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MISCELLANEOUS EXPERIMENTS RELATING TO THE PROCESSING OF CETR FUEL BY SULFEX-THOREX AND DAREX-THOREX PROCESSES

L. M. Ferris
A. H. Kibbey

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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OAK RIDGE NATIONAL LABORATORY
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ABSTRACT

Experiments with unirradiated Consolidated Edison reactor ThO_2-UO_2 fuel pellets indicated that uranium losses to Sulfex and Darex decladding solutions were proportional to the UO_2 content of the pellets. For example, after 7 hr, losses to boiling initial Darex solution (5 M HNO_3—2 M HCl) were 0.45 and 0.65% from pellets containing 3 and 9% UO_2, respectively. The initial rate of dissolution of these pellets in 200% excess boiling 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$ was essentially independent of the UO_2 content. Rates were 2.1, 3.0, and 2.4 mg min^{-1} cm^{-2} for pellets containing 3, 6, and 9% UO_2, respectively. The presence in the dissolvent of the soluble neutron poisons H_3BO_3 and $Cd(NO_3)_2$ in concentrations up to 0.1 M and 0.075 M, respectively, had little effect on the rate of dissolution of sintered UO_2-ThO_2 fuel pellets.

CONTENTS

1.0 INTRODUCTION

The purpose of this report is to summarize laboratory data obtained on the processing of unirradiated Consolidated Edison power reactor fuel between January 1, 1960 and January 31, 1961. Information relating to both the Sulfex-Thorex and Darex-Thorex processes (1) is included. Experimental work performed prior to January 1, 1960 and preliminary results with irradiated fuel specimens have been reported (1-7).

2.0 RESULTS

2.1 Physical Properties of Process Solutions

2.1.1 Densities of Stainless Steel--Sulfuric Acid Solutions

Density measurement may be suitable for regulation of the Sulfex-Thorex process. The densities of stainless steel--sulfuric acid solutions varying in composition from 0 to 10 N H_2SO_4 and from 0 to 40 g of stainless steel per liter were determined over the temperature range 20 to 80°C. The data were correlated by means of the equation

$$d = 1 - 4.31 \times 10^{-6}(t^2) + 0.0275\,(H^+) + 0.00262\,(SS)$$

in which the variables were

d = density, g/ml
t = temperature, °C
(H^+) = sulfuric acid normality
(SS) = stainless steel concentration, g/liter

Data for solutions at 26°C are presented in Fig. 1.

2.1.2 Densities of Nitric Acid-Thorium Nitrate Solutions

Densities of HNO_3-$Th(NO_3)_4$ solutions, similar to those expected from dissolution of the CETR ThO_2-UO_2 fuel pellets, were reported by other workers (8,10).

2.1.3 Boiling Points of Some Process Solutions

Although a systematic determination of the boiling points of Sulfex, Darex, and Thorex process solutions was not made, some data were collected in the course of the work. These data are listed in Table 1.

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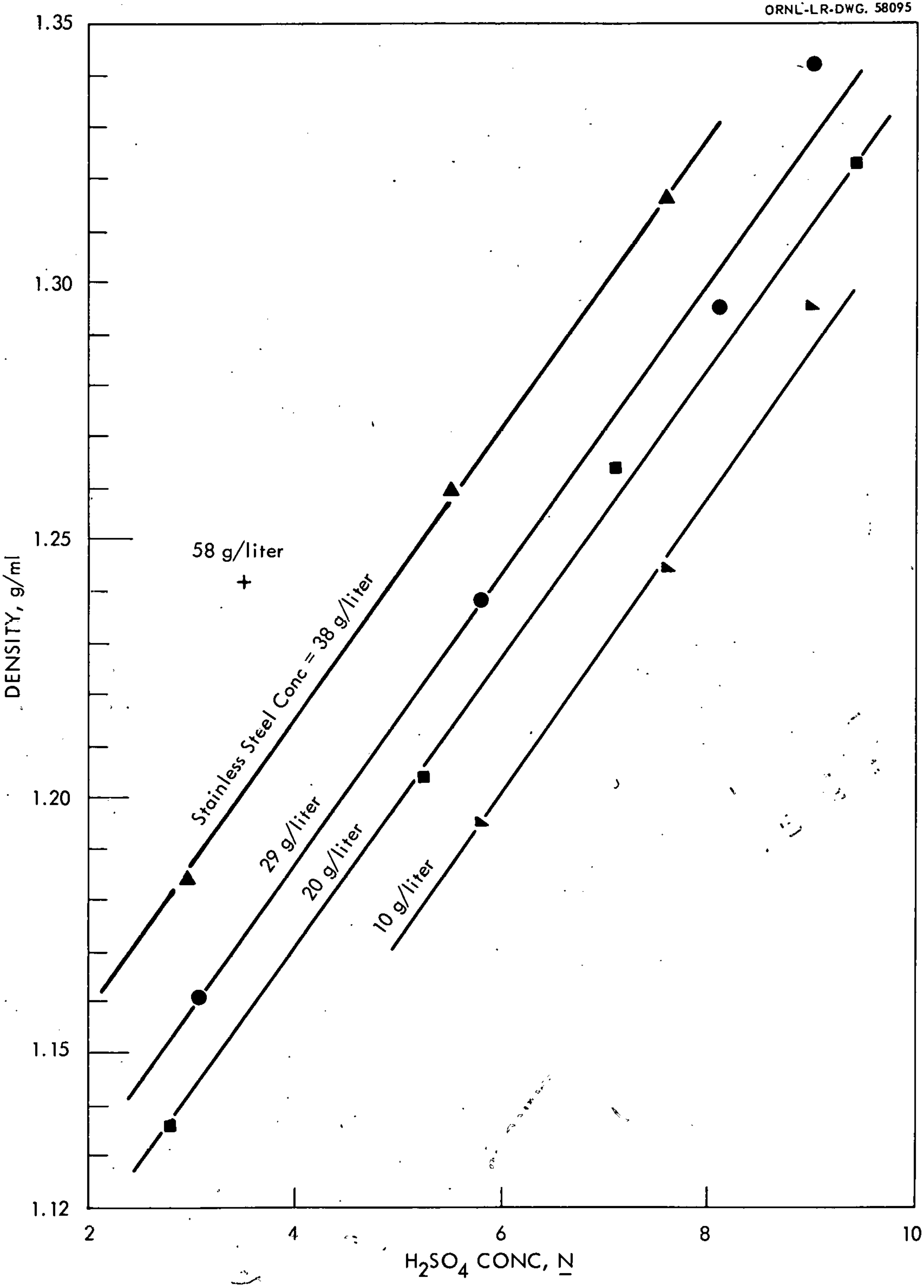

Fig. 1. Densities of stainless steel—sulfuric acid solutions at 26°C.

Table 1. Boiling Points of Some Darex, Sulfex, and Thorex Process Solutions

Solution	Boiling Point, °C, at ~745 mm Hg
5 M HNO_3—2 M HCl	105-107
1.5 M HNO_3—1.5 M HCl—50 g SS/liter	109
4 M H_2SO_4	107-109
6 M H_2SO_4	115-117
6.5 N H_2SO_4—100 g SS/liter	115-116
4.5 N H_2SO_4—70 g SS/liter	107
13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$	118
8.5 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$—1 M $Th(NO_3)_4$	126-129

2.1.4 Solubility of Thorium Nitrate in Nitric Acid

In previous studies the solubility of thorium nitrate in nitric acid was measured only in solutions where the acidity was less than about 10 M (10-12). The results of this investigation, which are in excellent agreement with those of the other workers, show that at 26°C the solubility of thorium nitrate decreases from about 620 to 180 g of thorium per liter when the nitric acid concentration increases from 0 to 18.3 M (Fig. 2).

2.2 Studies Relating to Decladding Losses

2.2.1 Effect of UO_2 Content of Fuel Pellet on Decladding Losses

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Interest in this variable stems from the fact that the Consolidated Edison reactor will have a three-region core containing fuel pellets ranging in UO_2 content from 3 to 10% (13). Uranium and thorium losses were determined by exposing unirradiated pellets containing 3, 6, and 9% UO_2 to boiling Sulfex and Darex decladding solutions. The pellets were nominally 93% of theoretical density. The results, while not entirely conclusive, indicated that soluble losses were a function of the UO_2 content of the pellet. After a 7-hr exposure to boiling initial Darex solution (5 M HNO_3—2 M HCl), for example, uranium losses were 0.45 and 0.65% from pellets containing 3 and 9% UO_2, respectively (Fig. 3a). In the initial Darex solution, the losses from pellets containing 6% UO_2 were higher than those from pellets containing 9% UO_2; this effect may be due to the lower density of the pellets containing 6% UO_2 (Sect. 2.3.1). In the initial Sulfex solution (6 M H_2SO_4) the losses were in direct proportion to the UO_2 content (Fig. 3b). As expected from previous studies (1,6,7), the Sulfex losses were 2- to 10-times lower than those to Darex solutions.

Thorium losses were 2- to 10-times lower than the uranium losses in both the Sulfex and Darex systems, and were also somewhat dependent on the composition of the pellets (Table 2). Losses to final solutions, i.e., those containing dissolved stainless steel, were lower than those to the initial solutions (Table 2).

Table 2. Effect of UO_2 Content on Losses of Uranium and Thorium from Unirradiated CETR Fuel Pellets to Darex and Sulfex Decladding Solutions

Pellets[a] exposed to boiling solutions for 48 hr

Solution	Losses, % 3% UO_2 Pellets U	Th	6% UO_2 Pellets U	Th	9% UO_2 Pellets U	Th
5 M HNO_3—2 M HCl	0.37	0.03	2.17	0.31	1.32	0.36
1.5 M HNO_3—1.5 M HCl—50 g SS/liter	0.35	0.02	0.07	0.09	0.55	0.08
6 M H_2SO_4	0.17	0.05	0.20	0.08	0.20	0.06
5 M H_2SO_4—50 g SS/liter	0.05	0.02	0.09	0.05	0.52	0.15

[a] Pellets nominally 93% of theoretical density.

2.3 Studies Relating to Dissolution of ThO_2-UO_2 Fuel Pellets

2.3.1 Effect of UO_2 Content on Rate of Dissolution

Because the Consolidated Edison reactor will contain fuel pellets of varying UO_2 content, it is essential to determine the effect of this variation on the rate of dissolution of the pellets in the Thorex dissolvent, 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$. Preliminary studies indicated that the amount of UO_2, up to 10%, had no effect on the rate of dissolution:

UO2 in Pellet, %	O/U Ratio	Bulk Density, g/cc	Initial, 10-min, Dissolution Rate, mg min^{-1} cm^{-2}
3	2.005	9.2	2.1
6	2.005	9.1	3.0
9	2.005	9.3	2.4

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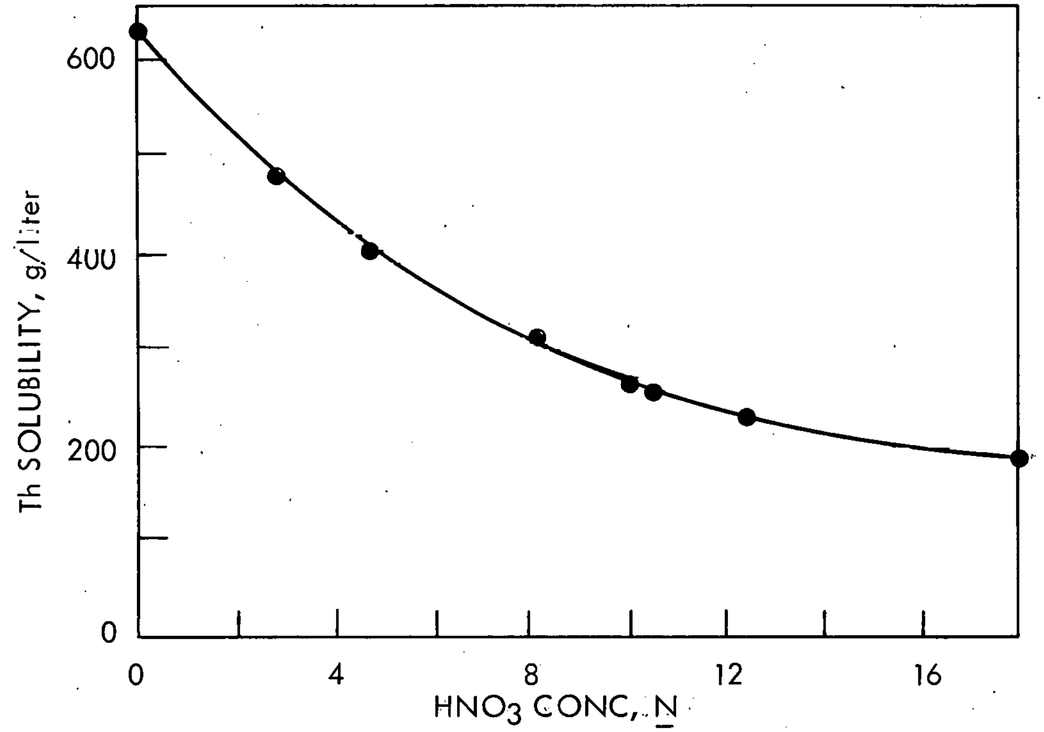

Fig. 2. Solubility of thorium nitrate in nitric acid solutions at 26°C.

The variation in rates is within the experimental error, and is probably due in part to the difference in densities of the pellets.

2.3.2 Effect of Dissolved Nuclear Poisons on the Dissolution Rate

Addition of soluble neutron poisons such as boron or cadmium to process solutions to aid in criticality control has been suggested (6). Preliminary studies have shown that the presence of up to 0.1 M H_3BO_3 or 0.075 M $Cd(NO_3)_2$ in boiling 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$ had little effect on the rate of dissolution of 95% ThO_2—5% UO_2 pellets whose densities were about 94% of theoretical.

The pellets used in these studies were supplied by the Universal Match Company and contained 5.3% $UO_{2.0}$. In three series of experiments, made to evaluate the effect of boron, initial 10-min rates of dissolution were determined

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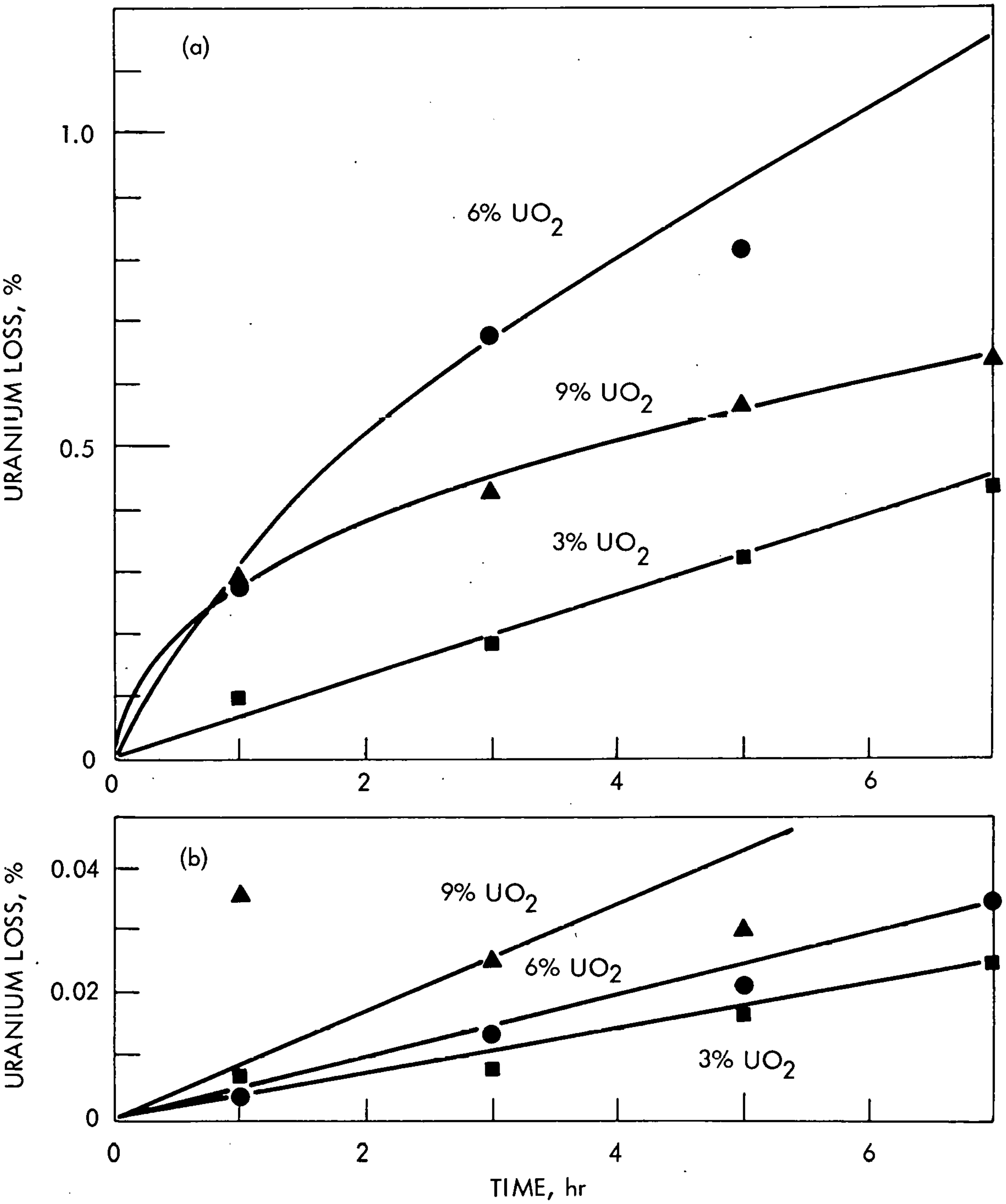

Fig. 3. Soluble uranium losses from ThO_2-UO_2 pellets containing 3, 6, and 9% UO_2 in boiling (a) 5 $\underline{M}$ HNO_3—2 $\underline{M}$ HCl and (b) 6 $\underline{M}$ H_2SO_4. Pellets were nominally 93% of theoretical density.

in boiling 13 M HNO_3—0.04 M NaF containing 0, 0.04, and 0.1 M $Al(NO_3)_3$ and boric acid in concentrations up to 0.2 M. A 200% stoichiometric excess of dissolvent was used in each case. The highest 10-min rate, about 2 mg min^{-1} cm^{-2}, was obtained when the dissolvent contained no aluminum and less than 0.1 M H_3BO_3 (Fig. 4). When the boric acid concentration was constant, the 10-min rate decreased with increasing aluminum concentration as expected (1). In solutions having a constant aluminum concentration, the 10-min rate was independent of boric acid concentration up to about 0.1 M; however, the rate in solutions containing no aluminum or 0.1 M aluminum decreased about 30% as the boric acid concentration increased from 0.1 to 0.2 M (Fig. 4). The rate in all solutions containing 0.04 M aluminum was nearly constant at 1.3 mg min^{-1} cm^{-2}. Solutions containing 1 M thorium were stable with respect to precipitation when the boric acid concentration was 0.1 M.

The effect of cadmium on the dissolution rate has not yet been clearly elucidated. In the first series of experiments (series 1, Table 3), the initial, 10-min, rate of dissolution in solutions containing no aluminum decreased from 2 to 0.7 mg min^{-1} cm^{-2} as the cadmium concentration increased from 0 to 0.075 M. However, with 0.1 M aluminum present, the rate increased from 0.7 to 2 mg min^{-1} cm^{-2} as the cadmium concentration increased from 0 to 0.075 M. These experiments were repeated with hopes of substantiating the results. On the contrary, data from the second series of experiments (series 2, Table 3) indicated that, at all aluminum concentrations, the initial rate decreased with increasing cadmium concentration from about 1.4 mg min^{-1} cm^{-2} to a minimum of about 0.8 mg min^{-1} cm^{-2} at about 0.05 M cadmium; the rates increased to about 1.3 mg min^{-1} cm^{-2} at 0.075 M cadmium. Because of the wide scatter in the data, it can be only tentatively concluded that the dissolution rate is not seriously affected by the presence of up to 0.075 M $Cd(NO_3)_2$.

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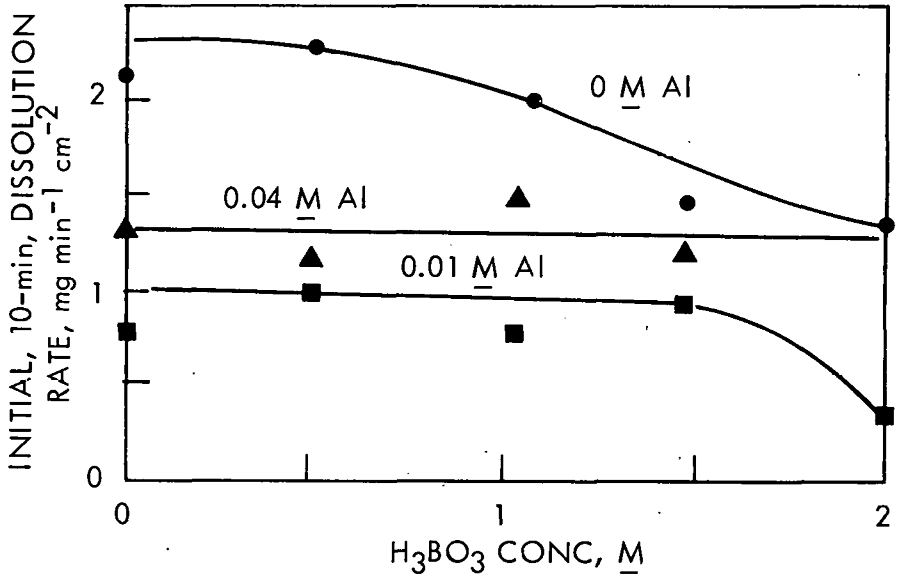

Fig. 4. Effect of boric acid and aluminum nitrate concentrations on the initial 10-min rate of dissolution of Universal Match Co. 94.7% ThO_2—5.3% UO_2 pellets in 200% stoichiometric excess of boiling 13 M HNO_3—0.04 M NaF. Pellet densities: 92-94% of theoretical.

2.3.3 Dissolution of ThO_2-UO_2 Fissia Pellets

Preliminary studies were conducted to determine the effect of fission product concentration on the rate of dissolution of UO_2-ThO_2 pellets. Unirradiated pellets used in these studies contained 5% UO_2 and either 2 or 4% "fissia", with the remainder being ThO_2.[a] Burnups of about 20,000 and 40,000 Mwd/ton of core were simulated by the addition of 2 and 4% "fissia", respectively. The "fissia" mix had the composition

Mo	13.8%	CeO_2	13.0%
ZrO_2	20.0	La_2O_3	6.5
Ru	7.8	Pr_6O_{11}	5.5
$BaCO_3$	12.2	Sm_2O_3	2.3
Nd_2O_3	14.4	Y_2O_3	2.5

[a] The pellets were made by J. Griffin, Y-12 plant, by mixing pure UO_2 and ThO_2 powders with the "fissia" mix, pressing the pellets, which were fired in hydrogen at 1700°C for 1 hr, and then cooled in hydrogen. The "fissia" mix was supplied by Atomics International.

The initial rate of dissolution of the pellets in 200% stoichiometric excess of boiling 13 $\underline{M}$ HNO_3—0.04 $\underline{M}$ NaF—0.1 $\underline{M}$ $Al(NO_3)_3$ was proportional to the "fissia" content:

Amount of Fissia in Pellet, %	Density of Pellet, g/cc	Initial, 10-min, Dissolution Rate, mg min^{-1} cm^{-2}
2	9.07	0.73
2	9.44	1.0
4	9.49	1.3
4	9.49	1.3

The tendency of the pellets containing 4% "fissia" to dissolve faster was substantiated in other experiments (Table 4). Even with crushed pellets, dissolution was incomplete in 85 hr. It was established that the residue contained UO_2 and ThO_2 in addition to zirconium and molybdenum oxides. Because of the low rate of dissolution and the extreme difficulty in analyzing the solutions for the fission product elements, work on these pellets was abandoned. The behavior of fission products during dissolution will probably be determined only when highly irradiated fuel specimens become available.

2.3.4 Dissolution of ThO_2 Pellets and Fused ThO_2

Scouting experiments were performed to determine whether the rate of dissolution of ThO_2 pellets in 200% excess of boiling 13 $\underline{M}$ HNO_3—0.04 $\underline{M}$ NaF—0.1 $\underline{M}$ $Al(NO_3)_3$ was different from those obtained with pellets containing up to 10% UO_2. The pellets were fabricated by the Davison Chemical Company and had densities varying between 85 and 99% of theoretical. Initial, 10-min, dissolution rates of pellets whose densities were 8.6 and 9.8 g/cc were 1.9 and 1.1 mg min^{-1} cm^{-2}, respectively. These rates are

Table 3. Effect of $Cd(NO_3)_2$ and $Al(NO_3)_3$ Concentrations on Initial, 10 min, Rate of Dissolution of 95% ThO_2—5% UO_2 Pellets in Boiling 13 M HNO_3—0.04 M NaF

200% stoichiometric excess of reagent used in each case

$Cd(NO_3)_2$ Conc, M	$Al(NO_3)_3$ Conc, M	Pellet[a] Density, g/cc	Initial, 10-min, Dissolution Rate, mg min^{-1} cm^{-2}
0.0	0.0	9.38	2.0
0.0	0.04	9.37	1.3
0.0	0.10	9.45	0.7
0.025	0.0	9.36	1.9
0.025	0.04	9.34	1.2
0.025	0.10	9.32	0.80
0.05	0.0	9.42	1.2
0.05	0.04	9.35	1.4
0.05	0.10	9.39	1.4
0.075	0.0	9.41	0.67
0.075	0.04	9.39	1.1
0.075	0.10	9.32	2.0
0.0	0.0	9.39	1.5
0.0	0.04	9.42	1.2
0.0	0.10	9.26	1.4
0.025	0.0	9.45	1.2
0.025	0.04	9.38	1.2
0.025	0.10	9.36	0.83
0.05	0.0	9.50	0.78
0.05	0.04	9.43	0.79
0.05	0.10	9.42	0.79
0.075	0.0	9.36	1.2
0.075	0.04	9.39	1.4
0.075	0.10	9.35	1.3

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[a] Theoretical density: 10.0 g/cc.

Table 4. Dissolution of ThO_2—5% UO_2—"Fissia" Pellets[a] in 200% Stoichiometric Excess of Boiling 13 M HNO_3—0.04 M NaF--0.1 M $Al(NO_3)_3$

Expt. No.	Amt. of Fissia in Pellet, %	State of Pellet	Dissolution Time, hr	Amt. Dissolved, %
1	2	Crushed	50	80
2	4	Crushed	50	92
3	2	Whole	21[b]	70
4	4	Whole	21[b]	91

[a] The pellets were made by J. Griffin, Y-12 plant, by mixing pure UO_2 and ThO_2 powders with the "fissia" mix, pressing the pellets, which were fired in hydrogen at 1700°C for 1 hr, and then cooled in hydrogen. The "fissia" mix was supplied by Atomics International.

[b] Three 7-hr digestions with fresh reagent.

Table 5. Dissolution of Sintered ThO_2 Pellets in 200% Stoichiometric Excess of Boiling 13 M HNO_3--0.04 M NaF—0.1 M $Al(NO_3)_3$

Time, hr	Hydrogen Ion Conc, M	Amount Dissolved, %	
		9.8-g/cc Pellet	8.6-g/cc Pellet
1	12.0	9.3	14.2
2.5	11.5	23.2	32.0
20	8.3	89.5	93.6
24	8.1	94.8	97.0
26.5	8.0	95.0	97.1

not significantly different from those obtained with pellets containing 3-10% UO_2 (Sects. 2.3.1 and 2.3.2; also see ref. 1). In 26.5 hr, 95-97% of the ThO_2 pellets dissolved (Table 5). A small residue, shown by x-ray analysis to be ThO_2, was visible at the end of each experiment.

The amount of fused ThO_2 dissolved in 200% stoichiometric excess of boiling 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$ was determined as a function of time and mesh size and the results compared to those obtained under similar conditions with 95.8% ThO_2—$UO_{2.36}$ which was 93% of theoretical density. As expected, the fused ThO_2 dissolved much more slowly; e.g., after 5 hr, only about 30% of the -4+8 mesh fused ThO_2 dissolved compared to about 88% of the other material (Fig. 5). Since the presence of up to 10% UO_2 apparently has no effect on the rate of dissolution (Sect. 2.3.1), the results obtained with fused ThO_2 are attributed mainly to its higher density.

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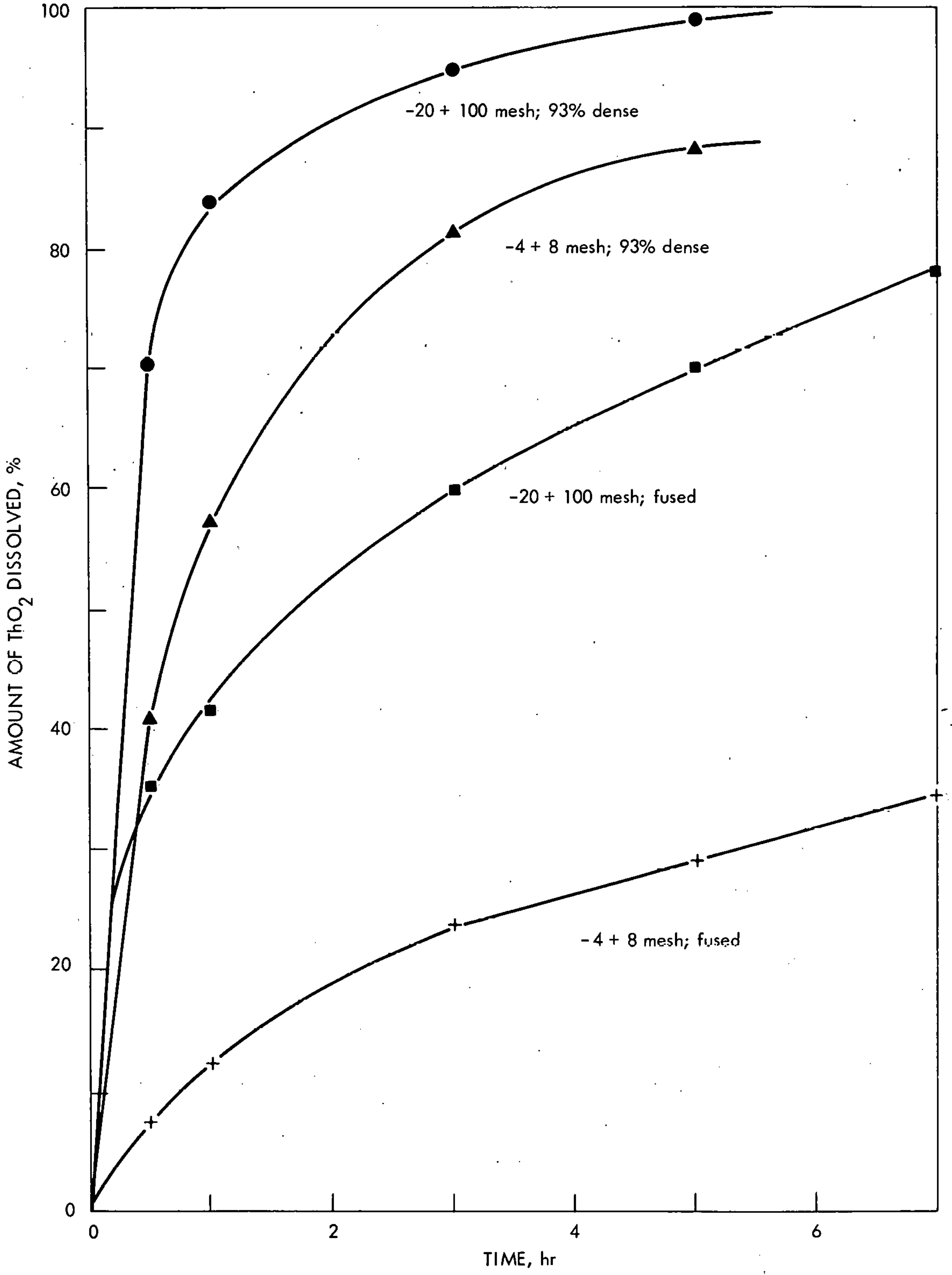

Fig. 5. Comparison of rates of dissolution of fused ThO_2 and 95.8% ThO_2—$UO_{2.36}$ (93% of theoretical density) in 200% excess of boiling 13 $\underline{M}$ HNO_3—0.04 $\underline{M}$ NaF—0.1 $\underline{M}$ $Al(NO_3)_3$.

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