table VIII). Such sintering produces a solid solution of UO_2 and PuO_2 (mixed oxide) which does not require the presence of fluoride for dissolution in HNO_3 .

Plutonium dioxide in unirradiated oxide mixtures (Fuels 1 and 3), however, was only sparsely soluble in HNO_3 in agreement with the results of many previous investigators. Data in Table VIII suggests the pneumatic densification technique used to prepare the Nupac fuel converts a small amount of oxide mixture to a solid solution.

Data in Table VIII also demonstrate significant in-reactor conversion of oxide mixtures to solid solutions occurs during irradiation of PRTR PuO_2 - UO_2 fuels. The degree of conversion increases with exposure level reflecting, probably, the effects of longer times at elevated temperature. One datum indicates essentially complete conversion to a mixed oxide occurs upon sufficient reactor irradiation (~5000 MWd/ton U).

In-reactor conversion of oxide mixtures of PuO_2 and UO_2 to solid solutions is, of course, a highly interesting phenomenon; and more data are needed to establish under what irradiation conditions this conversion occurs for various percentages of PuO_2 and UO_2 . Its occurrence, however, does not eliminate the need for a dissolver

TABLE VIII. Dissolution of PRTR $\text{PuO}_2\text{-UO}_2$ Fuels in HNO_3

Conditions: 2 hr in boiling 11M HNO_3

Fuel No. (a)	Fabrication Procedure (b)	Irradiation Mwd/ton U	Plutonium Soluble in HNO_3 , %
1	Incremental Loaded-Swaged	None	0.62
3	Nupac - Swaged	None	6.2
5	Sintered Pellets	None	96.1
5213	Nupac - Swaged	1068	60.5 (c)
5187	Incremental Loaded-Vibrational Packed	1972	74.5 (d)
5168	Incremental Loaded - Swaged	5000	99.0 (e)

(a) Numbers correspond to those used in Table I.

(b) See discussion of Fuel Description

(c) Average of triplicate determinations.

(d) Average of duplicate determinations.

(e) Single determination.

vessel resistant to boiling HNO_3 -fluoride solutions for routine reprocessing of $\text{PuO}_2\text{-UO}_2$. In handling such fuels, unirradiated fuel scrap as well as elements which, for one reason or another, have not been irradiated long enough to effect complete conversion will also have to be reprocessed. By judicious selection of dissolver charges, however, it might be possible to eliminate fluoride in reprocessing at least part of the total load and thereby reduce overall dissolver corrosion.

CHEMICAL FLOWSHEETS

The decladding and core dissolution flowsheets proposed by Hanson⁽²³⁾ for use in the Hanford Redox plant are shown in Figures 1 and 4 respectively. The core dissolution conditions are

essentially those referred to as Scheme 1 in this report.

Chemical flowsheets proposed by Ritter⁽²²⁾ for use in a service dissolver in the Hanford Purex plant are shown in Figures 2 and 3. Results obtained in this study (Tables II and VII) demonstrated the utility of these flowsheets.

REDOX PLANT DISSOLUTION EXPERIENCE

Table IX summarizes recent Hanford Redox plant experience in decladding and dissolving three charges (29 assemblies) of irradiated PRTR $\text{UO}_2\text{-0.48 wt% PuO}_2$ fuel. Decladding was accomplished with excellent results. Core dissolution according to the flowsheet shown in Figure 4 was only partially successful, however. Complete dissolution of UO_2 and PuO_2 was realized in each case. However, dissolver solution fluoride concentrations were much higher than

TABLE IX. Summary of Dissolution of PRTR UO_2 -0.48 wt% PuO_2 in the Redox Plant

	Charge 1	Charge 2	Charge 3
No. Assemblies	10.5	11	7.5
Uranium, lb	1100	1140	800
Plutonium, g	2330	2500	1730
Irradiation History			
Minimum MWd/ton U	290	112	390
Maximum MWd/ton U	11,400	5670	6400
Average MWd/ton U	3110	3720	4950
Zirflex Process Decladding ^(a)			
Time at Boiling			
Temperature, hr	8	8	8.75
Percent U Loss	0.72	0.44	0.50
Percent Pu Loss	0.43	0.24	0.46
Fuel Dissolution ^(b)			
Total time at 60 to 80 °C, hr	9.0	6.67	9.25
Time to dissolve 100% Pu and U ^(c) , hrs	1.4	4.0 - 6.67	~9.25
Terminal Composition, M			
HNO ₃	8.72	8.74	7.35
UO ₂ (NO ₃) ₂	0.61	0.57	0.49
Fluorine ²	0.271	0.36	0.77
Zirconium	0.036	0.029	0.044

(a) Conditions were those given in Figure 1.

(b) Conditions were those given in Figure 4 except fluoride was not added to initial dissolvent.

(c) Estimated from analyses of samples taken during dissolution.

the anticipated and desired 0.1M even though fluoride was not added to the initial dissolvent.

The source of the large amounts of fluoride is in doubt. One explanation, of course, is that conversion of UO_2 to UF_4 during decladding occurred to an extent greater than anticipated from laboratory studies. However, judging from the presence of zirconium in the dissolver solutions, a large

fraction of the fluoride appears to have originated by carryover of a heel of decladding waste solution. Redox plant personnel estimate as much as 25 to 50 gal of decladding waste could have been carried over to the core dissolution step.

It is also probable that at least some zirconium and fluoride were carried over to the core dissolution step as the compound $(NH_4)_3ZrF_7$. This

compound is reported⁽²⁰⁾ to precipitate when decladding wastes, in which the F:Zr mole ratio is greater than 7, are cooled to room temperature. The F:Zr ratio in decladding waste produced according to the flowsheet condition shown in Figure 1 is 16. In a laboratory test of these flowsheet conditions, white solids - presumably $(\text{NH}_4)_3\text{ZrF}_7$ - did precipitate when the solution was cooled to 25 °C. Because of the absence of a centrifuge, plant decladding wastes were also cooled to 25 °C before jetting to allow fines (UO_2 and UF_4) to settle.

ACKNOWLEDGEMENT

Grateful acknowledgement is made to G. L. Hanson and G. L. Ritter of Isochem Inc. for permission to reproduce certain chemical flowsheets. Thanks are due, also, to G. L. Hanson for his resume' of plant dissolution experience. The cooperation of members of the Ceramics Research and Development Reactor and Fuels Laboratory, Battelle-Northwest, and of L. D. Turner, Programming Section, Battelle-Northwest, in providing samples of mixed oxide fuels is also acknowledged.

REFERENCES

1. Staff of Ceramics Research and Development Operation. Specifications for Vibrationally Compacted, Mixed Oxide (UO_2 - PuO_2), Fuel Elements for the PRTR (Mark 1-L), HW-79291. General Electric Company, Richland, Washington, October, 1963.
2. Staff of Ceramics Research and Development Operation. Specifications for Swage Compacted, Mixed Oxide (UO_2 - PuO_2), Fuel Elements for the PRTR (Mark 1-M), HW-79290. General Electric Company, Richland, Washington, October, 1963.
3. H. J. Anderson and R. J. Anicetti. Specifications for High Density UO_2 (Nuclear Grade), HW-74204 REV 1. General Electric Company, Richland, Washington, September, 1963.
4. H. J. Anderson. Specification for Nuclear Grade PuO_2 , Sinterable Powder, HW-76566 REV 1. General Electric Company, Richland, Washington, September, 1963.
5. H. J. Anderson and J. P. Wisely. Specification for Nuclear Grade Mixed Oxides (UO_2 - PuO_2), Compactable Powder, HW-78878. General Electric Company, Richland, Washington, September, 1963.
6. A. S. Wilson. Uranium Dioxide-Plutonium Dioxide Ceramics, HW-55319. General Electric Company, Richland, Washington, March 1958.
7. A. L. Uriarte and R. H. Rainey. Dissolution of High-Density UO_2 , PuO_2 , and UO_2 - PuO_2 Pellets in Inorganic Acids, ORNL-3695. Oak Ridge National Laboratory, April, 1965.
8. T. D. Chikalla. Sintering Studies on the System UO_2 - PuO_2 , Part 1 - Solid Solution Formation, HW-60274. General Electric Company, Richland, Washington, April, 1959.
9. W. S. Gelman. A Review of the Dissolution of Plutonium Dioxide, MLM-1264. Mound Laboratory, May, 1965.
10. Dissolution of Plutonium Dioxide in Mixtures of Nitric and Hydrofluoric Acid, EURAEC-708. European Atomic Energy Commission, December, 1963.
11. J. M. Cleveland. J. Inorg. Nucl. Chem. vol. 26, p. 1470. 1964.
12. R. F. Maness. Corrosion of 304-L Stainless Steel and HAP0-20 in Acid Fluoride Systems, HW-80658. General Electric Company, Richland, Washington, January, 1964.
13. J. L. Swanson. "The Zirflex Process," Proceedings of Second U. N. International Conference on Peaceful Uses of Atomic Energy. p. 2429, vol. 17, p. 154. Geneva, 1958.

14. P. W. Smith. The Zirflex Process - Terminal Development Report, HW-65979. General Electric Company, Richland, Washington, August, 1960.
15. R. A. Schneider and K. M. Harmon, editors. Analytical Technical Manual, HW-53368. General Electric Company, Richland, Washington, 1961.
16. R. J. Kofoed, Jr., "Uranium and the Transuranium Elements Analysis by X-Ray Fluorescence Spectrometry-Hanford Techniques" Encyclopedia of X-Rays and Gamma Rays, G. L. Clark, editor p. 1108-1110. Reinhold, New York, 1963.
17. R. J. Baltisberger. Anal. Chem. vol. 36, pp. 2369-2370. 1964.
18. I. N. Kolthoff and E. B. Sandell. Textbook of Quantitative Inorganic Analysis, 3rd ed. p. 721-723. Macmillan, New York, 1952.
19. J. P. Young and J. C. White. Talanta, vol. 1, pp. 263-273. 1958.
20. Y. Marchant and J. Van Caeneghem. Citriflex: Decladding of Fuel Elements Clad with Zirconium or Zircaloy-2 by an Ammonium Fluoride-Citric Acid Solution, NP-15661 (ETR-170). Eurochemic, Mol, Belgium. 1965.
21. R. F. Maness, et al. Ind. & Eng. Chem., Product Research & Develop. Quarterly. vol. 2, pp. 163-167. 1963.
22. G. L. Ritter. Personal Communication, Isochem, Inc., Richland, Washington.
23. G. L. Hanson. Personal Communication, Isochem, Inc., Richland, Washington.

DISTRIBUTION

<u>Number of Copies</u>		<u>Number of Copies</u>	
184	<u>Division of Technical Information Extension</u>	17	<u>Battelle-Northwest</u>
10	<u>Isochem Inc., Richland</u>		J. M. Atwood
	O. F. Beaulieu		A. G. Blasewitz
	R. D. Ehrlich		R. E. Burns
	J. B. Fecht		K. M. Harmon
	G. L. Hanson		A. M. Platt
	O. F. Hill		W. W. Schulz
	J. B. Kendall		L. D. Turner
	T. R. McKenzie		E. E. Voiland
	H. C. Rathvon		M. T. Walling
	G. L. Ritter		Technical Information
	R. E. Tomlinson		Files (5)
			Technical Publications (2)
			G. M. Dalen
2	<u>Richland Operations Office</u>		
	R. K. Sharp		
	Technical Information		
	Library		