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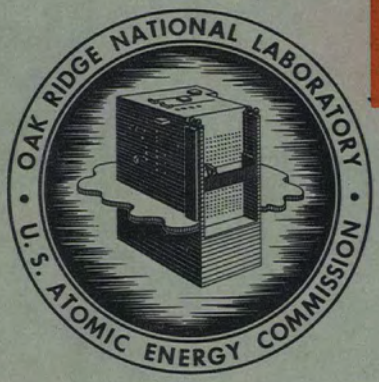
DECLADDING OF PWR BLANKET FUEL ELEMENTS WITH AQUEOUS AMMONIUM FLUORIDE SOLUTIONS

L. M. Ferris



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Chemical Development Section B

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L. M. Ferris

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ABSTRACT

A flowsheet is presented for decladding PWR blanket fuel elements with boiling 6 M NH_4F--1.0 M NH_4NO_3. After decladding, the pelleted UO_2 core is dissolved in 10 M HNO_3 to produce a solution suitable for recovery of uranium by solvent extraction. A 30-mil Zircaloy-2 jacket can be completely dissolved in about 2 hr with a uranium loss of less than 0.02%. The total uranium loss after decladding and core dissolution was less than 0.022%. All experiments were performed with unirradiated fuel specimens. Ammonium nitrate was added to the decladding solution to dissolve the tin in Zircaloy-2 and thus prevent excessively high uranium losses (~0.8%) to a nitric acid--insoluble residue comprised mainly of SnO_2.

Another dissolvent for tin, cupric chloride, caused higher uranium losses (0.13%) to the dejacketing solution.

CONTENTS

1.0 INTRODUCTION

Several methods for decladding zirconium- or stainless steel-clad fuels are being evaluated as part of the Power Reactor Processing Program at ORNL. This report is a summary of recent experiments on the development of a flowsheet for the decladding of zirconium-clad oxide fuels with aqueous ammonium fluoride. Methods which appear feasible for decladding this type of fuel include the gas-phase Zircex process,[1] which utilizes anhydrous HCl, and decladding with aqueous HF.[2] Each of these methods has serious disadvantages, so that work on alternative processes is warranted. Included here are the results of several laboratory flowsheet demonstrations with unirradiated PWR blanket rods (Zircaloy-2 clad, pelleted UO_2 core). These results will serve as a basis for further work with fuel irradiated to 5000-10,000 Mwd/ton. During the tests with irradiated fuel, the effect of irradiation on uranium losses will be evaluated.

The use of aqueous ammonium fluoride as a dejacketing agent has been proposed by Hanford workers[3,4] and others.[5] At Hanford, the process was named "Zirflex." One of their more important findings is that stainless steel is but slightly corroded by ammonium fluoride. With no ammonium nitrate present, the dissolution reaction

$$Zr + 6NH_4F \longrightarrow ZrF_6^{=} + 2NH_4^{+} + 4NH_3 + 2H_2$$

proceeds at a high rate. Dissolution conditions are optimum when the NH_4F concentration is 6 M.

The author is indebted to J. F. Land who performed the experiments. Chemical and x-ray analyses were provided by the groups of G. R. Wilson and R. L. Sherman of the ORNL Analytical Chemistry Division.

2.0 FLOWSHEETS

Aqueous ammonium fluoride by itself is unsuitable as a decladding reagent because of its failure to dissolve tin. If metallic tin remains after decladding, uranium losses in the subsequent core dissolution can be excessively high (Sec. 5.3). However, small amounts of ammonium nitrate or cupric chloride may be added to dissolve the tin (Sects. 4.2 and 4.3).

The main steps in the tentative flowsheet (Fig. 2.1) are (1) removal of the 30-mil Zircaloy-2 clad with a boiling solution of 6 M NH_4F-1.0 M NH_4NO_3 (the uranium lost during decladding should be less than 0.02%); (2) washing to remove most of the fluoride from the dissolver vessel; and (3) dissolution of the core (UO_2) in twice the stoichiometric amount of 10 M HNO_3 to obtain a suitable solvent extraction feed solution. The small amount of residual insoluble material in this step results in a uranium loss of ~0.002%.

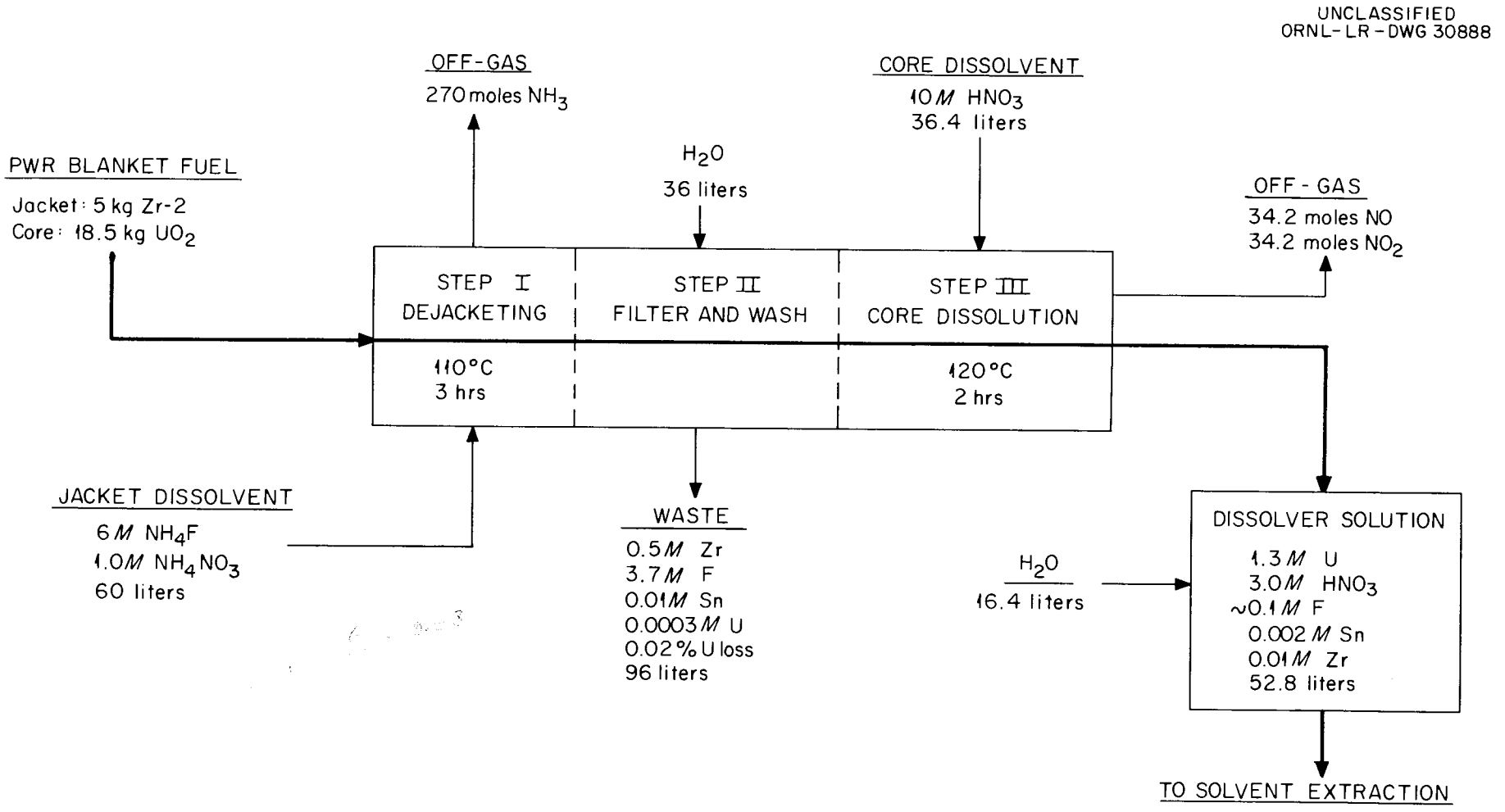

Fig. 2.1. Tentative flowsheet for dejacketing Zircaloy-2 jacketed fuels with aqueous NH_4F.

The flowsheet is based on the amount of uranium present before irradiation in one PWR blanket subassembly (120 fuel rods). The chemical composition of the subassembly was taken from a compilation by Ullmann.[6] It was assumed that Zircaloy-2 contains 1.5 wt % tin.[7] Other assumptions were that the jacket thickness was 30 mils, and that the final F/Zr mole ratio in the dejacketing solution should be 6.5. The amount of ammonium fluoride required for dejacketing was computed from the total quantity of zirconium present in the fuel, including end caps. The time required for complete dejacketing was estimated from rate data of Swanson.[3] The UO_2 core dissolves in nitric acid according to the equation

$$2UO_2 + 6HNO_3 \longrightarrow 2UO_2(NO_3)_2 + NO + NO_2 + 3H_2O.$$

3.0 EXPERIMENTAL PROCEDURE

All experiments were performed with unirradiated PWR blanket rods in the apparatus shown schematically in Fig. 3.1. On this scale, 500 ml of the 6 M NH_4F decladding solution was required per rod. The amount of uranium and zirconium in each rod was about 136 and 42 g, respectively.

The following general procedure was followed:

1. One PWR rod and 500 ml of the decladding solution were placed in the dissolver vessel, and the system was heated under reflux to 95-110°C, where decladding was complete in 2-3.5 hr.

2. The hot dejacketing solution was filtered off, leaving the UO_2 pellets, some insoluble tin, and the Zircaloy-2 end caps as a residue.

3. The UO_2 pellets were thoroughly washed with about 6 successive 65 ml portions of cold water, the wash water being used to dilute the decladding solution to prevent precipitation of such compounds as $(NH_4)_3ZrF_7$.

4. The end caps, tin and UO_2 were refluxed with 10 M HNO_3 for 1.5 hr.

5. The uranyl nitrate solution was passed through a filter, leaving the end caps and, in some cases, an insoluble residue.

4.0 RESULTS

4.1 Decladding with 6 M NH_4F

A total of six flowsheet demonstration experiments were performed using 6 M NH_4F as the decladding reagent. In each case decladding was

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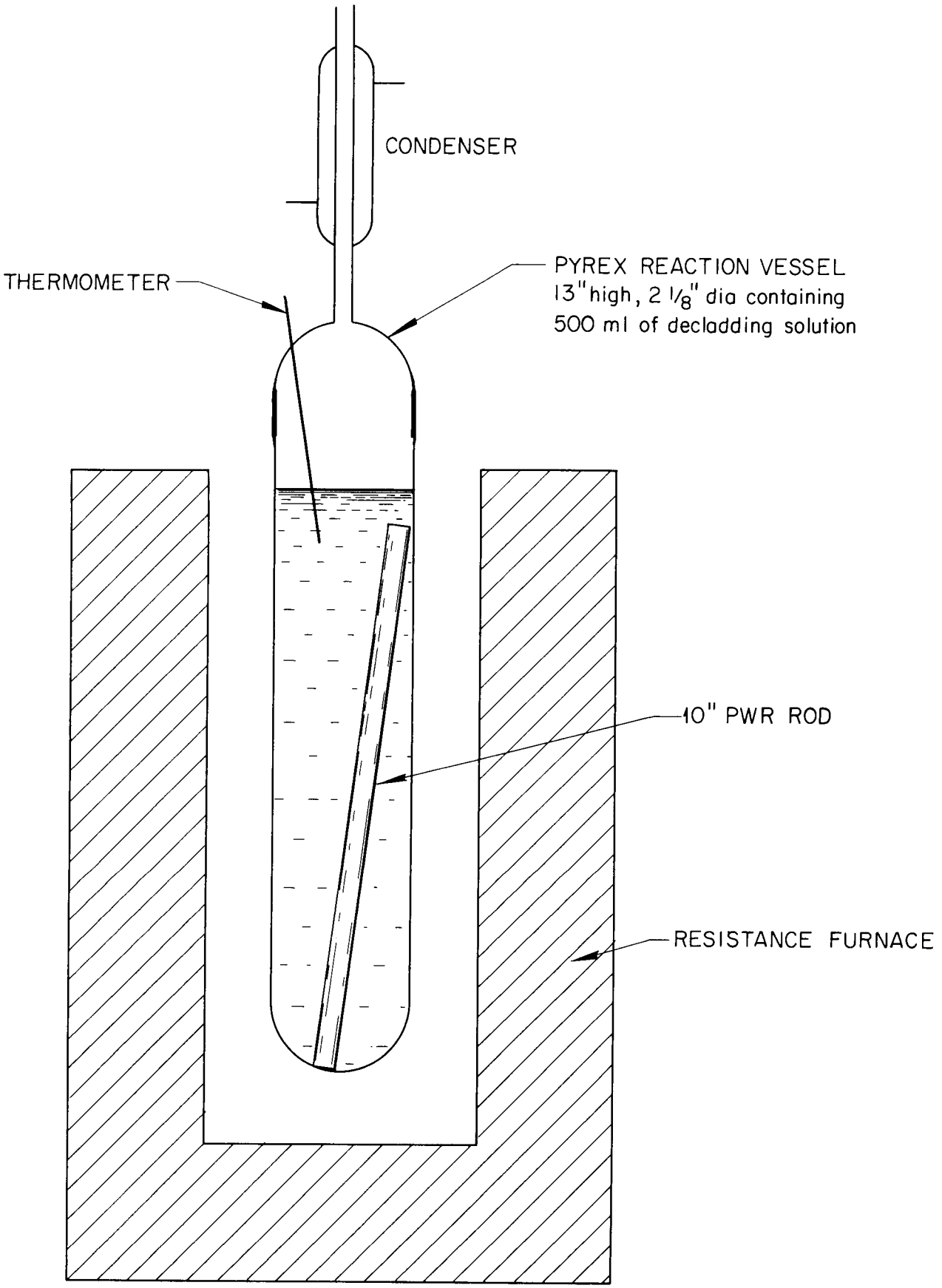

Fig. 3.1. Apparatus used for decladding PWR blanket rods with aqueous NH_4F.

complete in 3 to 3.5 hrs at 95-100°C. Complete dissolution of the UO_2 core in boiling 10 M HNO_3 was effected in 1.5 hrs. In all cases the uranium loss to the dejacketing solution was less than 0.002% (Table 4.1). However, losses to a nitric acid insoluble residue were excessively high (0.6-0.8%) in runs 3 and 4. These high losses are caused by formation, and subsequent dehydration, of solid metastannic acid during the core dissolution which occludes uranium oxide particles (Sec. 5.3).

Material balances for all experiments are given in Table 4.2.

4.2 Decladding with NH_4F-NH_4NO_3

Two experiments were performed with 6 M NH_4F-1.0 M NH_4NO_3 as the decladding reagent. Total uranium losses were 0.022% or less (Table 4.3). The uranium loss to the decladding solution (~0.02%) was higher than in the absence of nitrate, but is still quite acceptable. Uranium losses to the nitric acid--insoluble residue were less than 0.002%. Less nitric acid--insoluble residue was formed when ammonium nitrate was present in the decladding solution (cf. data in Tables 4.1 and 4.3). This is good evidence that nitrate ion oxidizes tin to a soluble species.

The average compositions of the various solutions are given in Table 4.4.

4.3 Decladding with NH_4F-$CuCl_2$

Several experiments were performed in which cupric ion, added as $CuCl_2$, was used as an oxidant for the dissolution of tin. The assumed reaction is

$$Sn + Cu^{++} \longrightarrow Sn^{++} + Cu.$$

In most cases the amount of nitric acid insoluble residue was very small and, in all cases, total uranium losses were less than 0.13% (Table 4.3). Since cupric ion also oxidizes zirconium in this system, it is desirable to add this reagent after the zirconium cladding has been essentially removed with NH_4F. However, an unidentified precipitate forms, which may contain up to 5% fluoride if the cupric chloride reagent is added rapidly, or if the solution is suddenly cooled. This precipitate is completely soluble in nitric acid, but, if present during core dissolution, would introduce undesired fluoride ion into the nitrate system.

The average compositions of the various solutions are included in Table 4.4.

Table 4.1. Dissolution of Prototype PWR Blanket Rods

Jacket Dissolvent: 6 M NH_4F

Core Dissolvent: 10 M HNO_3

Basis: 1 PWR rod containing ~136 g of uranium and ~45 g of Zircaloy-2

Exp. No.	Decladding time, hr	Temp., °C	Wt of HNO_3-insoluble Residue, g	Uranium Loss, % To Decladding Solution	Uranium Loss, % To HNO_3-insoluble Residue	Uranium Loss, % Total
1	3	90-95	---	0.00015	0.035	0.035
2	3.75	90-100	0.854	0.0011	0.0012	0.0023
3	3.5	90-100	1.612	0.001	0.61	0.611
4	3.5	90-100	1.783	0.0006	0.76	0.76
5	3.75	90-100	0.693	0.00056	0.0038	0.0044
6	---	90-100	0.689	0.002	0.011	0.013

Table 4.2. Material Balances for all Experiments

Basis: 1 PWR rod containing 42 g of Zr, 136 g of U, and 0.6 g of Sn; solution containing 53 g of F^-

Exp. No.	Decladding Reagent	Material Balance, % Zr	U	Sn	F	Cu
1	6 M NH_4F	---	---	---	---	---
2		116	98.0	98.3	92.8	---
3		114	98.0	66.7	85.6	---
4		117	102	76.7	84.5	---
5		125	96.1	80	87.1	---
6		119	98.0	113	105	---
7	6 M NH_4F-HCl wash	109	97.0	109	81	---
8	6 M NH_4F-1.0 M NH_4NO_3	136	97.3	226	83	---
9	6 M NH_4F-0.15 M $CuCl_2$	109	94.5	174	117	74.6
10	6 M NH_4F-0.1 M $CuCl_2$	118	98.0	109	63	53.7
11		117	96.7	205	107	66.2
12		126	96.9	171	103	187
13		119	98.2	113	103	120
14	6 M NH_4F-1.0 M NH_4NO_3	110	95.6	102	91.4	---
15		123	96.6	95.2	95.4	---
	Avg.	118	97.4	124	92.8	100

Table 4.3. Uranium Losses in Decladding

Temperature: ~ 110°C

Exp. No.	Decladding Reagent	Decladding time, hr	Wt of HNO_3-insoluble Residue, g	Uranium Loss, %		
				To Decladding Solution	To HNO_3-insoluble Residue	Total
8	6 M NH_4F-1.0 M NH_4NO_3	4	0.238	0.021	0.0015	0.022
14		2	0.089	0.0006	0.0025	0.0031
15		2	0.103	0.0005	0.0006	0.0011
9	6 M NH_4F-0.1 M $CuCl_2$	~ 2.5	0.121	0.0023	0.1 max*	---
10		3	0.734	0.0015	0.012	0.014
11		3	0.012	0.086	0.009 max*	0.09
12		3.5	0.021	0.032	0.015 max*	---
13		---	0.028	0.135	0.021 max*	~0.13

*In these cases the solid residue was not analyzed. The U loss recorded is that calculated assuming the residue was pure U, and therefore represents the maximum loss.

Table 4.4. Average Composition of Process Solutions

Exp. No.	Decladding Solution	U, M	Zr, M	F, M	Sn, M	Cu, M	H^+, M
		Diluted Waste Solution from Decladding Step					
9	6 M NH_4F-0.15 M $CuCl_2$	0.13×10^{-4}	0.40	3.21	7.9×10^{-3}	2.2×10^{-2}	---
10	6 M NH_4F-0.1 M $CuCl_2$	0.10×10^{-4}	0.45	1.96	1.1×10^{-3}	---	---
11		5.9×10^{-4}	0.57	3.70	7.6×10^{-3}	1.5×10^{-2}	---
12		2.2×10^{-4}	0.60	3.38	9.2×10^{-3}	4.4×10^{-2}	---
13		9.5×10^{-4}	0.58	3.52	6.7×10^{-3}	2.5×10^{-2}	---
14	6 M NH_4F-1.0 M NH_4NO_3	0.04×10^{-4}	0.43	3.12	28×10^{-3}	---	---
8		1.8×10^{-4}	0.78	3.65	12.9×10^{-3}	---	---
15		9.3×10^{-4}	0.62	3.80	4.6×10^{-3}	---	---
	Avg.	3.62×10^{-4}	0.55	3.29	9.8×10^{-3}	2.65×10^{-2}	---
		Solution Resulting from Core Dissolution					
9		1.29	11.9×10^{-3}	0.13	3.0×10^{-4}	8.0×10^{-2}	2.78
10		1.19	32.5×10^{-3}	0.18	8.4×10^{-4}	5.7×10^{-2}	3.90
11		1.30	19.6×10^{-3}	0.04	84×10^{-4}	4.2×10^{-2}	2.86
12		1.30	4.0×10^{-3}	0.37	8.4×10^{-4}	11.6×10^{-2}	3.05
13		1.30	4.7×10^{-3}	0.11	8.4×10^{-4}	8.1×10^{-2}	2.95
14		1.29	2.2×10^{-3}	0.13	8.4×10^{-4}	---	2.70
8		1.28	3.7×10^{-3}	0.05	21×10^{-4}	---	2.81
15		1.30	9.4×10^{-3}	0.006	4.2×10^{-4}	---	3.43
	Avg.	1.28	11.0×10^{-3}	0.13	18.2×10^{-4}	7.5×10^{-2}	3.06

4.4 Corrosion Tests

Scouting tests were performed to determine, in a cursory manner, the effect of nitrate and cupric ion on the rate of corrosion of several stainless steels. Type 304L stainless steel was corroded at a rate of only 0.1 mil/mo by boiling 4 M NH_4F.[8] Alternate cycles of boiling NH_4F and nitric acid did not result in a higher corrosion rate.[9] In 24-hr tests in this laboratory types 347, 304L and 309Cb stainless steels were corroded at rates of 2.6, 1.5 and 1.1 mils/mo, respectively, in boiling 6 M NH_4F-0.1 M $CuCl_2$. In similar tests with 6 M NH_4F-1.0 M NH_4NO_3, corrosion rates of 20, 17 and 39 mils/mo were obtained for the respective stainless steels.

In one test stainless steel coupons were exposed alternately to boiling 6 M NH_4F-0.1 M $CuCl_2$ and boiling 10 M HNO_3 for 1.5 cycles. Each exposure period was 24 hr. The specimens were cleaned in 3 M HNO_3 after the final test. Corrosion rates were 18.3, 8.5 and 5.7 mils/mo for types 347, 304L and 309Cb stainless steels, respectively.

5.0 DISCUSSION

5.1 Foaming During Decladding

Dissolution of zirconium in aqueous NH_4F produces foam. As an illustration, the foam level was 4-6 inches above the liquid level when the apparatus shown in Fig. 3.1 was used. This corresponds to a volume of foam to volume of solution ratio of about 0.6. In cases where too little free space was provided between the liquid level and the condenser, the buildup of foam caused serious "burping." The amount of foam decreases as the reaction proceeds. No attempt was made to reduce the foaming with an antifoaming agent.

5.2 Volume Reduction During Decladding

A volume reduction of approximately 15% occurs during the decladding step. The amount of volume reduction is important when calculating the amount of water needed to dilute the decladding waste solution and the amount of $CuCl_2$ solution required.

The volume of water used to dilute the decladding waste solution is at least equal to the final volume of the decladding solution. Dilution of the waste stream in this manner prevents precipitation of $(NH_4)_3ZrF_7$ and similar compounds.

To conserve reagent, $CuCl_2$ is added to dissolve tin after the zirconium cladding has been removed with NH_4F (Sec. 5.3). Cupric chloride solution, of appropriate concentration, is added to restore the system to its original volume, and to produce a final solution which is 0.1 M in Cu^{++}.

5.3 Formation of Stannic Oxide During Core Dissolution

Metallic tin, present in Zircaloy-2, is not soluble in aqueous NH_4F, and will therefore be present during core dissolution unless treated specifically. When tin is contacted with cold concentrated nitric acid the reaction is[10]

$$Sn + 2HNO_3 \longrightarrow H_2SnO_3 + NO_2 + NO.$$

As the temperature of the system is raised, metastannic acid (H_2SnO_3) dehydrates to stannic oxide, viz., $H_2SnO_3 \longrightarrow SnO_2 + H_2O$. Both H_2SnO_3 and SnO_2 are insoluble in nitric acid. Occlusion of small UO_2 particles by deposition of SnO_2 on their surfaces is probably the cause of the excessively high uranium losses to the solids which remain after core dissolution (Table 4.1). This hypothesis is further substantiated by x-ray and chemical analyses of the solids which showed the presence of UO_2 (Table 5.1). While occlusion of UO_2 did not occur in all cases, the incidence was high enough to require special treatment for the removal of tin prior to core dissolution. A precipitate of H_2SnO_3, produced by the reaction of a 3 M HNO_3-1.3 M $UO_2(NO_3)_2$ solution with metallic tin, contained only 0.03% of the uranium.

5.4 Exposure of Core to Decladding Solution

Several cases may be cited where the uranium-bearing core material would be exposed to the decladding solution for long periods of time, and could conceivably result in very high uranium losses: (1) failure of an operator to drain the decladding solution from the dissolver at the prescribed time, (2) addition of $CuCl_2$ to dissolve tin after the zirconium cladding has been removed with NH_4F, and (3) formation of holes in the cladding in those cases where the decladding solution can only penetrate through defects in the ZrO_2 surface film. For further discussion of this last point, see Sec. 6.0. The buildup of uranium in boiling 6 M NH_4F-0.1 M $CuCl_2$ is much more rapid (0.14 mg/min·cm^2) than that (0.02 mg/min·cm^2) in boiling 6 M NH_4F or 6 M NH_4F-1.0 M NH_4NO_3 (Fig. 5.1). The data given in Fig. 5.1 were obtained by exposing sintered UO_2 pellets to the boiling solutions. The reported solubility of U(IV) in 6 M NH_4F solutions containing zirconium is about 0.0023 M.[9] Uranium(VI) is reportedly less soluble than U(IV) in concentrated NH_4F solutions.[8]

Table 5.1. Comparison of Uranium Losses to the Weight and Composition of the HNO_3-insoluble Residue

Run No.	Wt of HNO_3-insoluble Residue, g	U Lost to Residue, %	X-ray Analysis of Residue	Sn in Residue, %
2	0.854	0.0012	$SnO_2 + ZrO_2$	44.7
3	1.612	0.61	$SnO_2 + UO_2 + ZrO_2$	12.7
4	1.783	0.76	$SnO_2 + UO_2$	12.5
5	0.693	0.0038	SnO_2	34.8
6	0.689	0.011	----	66.0
7	~0.1	----	$UO_2 + SnO_2$	----
8	0.238	0.0015	SnO_2	55.0
9	0.121	0.1 max[a]	SnO_2	----
10	0.734	0.012	SnO_2	52.0
11	0.0119	0.009 max[a]	----	----
12	0.0211	0.015 max[a]	$ZrO_2 + \beta$ Sn	----
13	0.0285	0.021 max[a]	$ZrO_2 + \beta$ Sn	----
14	0.0891	0.0025	SnO_2	----
15	0.103	0.0006	----	----

[a]The maximum loss was computed by assuming that the residue was pure uranium.

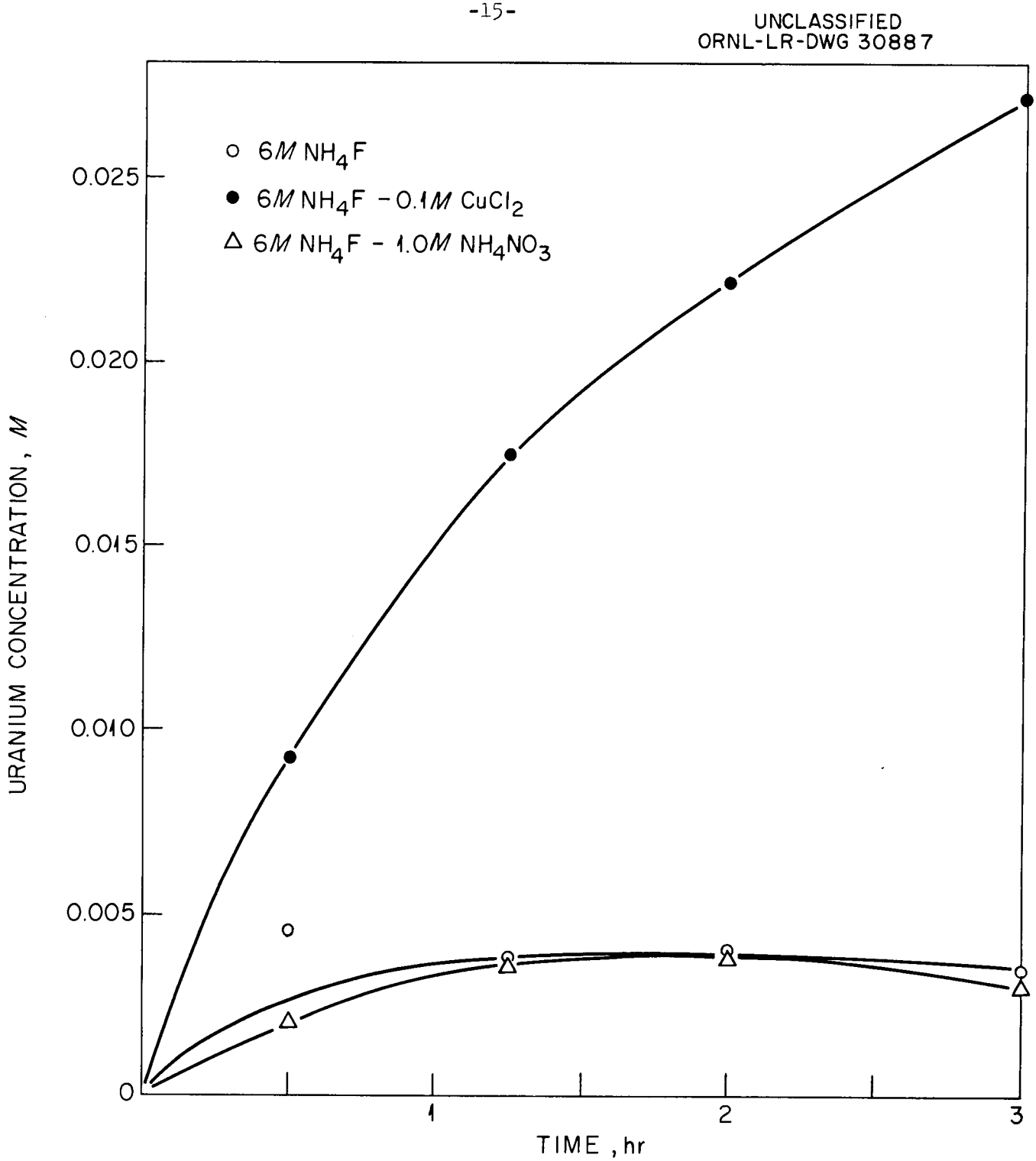

Fig. 5.1. Exposure of sintered UO_2 pellets to various boiling decladding solutions.

5.5 Fluoride Ion Concentration in the Solvent Extraction Feed

The concentration of fluoride ion in the solvent extraction feed solution averaged 0.1 M (Table 4.4). This concentration is very high and could lead to serious corrosion problems. In each case, the UO_2 pellets were carefully washed after the decladding step with six successive portions of cold water. Therefore, the fluoride which appeared in the solvent extraction feed solution was probably very strongly sorbed by the UO_2 pellets.

5.6 Regeneration of NH_4F from Waste Decladding Solutions

Decladding of zirconium fuels produces aqueous waste solutions of $(NH_4)_3ZrF_7$ and similar compounds. It has been reported[5] that the addition of NH_3 to such solutions regenerates NH_4F with simultaneous precipitation of hydrous ZrO_2. The reactions involved may be

$$NH_3 + H_2O \longrightarrow NH_4OH$$

and

$$4NH_4OH + (NH_4)_3ZrF_7 \longrightarrow 7NH_4F + Zr(OH)_4.$$

Very preliminary work in this laboratory has shown that about 98% of the zirconium is precipitated at 25°C and that the precipitate is completely soluble in nitric acid. Recycle of NH_4F therefore appears feasible, and would not only reduce chemical costs, but also reduce the volume and corrosiveness of the final waste solution.

6.0 FUTURE WORK

In future work the effect of the tenacious ZrO_2 coating present on fuel exposed to high temperature water must be evaluated. The rate of penetration of unoxidized zirconium by aqueous NH_4F is nearly linear[3] so that the time required for dejacketing is easily computed. However, penetration through a ZrO_2 coating will occur only where there is a defect in the coat.[11] Formation of holes in the cladding would lead to longer exposure of the core material than predicted from decladding rates of unoxidized zirconium. The uranium loss to the decladding solution could therefore be correspondingly higher.

Addition of aluminum nitrate to the solvent extraction feed solution may be required to reduce corrosion by fluoride ion. Washing of the UO_2 pellets with cold water after decladding did not completely remove the fluoride. Future work will include the determination of better methods for removing fluoride prior to core dissolution.

Extensive corrosion tests will be required to determine if stainless steel will be a suitable material of construction. Corrosion rates were very high in 24-hr tests (Sec. 4.4) when either ammonium nitrate or cupric chloride was used in the decladding solution.

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