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SULFEX PROCESS: ENGINEERING-SCALE SEMICONTINUOUS DECLADDING OF UNIRRADIATED STAINLESS STEEL-CLAD UO_2 AND UO_2-ThO_2

B. C. Finney
B. A. Hannaford

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Page 6, paragraph 3, line 3 reads

20.01% SO_2, 0.02% A, < 0.1% H_2S, ...

It should read

< 0.01% SO_2, 0.02% A, < 0.1% H_2S, ...

Corrected
5/19/61

ORNL-3072

Contract No. W-7405-eng-26
Chemical Technology Division

SULFEX PROCESS: ENGINEERING-SCALE SEMICONTINUOUS DECLADDING OF UNIRRADIATED STAINLESS STEEL-CLAD UO_2 AND UO_2-ThO_2

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ABSTRACT

An engineering-scale demonstration of the Sulfex (4-6 M H_2SO_4) process indicated that semicontinuous decladding of unirradiated stainless steel-clad UO_2 or UO_2-ThO_2 fuels is feasible. In a long cylindrical dissolver, 4-in. i.d. by 19-ft high, solution bumping or foaming did not cause serious operational problems.

For decladding of prototype Consolidated Edison UO_2-ThO_2 fuel assemblies, 2.8-4.0 hr was required. Soluble thorium losses to the clad solution varied from 0.022% in the absence of a heel to 0.25% in the presence of a heel. Soluble uranium losses were fairly constant, ~0.02%.

Clarification of Sulfex clad solutions by gravity sedimentation and centrifugation produced a solid waste equivalent to 1.3 to 2.7% of the weight of the stainless steel dissolved in 4 M H_2SO_4. Mean particle size ranged from 3 to 7 μ. Complete dissolution of waste solids by common aqueous reagents was not possible.

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1.0 INTRODUCTION

The semicontinuous Sulfex head-end treatment, decladding of stainless steel-clad UO_2 and UO_2-ThO_2 fuel elements with boiling 4 or 6 M H_2SO_4, was demonstrated on an engineering scale (4% of the scale of the proposed Power Reactor Fuel Processing Pilot Plant). The equipment was fabricated of glass, Carpenter 20, and Ni-O-Nel. Initial laboratory-scale development of the Sulfex process was based on batch processing.[1-3] Some reactor fuels are of sufficient enrichment that criticality control must be considered. Because of the relative size and cost of a critically safe batch dissolver as compared to a long cylindrical dissolver, the decision was made that the engineering development of Sulfex would utilize semicontinuous decladding and core dissolution (batch with respect to the solid phase and continuous with respect to the liquid phase).

The experiments included dissolution of the stainless steel and decladding of unirradiated prototype fuel elements. Cores were dissolved for removal from the equipment. Both clad and core solutions were clarified, and the nature of the waste solids was investigated briefly.

Acknowledgment. The authors wish to acknowledge the experimental work performed by F. L. Rogers, T. D. Napier, and G. B. Dinsmore. Chemical analyses were provided by the groups of G. R. Wilson and W. R. Laing of the ORNL Analytical Chemistry Division. Particle size analyses were provided by L. C. Bate and T. E. Willmarth of the Analytical Chemistry Division.

2.0 EQUIPMENT

The equipment (Fig. 1) consisted of a dissolvent acid feed system, a dissolver and auxiliary equipment, and an off-gas system. Hot dissolvent flowed into the bottom of the dissolver, and the metal-laden product was removed at a point sufficiently high above the bottom that the fuel elements were totally submerged during dissolution. The product flowed from the dissolver to a vertical settler, to a horizontal settler, and then to a catch tank, centrifuge, or acid feed tank for recirculation back to the dissolver.

The dissolvent acid feed system consisted of a 55-gal polyethylene-lined stainless steel drum for sulfuric acid and a 55-gal stainless steel drum for Thorex dissolvent (13 M HNO_3—0.04 F^-—0.1 M Al^{3+}). An Eco Teflon 3-gpm gear pump with Carpenter 20 housing and variable-speed drive transferred the dissolvent from the feed tank to a 4-in.-i.d. x 1-ft-high Pyrex head tank. The head tank was equipped with a standpipe overflow back to the feed tank to maintain a constant dissolvent level. The dissolvent flowed from the head tank through a rotameter and acid heater (1-ft^2 jacketed Carpenter 20 coil) into the bottom of the dissolver.

The dissolver was a 4-in.-i.d. x 19-ft-high Pyrex pipe with Carpenter 20 flanges and a 4-ft^2 Carpenter 20 coil at the bottom for heating and cooling. The dissolvents were maintained at boiling during both clad and core dissolutions. Fuel elements were inserted into the dissolver through a flange at the top.

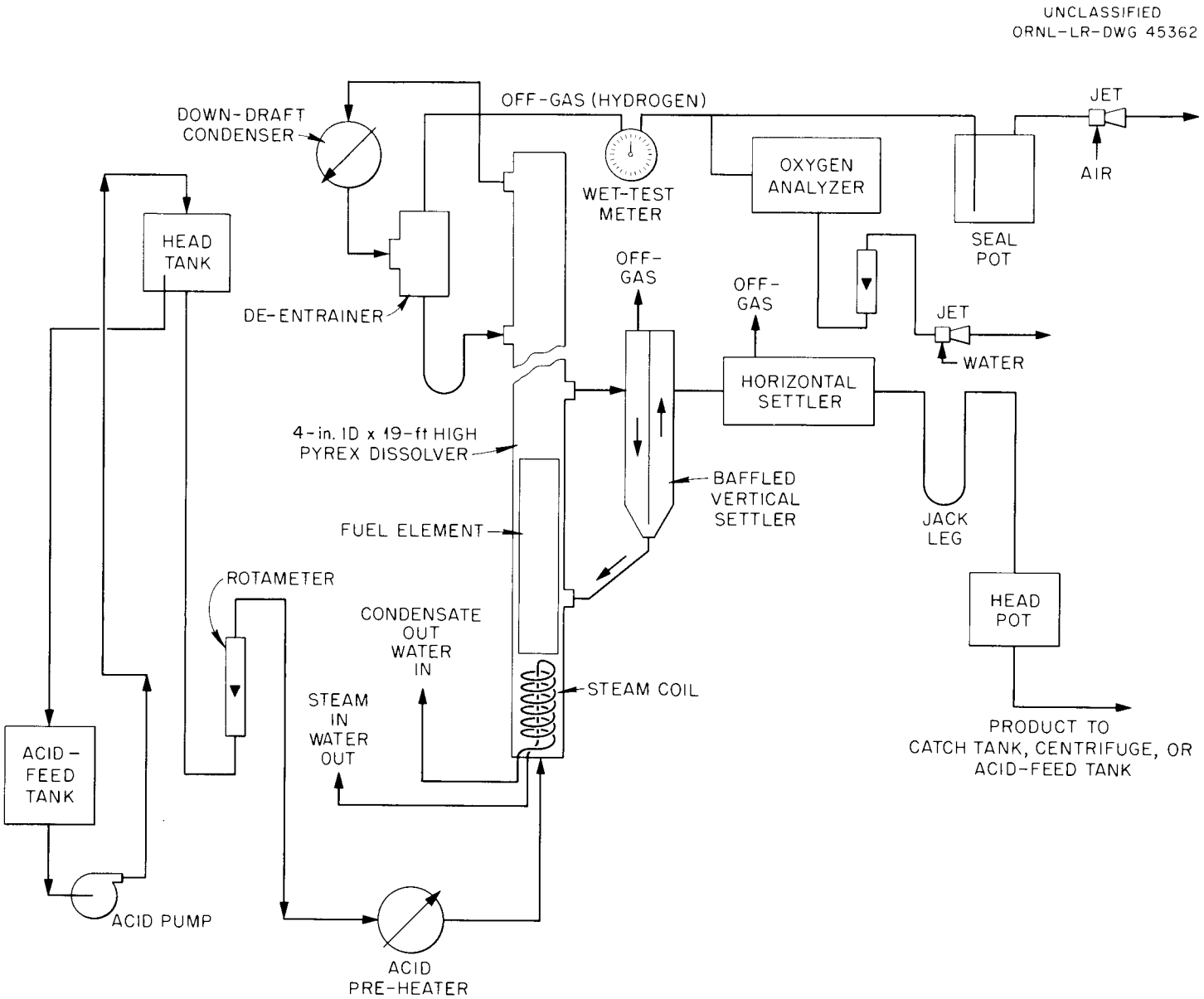

Fig. 1. Schematic diagram of semicontinuous Sulfex equipment.

The dual settler system consisted of a 4-in.-i.d. x 3-ft-high baffled Ni-O-Nel vertical settler and a 4-in.-i.d. x 3-ft-long Pyrex horizontal settler. A double piping arrangement between the vertical settler and dissolver resulted in a flow of liquid from the vertical settler to the dissolver, and consequently large particles were recirculated to the dissolver. At a nominal dissolvent flow rate of 1 liter/min there was ~3 min residence time in the horizontal settler, and approximate calculations indicated that waste particles 10 μ and larger would settle. Both settlers were vented to the off-gas system. A jackleg served as a seal between the system and the atmosphere and cooled the liquor to about 27°C in order to minimize corrosion of the stainless steel centrifuge. The 12-in.-dia centrifuge, with 4.4 liters dynamic volume separated into three compartments by horizontal ported baffles, developed an average centrifugal force of 650 G, sufficient to recover waste solids larger than ~0.5 μ at the feed rates used. The dissolver and horizontal settler were equipped with steam jets for emptying.

The dissolver overhead passed through a downdraft condenser (4 ft^2 area, four 0.5-in. Carpenter 20 tubes wound in a helix, Carpenter 20 heads, and stainless steel shell). A 6-in.-dia Pyrex pipe T was used as a de-entrainer for separation of the condensate and noncondensable off-gas. The condensate was returned to the dissolver, and the noncondensables were removed through the off-gas system.

An analysis of the dissolver noncondensable off-gas was 1.7% N_2, 0.3% O_2, < 0.01% SO_2, 0.02% A, < 0.1% H_2S, < 0.01% SO_3, and the remainder H_2. A wet-test meter was used to measure the hydrogen evolution. A Model C2 Beckman oxygen analyzer was used to continuously monitor the off-gas for oxygen concentration. An air jet was used to maintain the equipment at a slightly reduced pressure (2-4 in. H_2O), and hydrogen was diluted with air at the jet throat.

In both decladding and core dissolution the equipment was operated "semicontinuously," i.e., batch with regard to the solid phase and continuous with regard to the liquid phase.

3.0 EXPERIMENTAL RESULTS

In 36 semicontinuous runs (Tables 1-3) in a tall cylindrical dissolver (Sect. 2.0) there was no inherent difficulty in operation. There was some bumping and foaming of the dissolver solution, with a maximum height of 3-4 ft above the top of the fuel element being reached. Considerable foam was produced on the surface of the stainless steel during dissolution, but the foam was very unstable and none was in the liquid above the metal. In some runs type 304L stainless steel plate assemblies, simulating NMSR fuel cladding, were dissolved in 4 and 6 M H_2SO_4, and type 304L stainless steel tube assemblies were dissolved in 4 M H_2SO_4. In other runs unirradiated stainless steel-clad UO_2 fuel pins and stainless steel clad UO_2-ThO_2 (prototype Consolidated Edison) fuel assemblies were declad. After being declad, the UO_2 cores were dissolved in nitric acid and the UO_2-ThO_2 cores were dissolved in fluoride-catalyzed nitric acid to remove them from the dissolver in preparation for the next run. Core dissolution rate data determined at this time confirmed laboratory-scale experiments.[4]

Table 1. Summary of Semicontinuous Sulfex Run Data (Dissolution of 304L Stainless Steel Plate)

Fuel charge, Type I: 11 plates, 4.5 ft x 3-1/16 in. x 0.038 in. with 1/4 in. plate spacing, 2.2 x 10^4 cm^2 surface area, 7.75 - 8.25 kg

Type II: APPR type plate assembly, 16 plates, 23 x 2.75 x 0.025 in. and two side plates, 23 x 2.5 x 0.025 in. with 1/8-in. plate spacing, 1.46 x 10^4 cm^2 surface area, 4.7 kg

Run No.	Time, hr	Dissolvent			Fuel Elements	Product			H_2 in Off-gas		F/S, cm/min	Reaction Rate (based on total dissolution), $mg/cm^2 \cdot min$	Max Liquid Height in Dissolver, ft
		H_2SO_4, M	Flow Rate, liters/min	Temp., °C		Vol, liters	H_2SO_4, M	SS in solution, g/liter	Vol, ft^3 (STP)	g moles per g mole SS			
SX-2	3.85	6	1.03	114	2 type I	238	4.7	65.0	258	1.17	0.0228	1.52	~ 16
SX-3	7.60	6	0.626	116	2 type I	285	4.81	54.5	347	1.57	0.0142	0.77	~ 18
SX-4	3.50	6	1.30	118	2 type I	270	4.87	58.0	248	1.13	0.0341	1.68	~ 16
SX-5	3.75	6	1.06	115	2 type I	238	4.60	65.0	248	1.13	0.024	1.57	~ 16
SX-6	2.66	6	1.09	115	2 type II	174	4.83	57.1	147	1.11	0.037	2.00	~ 14
SX-7	2.42	6	1.91	118	2 type I	277	5.15	57.5	242	1.07	0.0435	2.49	~ 16
SX-8	1.92	6	2.40	118	2 type I	275	5.23	58.2	260	1.14	0.0545	3.16	~ 19
SX-9	3.5[a]	6	0.626 for 1st hr 1.06 for 2nd hr 1.60 for 3rd hr	118	2 type I	249	5.16	62.0	199	1.11		1.44	~ 16
SX-10	3.5[b]	6	1.3 for 1st hr 1.06 for 2nd hr 1.06 for 3rd hr	118	2 type I	246	4.70	63.7	218	1.12		1.40	~ 16
SX-11	3.42[c]	6	(see footnote d)	118	2 type I	241	4.78	61	230	1.11		1.46	~ 16
SX-12	2.75[e]	4	1.42	115	2 type I	235	2.58	75.2	284	1.13	0.0323	2.43	~ 18
SX-13	2.25	4	1.84	112	2 type I	248	2.86	66.3	(see footenote f)		0.042	2.76	~ 19
SX-14	4.42	4	0.94	112	2 type I	240	2.49	69	269	1.14	0.021	1.42	~ 16
SX-15	2.05	4	0.94	110	1 type I	116	2.50	70.5	139	1.19	0.0427	3.00	~ 14
SX-16[g]	3.25	4	0.718	110	1 type I	138	3.00	58.7	135	1.16	0.0326	1.92	~ 14
SX-17	2.83	4	0.751	105	1 type I	134	2.70	63.8	133	1.15	0.034	2.18	~ 13

[a]Approximately 12.7 kg of fuel element dissolved in 3.0 hr; 52 liters of dissolvent used for heel (~ 3.4 kg) cleanup (0.5 hr).
[b]Approximately 12.9 kg of fuel element dissolved in 3.0 hr; 40 liters of dissolvent used for heel (~ 3.0 kg) cleanup (0.5 hr).
[c]Approximately 13.14 kg of fuel element dissolved in 3.25 hr; 26 liters of dissolvent used to batch dissolve (0.17 hr) an additional 1.5 kg, leaving a 1.2-kg heel.
[d]Combination semicontinuous and batch dissolution used; dissolvent fed continuously at average flow rate of 2 liters/min for 15 min and then the dissolver contents digested 15 min. This procedure repeated 7 times.
[e]17.67 kg of fuel element dissolved, including 1.2-kg heel from run SX-11.
[f]De-entrainer flooded at startup. Condensate carried into wet-test meter, resulting in inaccuracy of off-gas rate and volume measurements.
[g]Line between dissolver and vertical settler intermittently plugged with platelets of stainless steel; for reasons unknown, dissolution ceased for ~0.5 hr.

- -

Table 2. Summary of Semicontinuous Sulfex Run Data (Dissolution of 304L Stainless Steel Tubing)

Dissolvent: Boiling 4 M H_2SO_4

Fuel charge: 3-ft sections of welded 304L stainless steel tubing, 0.5 in. o.d. x 0.035 in. wall thickness

Run No.	Time, hr	Fuel Element			Dissolvent Flow Rate, liters/min	Fuel Element Dissolved, %	Wt of Heel, g	F/S, cm/min	Reaction Rate, $mg/cm^2 \cdot min$	Off-gas		Product	
		No. of tubes	Wt, g	Outer Surface Area, cm^2						H_2 Evolved, ft^3 (STP)	g moles H_2 / g mole SS	SS in Solution, g/liter	Vol, liters
SX-25	5.8	12	2967	4430	0.218	88	377	0.049	1.70	41.4	1.12	30.6	96
SX-26	5.1	14	3411	5150	0.397	91	316	0.077	1.96	a	a	25.2	131
SX-27	6.0	14	3485	5275	0.422	100		0.080	1.84	55.4	1.12	22.9	150
SX-28[b]	6.75	14	3470	5275	~ 0.400	86.6	465	0.078	1.40	48.5	1.13	41.0	70
SX-29	7.17	14	3402	5200	0.229	91.7	284	0.044	1.36	50.9	1.15	30.2	108

[a]Wet-test meter had corroded, giving inaccurate measurements.
[b]Initially the dissolvent was 4 M H_2SO_4; dissolver product (clad solution) flowed to acid feed tank and was recycled through dissolver.

Table 3. Sulfex Semicontinuous Cyclic Decladding and Core Dissolution

Fuel charge: Two prototype Consolidated Edison fuel assemblies (unirradiated)[a]

Decladding

Run No.	Time, hr	Dissolvent Composition	Dissolvent Flow Rate, liters/min	Dissolvent Temp., °C	Stainless Steel 2 assemblies + heel, g	Stainless Steel Dissolved g	Stainless Steel Dissolved %	Heel SS + braze, g	F/S, cm/min	Reaction[b] Rate, mg/cm²·min	Product Vol, liters	Product H_2SO_4 M	Product SS g/liter	Product NO_3, M	Losses to Clad Solution, % Th	Losses to Clad Solution, % U
SX-30	2.8	4 M H_2SO_4	0.797	105-107	3976	3852	96.9	64	0.078	2.27	133	2.91[c]	28.9	-	0.022	0.017
SX-32	4.0	4 M H_2SO_4	0.480	105-101	4240	4022	94.8	158	0.047	1.64	115	3.07	35.0	0.004	0.08	0.10
SX-34	3.3	4 M H_2SO_4	0.613	105-101	4434	4066	91.7	308	0.060	2.00	122	3.53	33.4	~ 0.001	0.2	0.02
SX-36	3.9	4 M H_2SO_4-0.1 M HCOOH	0.661	105-101	4364	4056	92.9	248	0.065	1.70	156	3.60	26.1	~ 0.001	0.25	0.04

Core Dissolution

Run No.	Time, hr	Dissolvent[d] Composition	Dissolvent Vol, liters	Dissolvent Flow Rate, liters/min	Core UO_2-ThO_2 + heel, g	Core Dissolved Thorium g	Core Dissolved Thorium %	Core Dissolved Uranium g	Core Dissolved Uranium %	Heel[e] UO_2-ThO_2 g	Reaction[b] Rate, mg/cm²·min	Product Composition H^+ M	Th, g/ℓ	U, g/ℓ	$SO_4^=$, M	Cr. M	Ni, M	Fe, M	F^-, M	PO_4, M	B, M	Cd, M	Al. M
SX-31	9.5	13 M HNO_3-0.05 M F^--0.12 M Al	93	0.60	11614	8561	89.6	468	88.8	1341	1.9	10.0	83	4.53	0.006	0.001	0.01	0.003	0.048	0.0001			0.11
SX-33	9.5	13 M HNO_3-0.048 M F^--0.11 M Al-0.056 M B	63	1.0	12955	9643	91.4	557	95	1343	2.2	9.6	141	7.57	0.001	0.002	0.011	0.005	0.05	0.002	0.055		0.11
SX-35	10	13.4 M HNO_3-0.04 M F^--0.12 M Al-0.06 M B	66	1.43	12957	8924	85	478	90	2260	1.9	9.71	126	6.76	0.001	0.006	0.01	0.006	0.04	0.005	0.05		0.11
SX-37	10	12.7 M HNO_3-0.04 M F^--0.10 M Al-0.06 M B-0.025 M Cd	65	1.43	13874	9075	80	508	80	2971	2.05	9.4	137	7.7	0.007	0.006	0.01	0.008	0.04	0.005	0.06	0.021	0.10

[a] Each fuel assembly consists of thirty-six 23-in.-long x 5/16-in.-o.d. x 0.020-in.-wall-thickness 304L stainless steel tubes filled with UO_2-ThO_2 pellets (Davison Chemical Co.), 4.2% UO_2-95.8% ThO_2, $\rho \approx 9.6$ g/cc, H_2 annealed; wt of stainless steel ≈ 2025 g; outer surface area ≈ 5100 cm²; tubes brazed together, using two rows of ferrules.

[b] Reaction rates calculated from weight of material dissolved and initial outer surface area of tubing and pellets; area of heel not considered.

[c] Values probably low because of jetting dilution.

[d] Designated volume of dissolvent recycled through dissolver.

[e] Weight of heel based on known weight charged to dissolver and analysis of core solution; actual weight of heel at end of SX-37 was 2058 g, of which ~1686 g was UO_2-ThO_2 pellet fragments (3.6 of total weight charged).

3.1 Clad Dissolution Rates

Stainless steel clad dissolution rates, R $mg/cm^2 \cdot min$, were calculated from the rate equation

$$R = W/At$$

where W is the weight of metal dissolved, mg (generally a run was continued until the fuel was completely dissolved); A is the initial surface area, cm^2; and t is the dissolution time, min. Average rates were then correlated with the ratio of the dissolvent flow rate to initial fuel element surface area, F/S cm/min.

The stainless steel dissolution rate increased proportionately to an increase in F/S, i.e., to an increase in the dissolvent flow rate (Fig. 2). The plate dissolution rate increased from 1.45 to 3.0 $mg/cm^2 \cdot min$ as the F/S was increased from 0.021 to 0.043 cm/min in 4 M H_2SO_4 and from 0.75 to 3.1 $mg/cm^2 \cdot min$ as the F/S was increased from 0.014 to 0.055 cm/min in 6 M H_2SO_4. The tubing (1/2 and 5/16 in. o.d.) dissolution rate in 4 M H_2SO_4 increased from 1.2 to 2.25 $mg/cm^2 \cdot min$ as the F/S was increased from 0.034 to 0.08 cm/min. The average rate of the plate was higher in 4 M than in 6 M H_2SO_4, which may be partially explained by the inverse relation of the solubilities of iron, nickel, and chromium sulfates to hydrogen ion concentration.[5]

Although the fuel charges consisted of metal of identical chemical and physical properties, dissolution was not uniform from one piece of metal to the next or even in all parts of the same piece of metal. The difference was more pronounced in tubing than in plate, and the reaction rate and dissolver product metal loading were lower for tubing than for plate under comparable operating conditions. In all cases the fuel dissolved completely after a sufficient amount of acid was added. In processing irradiated fuel assemblies, leaving of a stainless steel heel, which could contain some intact fuel rods, could lead to difficulty in charging additional fuel.

The nonuniformity of dissolution is shown in Figs. 3 and 4. Figure 3 shows the 8% heel (284 g) remaining at the end of run SX-29 (Table 2) in which the fuel charge consisted of empty 3-ft sections of 0.5-in.-o.d. x 0.035-in.-wall-thickness welded 304L stainless steel tubing. Two tubes are still the original length. All the tubes were from the same heat. The sections of tubing have a "barber pole" appearance with alternating dark and light stripes, penetration being slightly greater in the dark sections. Figure 4 shows the dissolver contents after 2.7 hr in 4 M H_2SO_4. Of the 40 prototype pins (0.5 in. o.d. x 16 in. long x 0.024 in. wall thickness and 0.020-in. end caps filled with UO_2 pellets) charged, 23 remained essentially intact. Some of the end caps were missing and there was only slight attack on the side walls. The decladding was subsequently successfully completed, indicating a partial rather than a total passivation. Fourteen of the 40 pins had been twice heated to 1500°F and water quenched in an unsuccessful attempt to shatter the pellets. They were held at temperature for 3 hr the first time and for 16 hr the second.

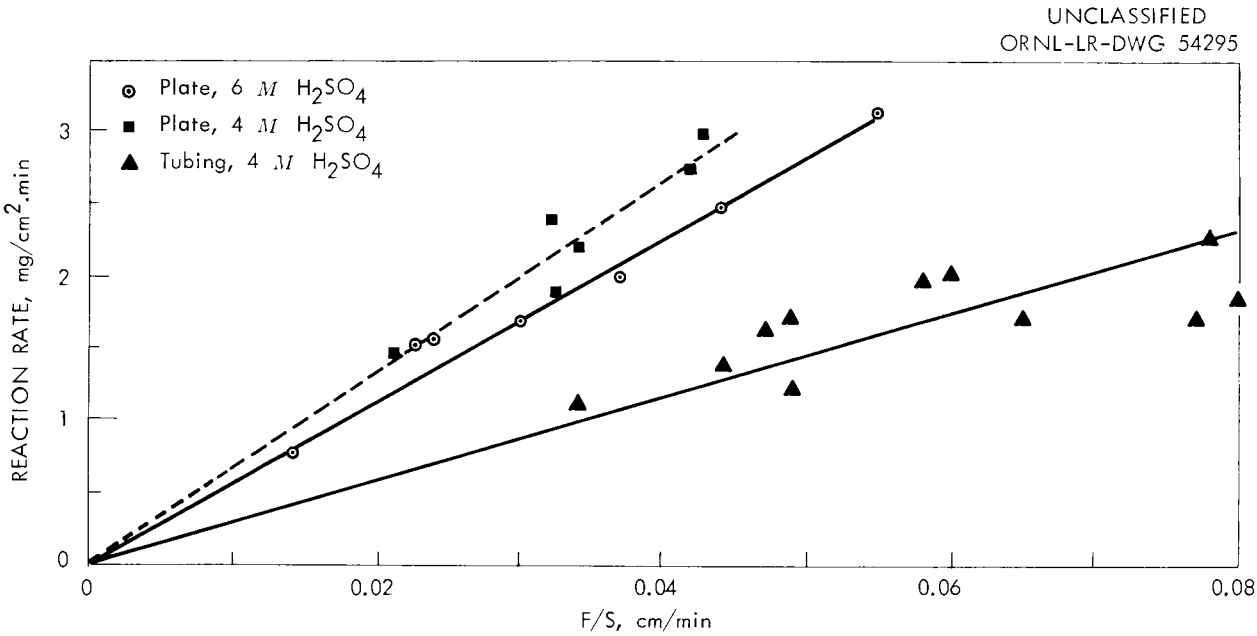

Fig. 2. Reaction rate as a function of F/S (dissolvent flow rate/initial surface area) in semicontinuous dissolution of 304 stainless steel plate and tubing in 4 M and 6 M H_2SO_4.

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Fig. 3. Dissolver contents at the end of Run SX-29, showing the non-uniformity in dissolution of "identical" pieces of 304 stainless steel tubing.

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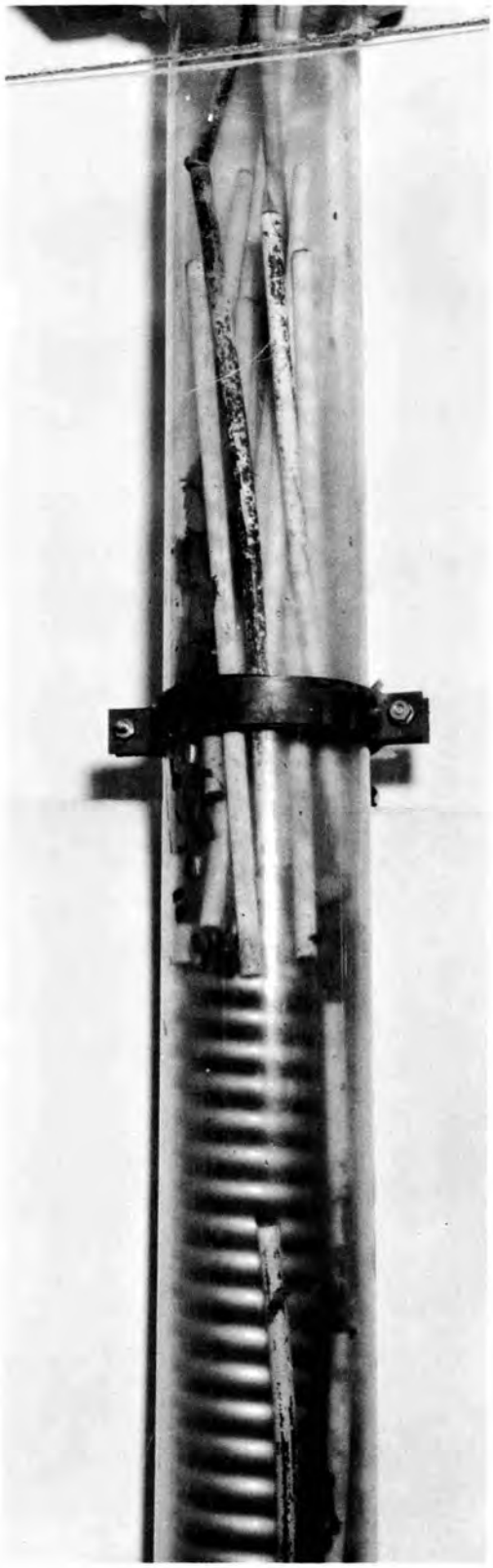

Fig. 4. Undissolved 304L stainless steel tubes at the end of a 2.7-hr semicontinuous dissolution run with 4 *M* H_2SO_4.

3.2 Hydrogen Evolution

The instantaneous hydrogen evolution rate can be used to follow the dissolution rate, and the accumulated amount of hydrogen can be used to calculate the weight of metal dissolved at any given time. Approximately 1.1-1.2 g moles of hydrogen was liberated per gram mole (55.4 g) of stainless steel dissolved (Tables 1 and 2), which agrees very well with the amount (1.1 moles) expected from the dissolution of 18-8 stainless steel in sulfuric acid:

$$Fe + H_2SO_4 \rightarrow FeSO_4 + H_2$$

$$2Cr + 3H_2SO_4 \rightarrow Cr_2(SO_4)_3 + 3H_2$$

$$Ni + H_2SO_4 \rightarrow NiSO_4 + H_2$$

The instantaneous hydrogen evolution rate in general decreased throughout a dissolution run (Fig. 5), but there were fluctuations, showing the non-uniformity of dissolution. If all plates dissolved uniformly over the entire length, the surface area and dissolution rate would remain essentially constant until the final instant in which the plates were completely dissolved. The integrated form, or the accumulated hydrogen evolved, plotted as a function of elapsed time for the dissolution of stainless steel plate in 6 M H_2SO_4 is a smooth curve (Fig. 6).

For comparison purposes, the instantaneous hydrogen evolution rate, accumulated hydrogen evolved, and instantaneous dissolver product metal loadings as a function of elapsed time for typical runs in which stainless steel plate was dissolved in 4 and 6 M H_2SO_4 are presented in Figs. 7 and 8 and dissolution of stainless steel tubing in 4 M H_2SO_4 in Fig. 9. The fluctuations in the instantaneous hydrogen evolution rate were much greater in the dissolution of tubing than of plate, with the rate increasing 6- to 14-fold in a 15- to 30-min period. The initial instantaneous hydrogen evolution rate of 17.8 slpm for tubing (Fig. 9) corresponds to an instantaneous reaction rate of 3.8 $mg/cm^2 \cdot min$ based on the initial outer surface area of the assemblies (10,200 cm^2). The series of plateaus in the accumulated hydrogen curve for tubing shows that flattening of the curve does not necessarily indicate completion of clad dissolution. Therefore the weight of cladding charged to the dissolver should be known fairly accurately so that the total hydrogen evolved would be an indication of the amount of clad dissolved at any given time based on the generation of 1.1-1.2 g moles of hydrogen per gram mole of stainless steel dissolved.

3.3 Dissolver Product Metal Loading

The instantaneous dissolver product metal loading followed the same general trend as the instantaneous hydrogen evolved (Fig. 10) and was inversely proportional to the dissolvent flow rate. The composite metal loading was equal (mg/ml) to the slope of the average reaction rate vs the F/S line (Fig. 2).

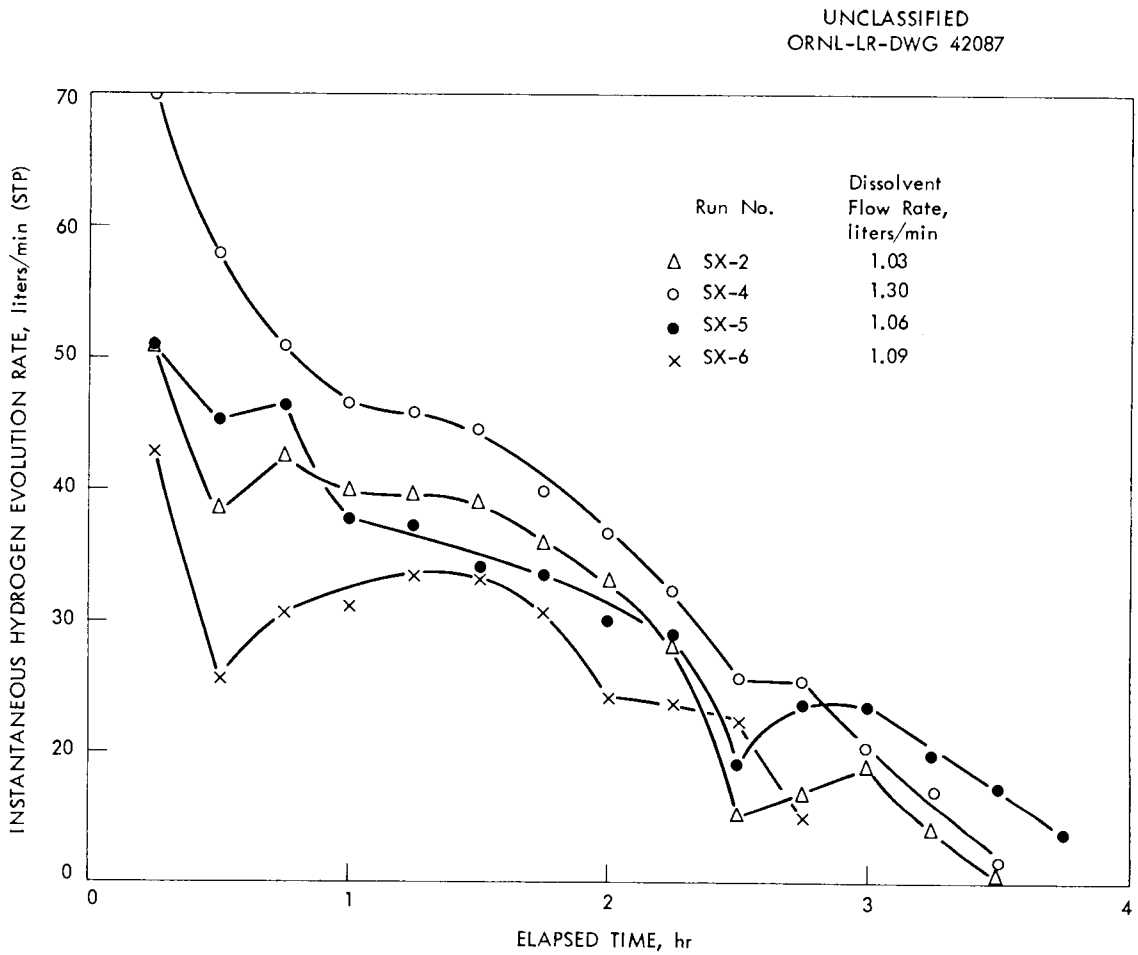

Fig. 5. Instantaneous hydrogen evolution rate as a function of time for semicontinuous dissolution of stainless steel plate using 6 *M* H_2SO_4.

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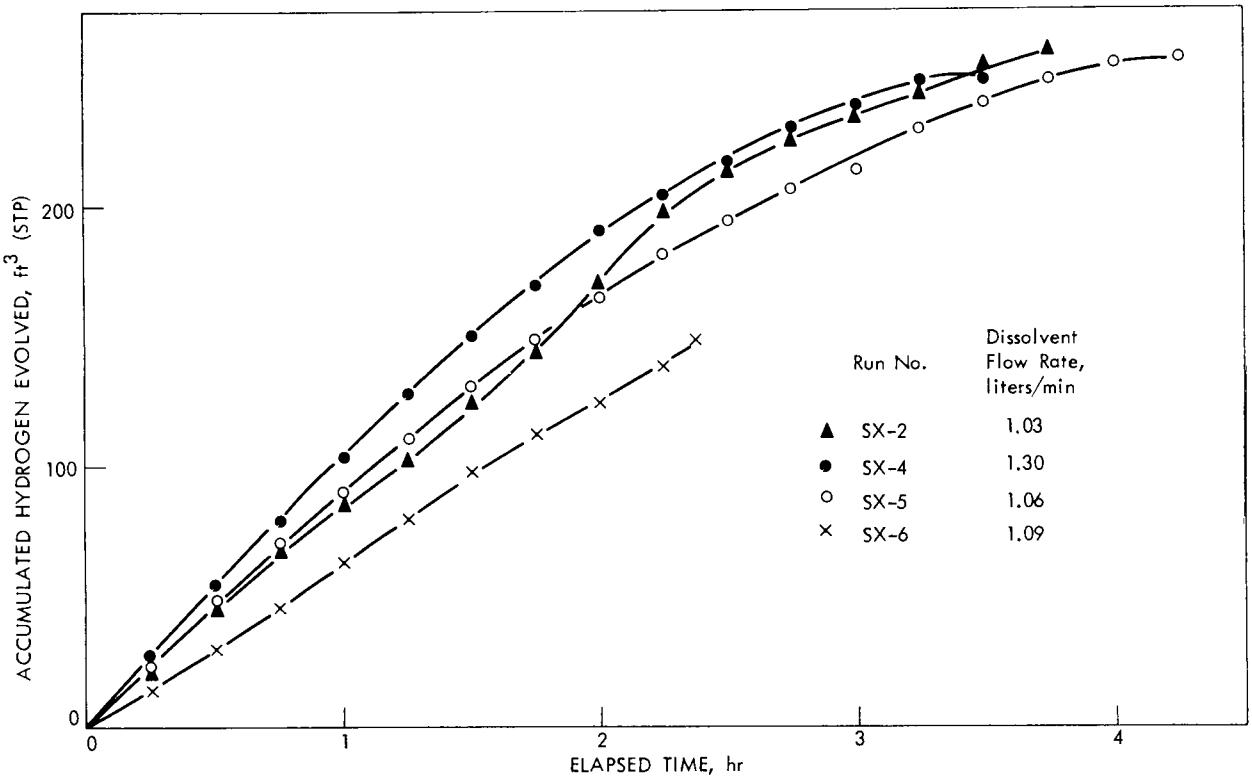

Fig. 6. Accumulated hydrogen evolution as a function of time for the semicontinuous dissolution of 304L stainless steel plate in 6 M H_2SO_4.

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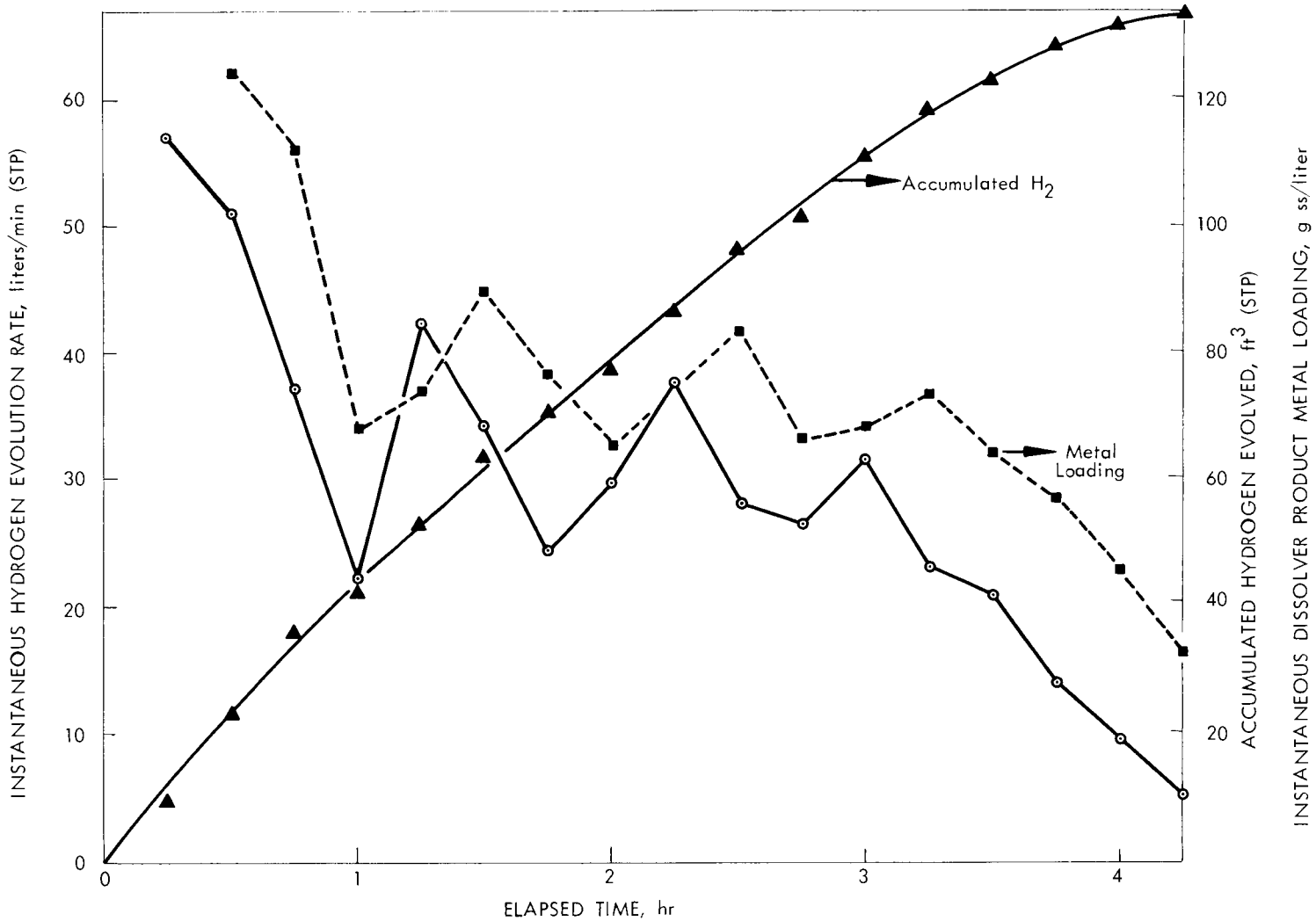

Fig. 7. Instantaneous Hydrogen evolution rate, accumulated hydrogen, and instantaneous dissolver product metal loading as a function of time in semicontinuous dissolution of 304 stainless steel plate in 4 *M* H_2SO_4.

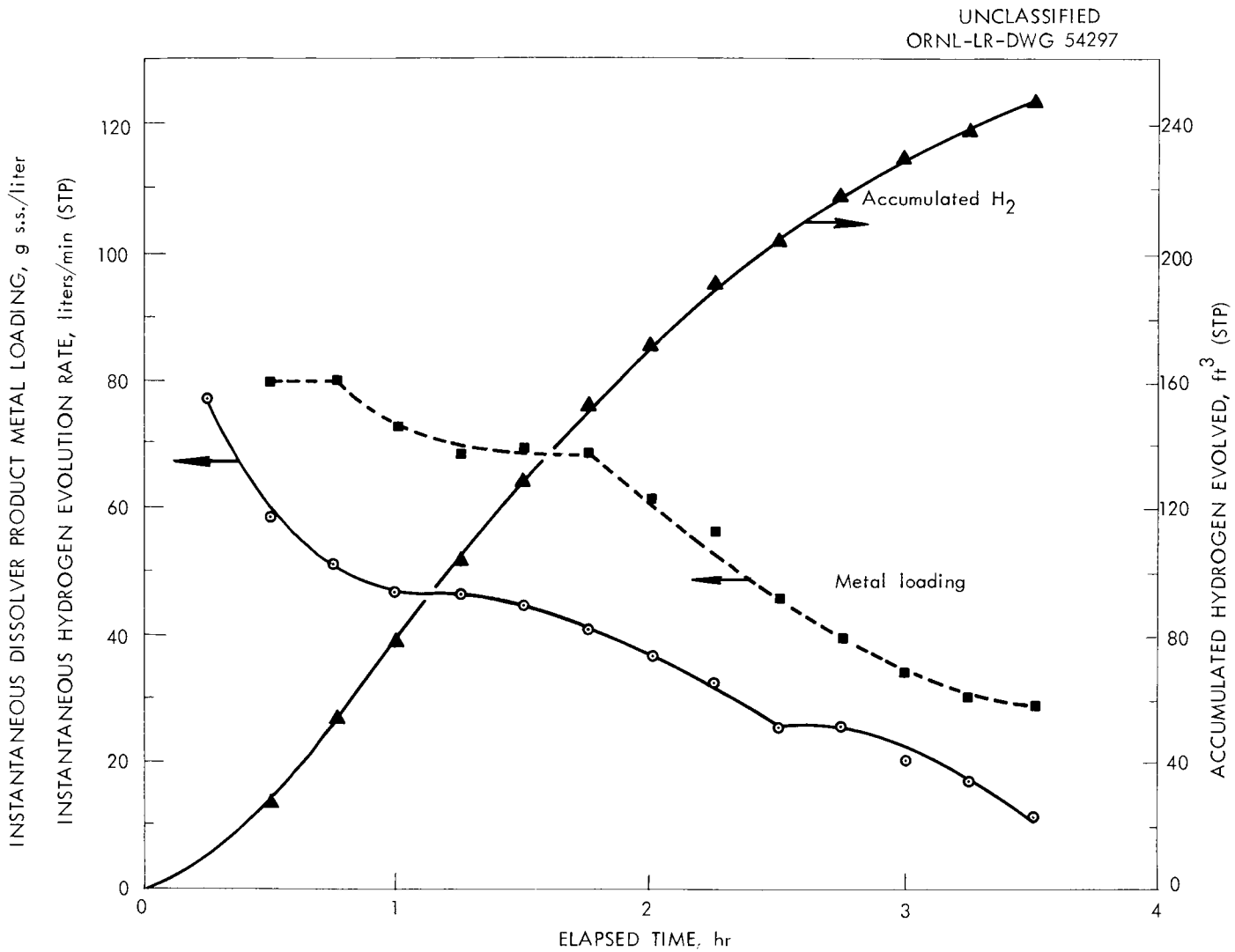

Fig. 8. Instantaneous hydrogen evolution rate, accumulated hydrogen, and instantaneous dissolver product metal loading as a function of time in semicontinuous dissolution of 304 stainless steel plate in 6 *M* H_2SO_4.

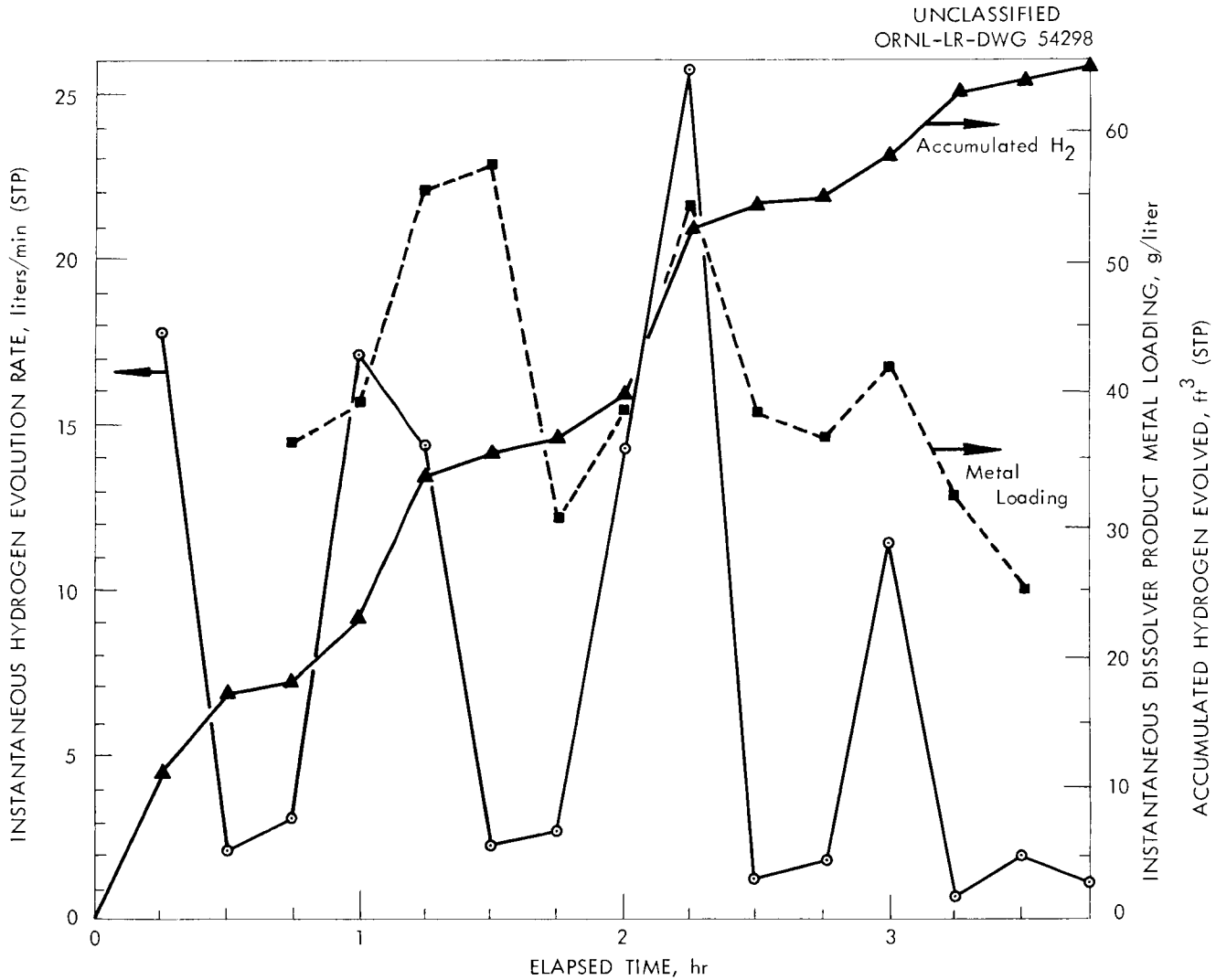

Fig. 9. Instantaneous hydrogen evolution rate, accumulated hydrogen, and instantaneous dissolver product metal loading as a function of time in semicontinuous decladding of prototype Consolidated Edison fuel assemblies (stainless steel-clad UO_2-ThO_2 Pellets) with 4 *M* H_2SO_4.

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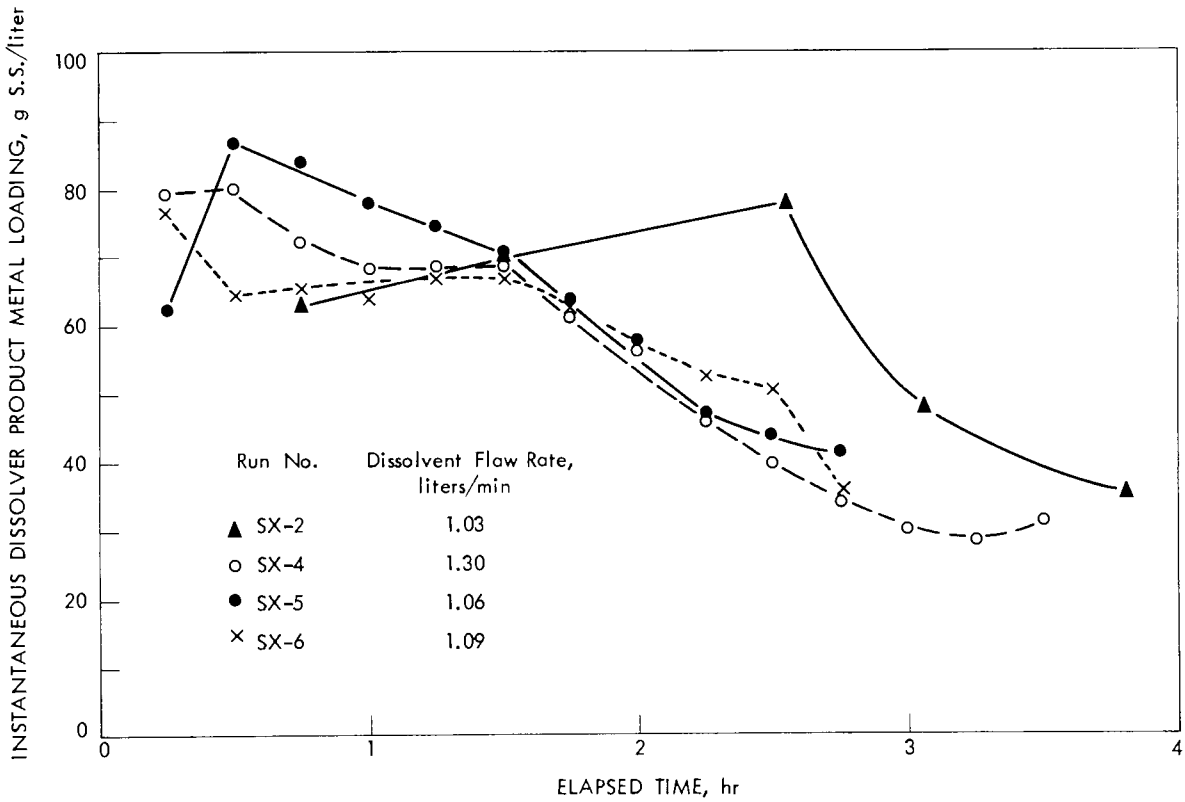

Fig. 10. Dissolver product metal loading as a function of time for semicontinuous dissolution of 304L stainless steel plate in 6 M H_2SO_4.

For a given dissolvent composition the composite dissolver product metal loading was insensitive to variations in dissolvent flow rate. Average loadings were 67 g/liter for the dissolution of plate in 4 M H_2SO_4, 57 g/liter for plate in 6 M H_2SO_4, and 29 g/liter for tubing in 4 M H_2SO_4.

Attempts to markedly increase the product loading by varying the dissolvent flow rate within a given run (runs SX-9 and 10, Table 1), combination batch and semicontinuous dissolution (run SX-11, Table 1), and recycling the dissolvent (run SX-28, Table 2) were unsuccessful. In run SX-28 (Table 2), the dissolver was charged with 304L stainless steel tubing (0.5 in. o.d. x 0.035 in. wall thickness), and sufficient 4 M H_2SO_4 to give a product stainless steel loading of 50 g/liter, based on total dissolution of the tubing, was recycled through the dissolver. After 6.75 hr, approximately 87% of the fuel had been dissolved and dissolution had essentially ceased. The instantaneous product metal loading became constant at approximately 43 g/liter (Fig. 11) after 4.5 hr dissolution. Consequently, complete dissolution could not have been accomplished in this particular run within a reasonable dissolution time. This is substantiated by a plot of metal dissolved (expressed as percent of original) vs time, which tends to be asymptotic (Fig. 12).

3.4 Decladding of Stainless Steel-clad UO_2 and Core Dissolution

A series of runs (SX-18 through 24) was made to determine dissolution rates and soluble uranium losses in chemical decladding by boiling 4 M H_2SO_4 of unirradiated prototype stainless steel-clad UO_2 fuel elements. In each run the fuel charge consisted of tubular 304L stainless steel-clad UO_2 plus additional plate stainless steel to provide a UO_2-to-stainless steel ratio of approximately 2.7 to simulate NMSR fuel. The fact that some of the cores were powdered instead of pelleted UO_2 and partial passivation of the clad presented operational difficulties, and the data from these runs are therefore not presented in tabular form. Partial passivation of the clad resulted in an approximately 2-fold increase in dissolution times. The UO_2 pellets dissolved readily in 8 M HNO_3.

Run SX-18. The first run was made with nine prototype 304L stainless steel-clad UO_2 fuel elements 66 in. long x 0.5 in. o.d. with approximately 25 mils wall thickness. Due to misinformation, it was assumed that the UO_2 was in the form of pellets. At the conclusion of the run, physical inspection of a similar fuel element showed the UO_2 as a finely divided powder. A Cintel particle size evaluation based on 510 particles measured showed 50% < 0.7 μ, 90% < 3 μ, and 100% < 16 μ.

Initially the clad dissolution proceeded smoothly, but upon penetration of the clad the dissolver solution immediately assumed a reddish-brown, muddy appearance. As the run progressed, the dissolver and auxiliary equipment became choked with the powder and a large quanity of it was carried out in the product stream. At the end of the clad dissolution, the UO_2 remaining in the dissolver settled to the bottom in a compacted mass in approximately 0.5 hr. The dissolver could not be emptied by either jetting or draining, and the UO_2 was flushed from the dissolver by fluidizing with a large quanity of water. It was estimated that the soluble uranium loss to the clad solution

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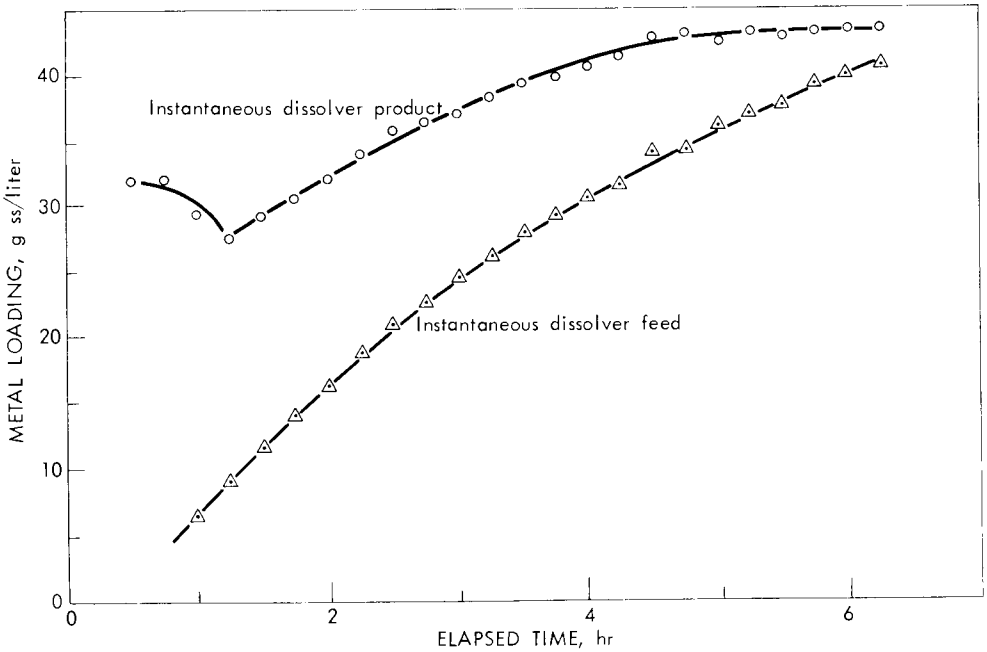

Fig. 11. Instantaneous metal loading of dissolver product and feed as a function of time in semicontinuous dissolution of welded stainless steel tubing (run SX-28) with recycling of the clad solutions.

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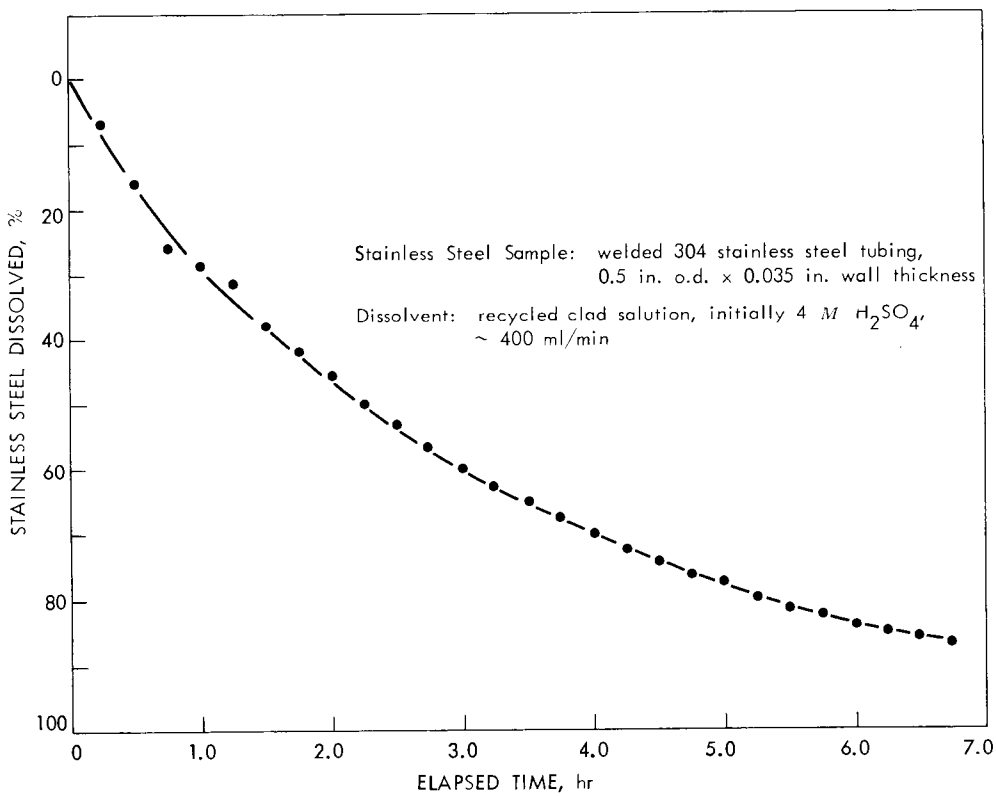

Fig. 12. Per cent dissolved as a function of time in semicontinuous dissolution of stainless steel tubing (run SX-28); clad solution recycled.

was approximately 1.3%, and it is evident from the difficulties encountered that a semicontinuous Sulfex process in the present equipment is not feasible for processing of fuel elements in which the core material is in a finely divided state.

Runs SX-19 and 21 were made with a fuel charge in each run of 40 prototype pins (0.5 in. o.d. x 16 in. long x 0.024 in. wall thickness and 0.020 in. end caps) filled with UO_2 pellets (0.370 in. o.d. x 0.375 in. long) with a density of 10.36 g/cm^3, plus 780 g of 38-mil 304L stainless steel plate and boiling 4 M H_2SO_4 as the dissolvent. The total weight of stainless steel was 9088 g and the weight of all UO_2 pellets was 11,157 g. In each run the plate dissolved completely but the cladding was partially passivated (Sect. 3.1). In the first run, after 2.7 hr dissolution, 23 pins remained essentially intact, and in the second, 11 pins remained after 6.4 hr. In both cases (run SX-20), the undissolved cladding was then batch dissolved using 6 M H_2SO_4 to initiate dissolution and diluted to 4 M H_2SO_4 in the first run and starting with 4 M H_2SO_4 in the second. Soluble uranium losses to the clad solution in the two runs were 0.04 and 0.05%, respectively.

Run SX-23. In the final decladding run, the dissolver charge consisted of 14 pieces (thirteen 4 ft long and one 2 ft long) of 0.5-in.-o.d. x 0.035-in.-wall-thickness 304L stainless steel tubing (total of 4.56 kg) filled with the UO_2 pellets left from run SX-22. The clad was dissolved in 5.25 hr at an average 4 M H_2SO_4 flow rate of 0.379 liter/min (F/S = 0.058 cm/min), resulting in an average reaction rate of 2.2 mg/cm^2·min. Soluble uranium loss to the clad solution was 0.0007%.

Core Dissolution. The UO_2 pellets from run SX-23 (11.2 kg) were completely dissolved in 3.7 hr in 20 liters of 8 M HNO_3 (run SX-22). Although the nitric acid was fed to the dissolver continuously (0.091 liter/min), the dissolver holdup was 20 liters so that the run was essentially a batch dissolution.

3.5 Dejacketing of Stainless Steel-clad UO_2-ThO_2

Four semicontinuous cyclic Sulfex decladding and Thorex core dissolutions of prototype Consolidated Edison fuel assemblies were made (Table 2). Decladding times varied from 2.78 to 4.0 hr as the ratio of dissolvent flow rate to outside surface area (F/S) was varied from 0.078 to 0.047 cm/min; corresponding average reaction rates varied from 2.27 to 1.64 mg/cm^2·min. The dissolver product metal loading was inversely proportional to the dissolvent flow rate, varying from 26 to 35 g/liter. The soluble thorium loss to the clad solution increased from 0.022% in the absence of a heel to 0.25% in the presence of a heel. Soluble uranium losses were fairly constant at 0.02-0.04% except for one run in which the loss was 0.1%. One fuel assembly that had been autoclaved 7 days at 300°C and 2000 psig (steam + O_2) to oxidize the surface behaved no differently from the unoxidized assemblies.

In each run the dissolver was charged with two fuel assemblies (Fig. 13), each assembly consisting of thirty-six 23-in.-long x 5/16-in.-o.d. x 0.020-in.-wall-thickness 304L stainless steel tubes brazed together, using two

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Fig. 13. Prototype (Davison Chemical Company) Consolidated Edison fuel assembly (stainless steel-clad UO_2-ThO_2 pellets).

rows of ferrules and Nicrobraze 50, filled with UO_2-ThO_2 pellets. The weight of the stainless steel was 2025 g and the outer surface area 5110 cm^2. Each assembly contained 11,614 g of Davison Chemical Company pellets (4.2% UO_2—95.8% ThO_2, density 9.6 g/cm^3, H_2 annealed). The clad dissolvent was 4 M H_2SO_4.

Clad Passivation. Passivation of the clad was encountered in runs SX-32 and 34. There was no dissolution of the clad in 15 min exposure to boiling 4 M H_2SO_4 (~10 liters of dissolvent fed continuously to the dissolver). When the dissolution reagent was removed and the clad contacted with fresh dissolvent, dissolution started immediately. Apparently the passivation was induced by excessive concentration of nitrate ion (0.05 M NO_3^- in the drained dissolvent) remaining from the preceding core dissolution run.[6] In run SX-36 the dissolvent was spiked with 0.1 M HCOOH and dissolution started as soon as the boiling dissolvent contacted the fuel assemblies.

The nitrate residue was in spite of the following thorough washing procedure: At the end of each decladding and core dissolution run, the dissolver was emptied and then washed with two batches of water of ~3.5 liters each. Each batch of wash water was boiled ~15 min to flush out the system. Instantaneous nitrate concentrations varied from an initial 0.015 M to a final 0.0005 M and the average concentration in the clad solution was 0.001-0.004 M.

Core Dissolution. Average rates of core dissolution in 13 M HNO_3—0.04 M F^-—0.1 M Al^{3+} were fairly consistent, 1.9-2.2 mg/cm^2·min (Table 3) with dissolvent recycle flow rates of 0.6-1.43 liters/min. Reaction rates were calculated from the weight of pellets dissolved in a given run (based on analysis and volume of dissolvent) and the surface area of the pellets charged to the dissolver prior to the run. The area and weight of the heel from the previous run were not considered.

In each core dissolution run, the dissolvent was recycled through the dissolver to increase the thorium and uranium loadings in the final product. Each run was continued until there was little or no change in dissolver product and density as a function of time. Equilibrium was reached in 9 hr (Fig. 14) with 80-95% of the core dissolved (based on analysis of product). The data are qualitative, but they illustrate the trend in the semicontinuous dissolution of UO_2-ThO_2 pellets with recycled dissolvent.

At the end of run SX-37 the residue was removed from the dissolver and separated into three fractions: -20 mesh fines, UO_2-ThO_2 pellet fragments, and stainless steel and braze metal fragnemts (Fig. 15). The total weight of residue was 2038 g consisting of 329 g fines, 1686 g UO_2-ThO_2 fragments (3.6% of total weight of pellets charged to the dissolver in the four cyclic runs), and 43 g of stainless steel and braze metal fragments (0.26% of total weight of stainless steel and braze). The fines consisted primarily of particles of stainless steel and braze metal and carbonaceous material formed during dissolution of stainless steel in sulfuric acid. It is important to note that the actual weight of the UO_2-ThO_2 heel was 1686 g, compared to the 2971 g estimated from analyses of core solutions (Table 3). There was no visual evidence of an appreciable heel buildup from one run to the next.

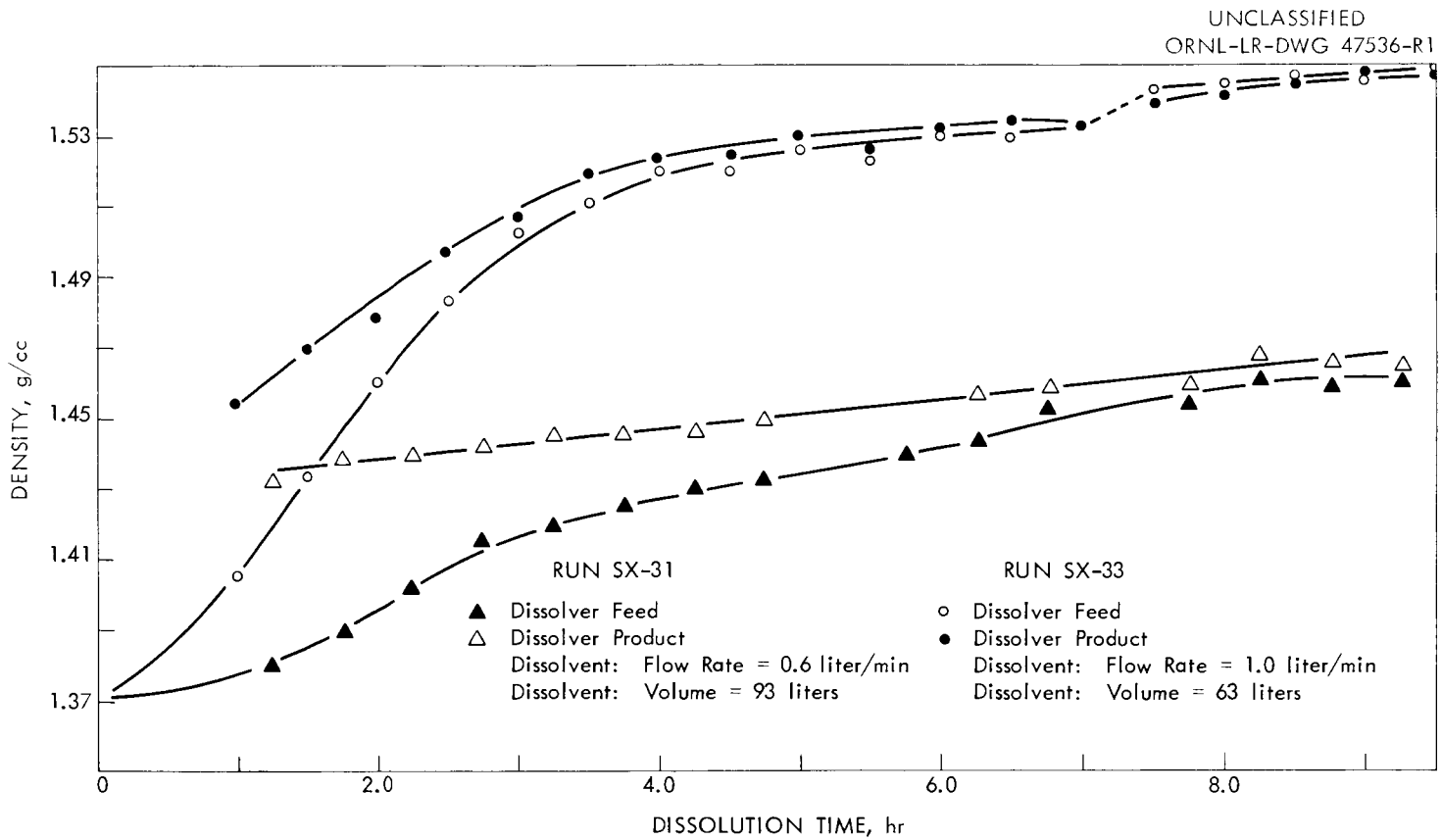

Fig. 14. Density of dissolver feed and product as a function of dissolution time in semicontinuous dissolution of UO_2-ThO_2 pellets (Davison 4.2% UO_2-95.8% ThO_2, $\rho \sim 9.6$ g/cc, H_2 annealed); dissolvent recycled.

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Fig. 15. Dissolver residue at end of run SX-37.

The total residue was batch dissolved in 300% excess dissolvent. After ~24 hr refluxing, 178 g of residue, consisting of 62 g of fines, 100 g of UO_2-ThO_2 fragments, and 16 g of stainless steel and braze metal fragments, remained. The 100 g of UO_2-ThO_2 fragments represented approximately 0.02% of the total weight (46,456 g) of UO_2-ThO_2 pellets charged to the dissolver.

When the dissolver was drained at the end of decladding run SX-30 a "honeycomb" section of ferrules and braze metal was undissolved (Fig. 16). Essentially all the "honeycomb" was dissolved in the subsequent core dissolution run (SX-31), and there was some contamination of the core solution with chromium, nickel, iron, and phosphorus. The concentration of $SO_4^{=}$ ion in the core solution varied from 0.001-0.007 M. A "honeycomb" section as such did not exist at the end of runs SX-32, 34, and 36 but there were individual pieces of undissolved braze metal and ferrules.

Criticality Control. Criticality control of solutions in the Fuel Processing Pilot Plant by using boron and/or cadmium as nuclear poisons has been proposed. The dissolvent was "spiked" with boron in run SX-33 and 35 and with both boron and cadmium in run SX-37. The data indicate that neither boron nor cadmium is lost through volatilization or precipitation during core dissolution.

3.6 Solution Clarification

In the dejacketing step small amounts of solid stainless steel corrosion products remain in the clad solution with metal loadings > 25 g/liter. Dense fuel oxide particles are entrained in both clad and core solutions, the amount depending on the degree of fuel fragmentation and the hydraulics of the dissolver, and, in the case of core dissolution, on the dissolution rate. Particulate fuel oxides must be removed from both solutions, and waste solids must be removed from the core solution before solvent extraction processing. The nature of the solids removed by both gravity sedimentation and centrifugation were briefly investigated.

Amount of Solids. The volume of centrifuge waste cake from stainless steel solution ranged from 0.2 to 0.5% of the total solution volume (Table 4); the weight was 1.3 to 2.7% of the weight of stainless steel plate dissolved. The total silicon content of the solution, which indicates the approximate precision in measurements of total weight and composition of waste solids, varied from 0.0028 to 0.0057 g/g SS vs a specified < 0.01 g/g SS for 304L.

Prototype CETR elements processed by the Sulfex-Thorex decladding-core dissolution cycle yielded centrifuged solids of fairly consistent quantity and composition from run to run (Table 5). The volume of waste solids produced was not measured; however, the weight ratio of waste clad solids produced to stainless steel dissolved ranged from 1.3 to 1.9%, which agrees fairly well with values obtained for plate stainless steel.

Solids Particle Size. A very small amount of particulate ThO_2-UO_2, of the order of 10 g, was collected from all streams of each fuel element run.

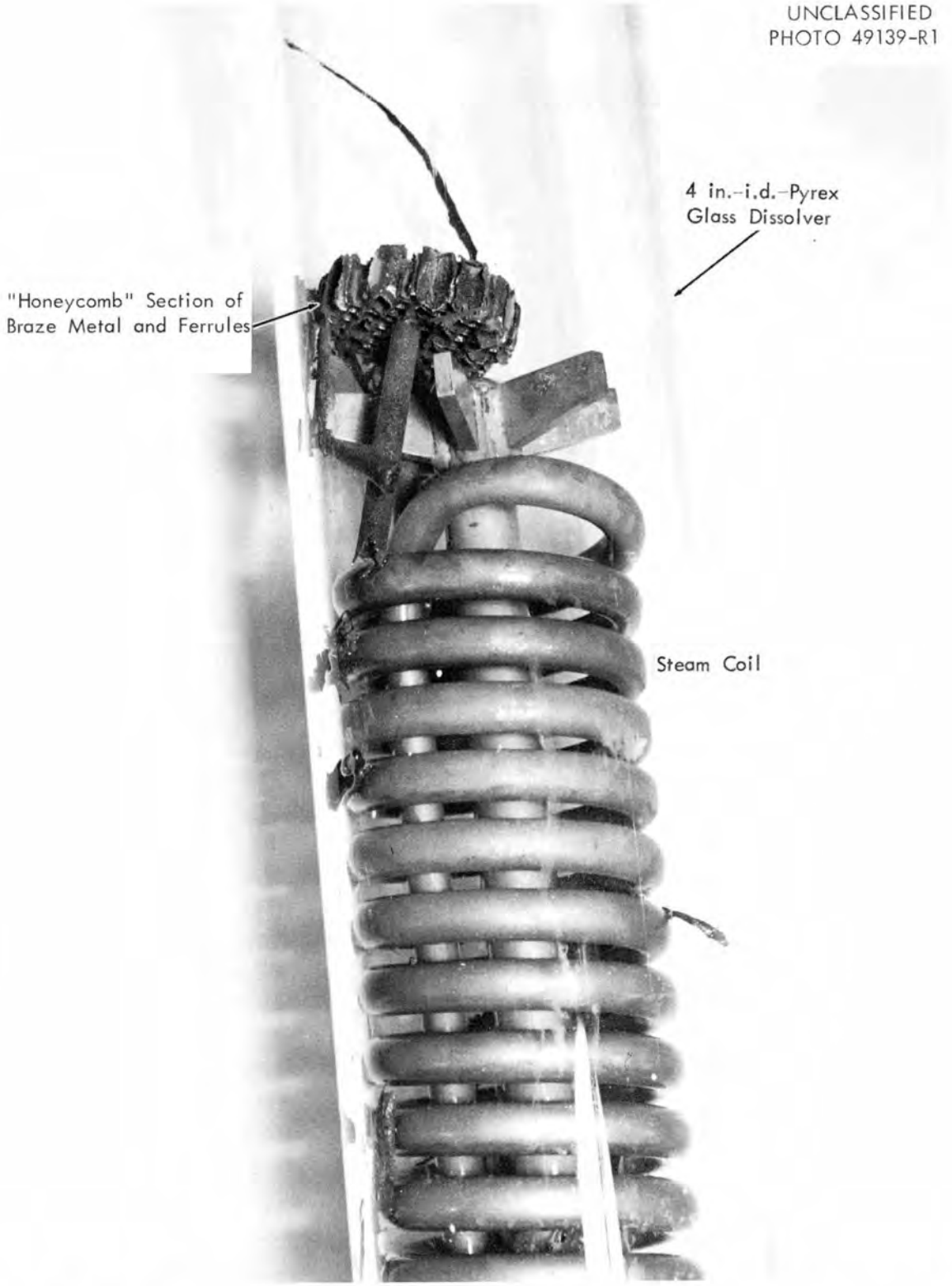

Fig. 16. Undissolved "honeycomb" section of braze metal and ferrules at end of decladding run SX-30.

Table 4. Waste Solids Remaining After Dissolution of 304L Stainless Steel Plate in 4 M H_2SO_4

SX Run No.	Waste Solids: Amount: Wet Solids,[a] ml/liter product	Dry Solids: g/liter product[b]	Dry Solids: g/g SS	Composition (Dry), %: Fe	Composition (Dry), %: Si	Silicon Balance, g/g SS: Soluble	Silicon Balance, g/g SS: Total[c]
12	4.5	2.0	0.027	9	8	0.00326	0.0054
13	5.0	1.4	0.022	15	19	0.00144	0.0056
14	5.0	0.83	0.013	22	20	0.00324	0.0057
15	4.0	1.5	0.022	14	6	0.00338	0.0047
16	2.5	1.0	0.017	16	2	0.00377	0.0042
17	2.0	0.89	0.015	7	2	0.00351	0.0038

[a]Based on centrifugation of 100 ml of mixed product in clinical tube centrifuge (1200 G); precision of measurement ± 20%.

[b]Solids from 100-ml sample dried at 110°C.

[c]A.I.S.I. specification for 304L sheet is 1.00% Si max.

Table 5. Waste Solids and Particulate ThO_2-UO_2 Remaining After Sulfex-Thorex Cyclic Decladding and Core Dissolution

Run No.: Decladding	Run No.: Core Dissolution	Dry Waste Solids: Weight: Total,[a] g	Dry Waste Solids: Weight: g/liter clad solution[b]	Dry Waste Solids: Weight: g/g SS	Dry Waste Solids: Composition, %: Fe	Dry Waste Solids: Composition, %: Si	Wt of Particulate ThO_2-UO_2, g: From Clad Solution	Wt of Particulate ThO_2-UO_2, g: Total
30	31	56	0.42	0.015	17	3.5	-	-
32	33	74	0.64	0.019	20	9.7	3.8	12
34	35	54	0.44	0.013	18	7.5	3.0	4.5
36	37	60	0.39	0.015	22	6.0	6.3	9.3

[a]Total weight of waste solids recovered by clarification of all streams in a 12-in. solid-bowl centrifuge. Of the total, 75-80% of the waste solids came from the Sulfex stream, 20-25% from the Thorex stream.

[b]Total weight of waste solids from both streams divided by Sulfex volume only.

Composition of Solids. The composition of SS corrosion product solids (SX-32) was reported[7] as:

SiO_2	- 28%
Cr	- 14%
Fe	- 10%
Cu	- 9%
P	- 4%
Ni	- 2%
Mn and Mo	- 0.1 to 1%
Ti	- 0.01 to 0.1%

While composition of the solids varied somewhat from run to run, in general the total waste solids produced in a run contained more iron and less silica than shown here (Tables 4 and 5). Determination of copper by neutron activation indicated 14 wt %, in substantial agreement with the above. The copper probably resulted from corrosion of Ni-O-Nel or Carpenter 20 materials of construction, both of which contain 2-3% copper. If true, a moderate increase in the quantity of waste solids produced in an all-metal dissolver system might be expected, since the subject dissolver was largely glass.

Magnetic solids, found in most solid waste samples, were shown by x-ray diffraction to be magnetic iron oxide, Fe_3O_4. The mean particle size of waste solids in the sulfuric acid clad solution was 3 to 7 μ (Fig. 17a), as determined on a few samples by neutron activation-sedimentation and Cintel flying-spot microscopy. One or two points on the size distribution curve inferred from run data on settler operation and centrifuge operation, assuming Stokes' law to hold, agree approximately with the other results. Samples a and b (Fig. 17a) are known to be biased toward the larger size particles because of the necessity of drying them prior to irradiation.

Because size analysis by sedimentation in the Sulfex liquor was believed to give more useful results than centrifugal classification or flying-spot microscopy, subsequent samples were analyzed by the neutron activation-sedimentation technique. Samples were activated in the wet condition. Separate samples of "slurry" collected in the horizontal settler and centrifuge during run SX-32 were analyzed (Fig. 17b), with qualitative results in line with those expected. Under run conditions all 10-μ particles should have been collected in the horizontal settler, and 70% of the solids in the settler were larger than 10 μ. Conversely, 70% of the solids collected by the centrifuge were smaller than 10 μ. The mean particle size for the combined solids is, by coincidence, also 10 μ. This compares fairly well with the 3- to 7-μ figure above, although the size distribution varied substantially even between similar runs. The flow rate, weight of waste solids produced, etc., were virtually the same for runs SX-32 and SX-36, but the mean particle sizes of solids collected in the centrifuge were 4.9 and 1.4 μ, respectively (Fig. 17b).

Application to Process. If the solid waste particle could be sufficiently degraded in size by some method, it might be possible to separate and recover entrained UO_2 or ThO_2-UO_2 by a sedimentation technique (e.g., gravity settling, centrifugation, hydraulic cyclone) while simultaneously rejecting the waste

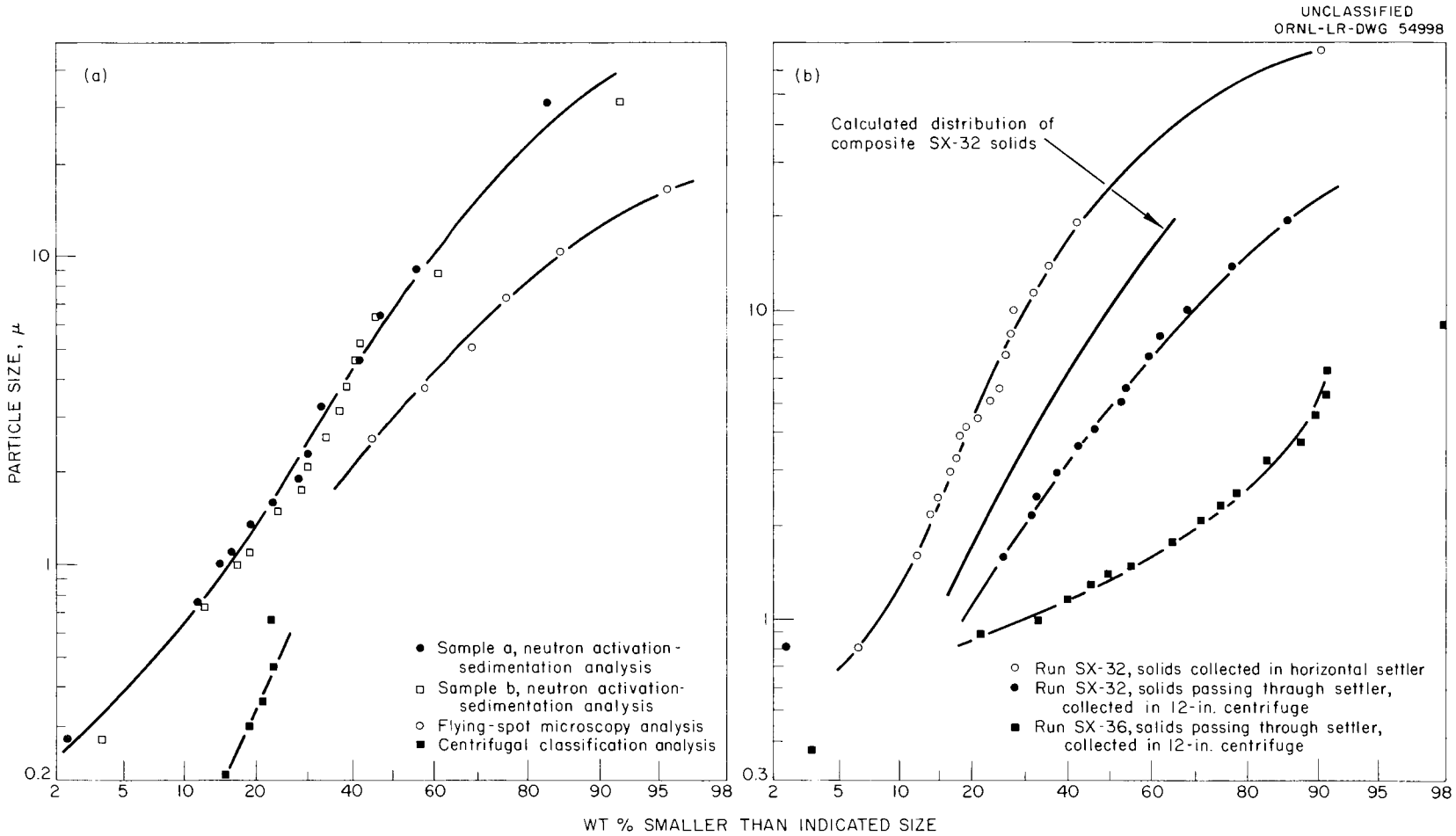

Fig. 17. Particle size distribution of waste solids produced in (a) Sulfex dissolution of stainless steel plate (run SX-13) as determined by three independent methods, and (b) Sulfex dejacketing of prototype CETR assemblies (runs SX-32, 36), as determined by neutron activation-sedimentation. Assumed specific gravity of solids: 5.

solids with the waste Sulfex liquor. An attempt to measure the extent to which waste solids might be degraded in size, by circulation through a steam jet-hydroclone combination, gave inconclusive results. Centrifuged SX-36 solids subjected to this treatment had a mean particle size of 7 μ, compared to 1.4 μ for the starting material (Fig. 17b). The anomaly may be due to incomplete collection of solids after the size degradation attempt.

For the Sulfex system (viscosity ~2-3 cp, ρ_ℓ ~1.2-1.3 g/cm^3, ρ waste = 5 g/cm^3 assumed) the ratio of the equal settling diameter of waste solids to that of UO_2 is approximately 1.5, by Stokes' law. Therefore, on the basis of waste solids size distribution shown in Fig. 17b, all UO_2 particles > 66 μ might be collected by a device that would reject all waste solids which are < 100 μ.

An alternative approach for disposal of waste solids is complete clarification of all streams and recovery of uranium and thorium values by leaching, followed by dissolution of waste residue and disposal as a solution. However, attempts at complete dissolution of waste solids by aqueous reagents were unsuccessful; chemical fusion was the only method shown to permit total dissolution.[7]

4.0 REFERENCES

1. M. E. Brennan et al., "Fuel Recovery Process," KAPL-933 (July 17, 1953).

2. J. R. Flanary et al., "Development of the Sulfex Process for Decladding Stainless Steel-Clad Power Reactor Fuel Elements with Sulfuric Acid," ORNL-2461 (March 13, 1959).

3. L. M. Ferris, "Decladding of Consolidated Edison Power Reactor Fuel by Sulfex and Darex Processes: Cyclic Dissolution Experiments," ORNL-2822 (Jan. 13, 1960).

4. W. D. Bond, "Dissolution of Sintered Thorium-Uranium Oxide Fuel in Nitric Acid-Fluoride Solutions," ORNL-2519 (Oct. 28, 1958).

5. L. M. Ferris, ORNL, unpublished data.

6. T. A. Gens, "Sulfex Process: Depassivation of Stainless Steel," ORNL-2785 (Nov. 6, 1959).

7. L. M. Ferris, A. H. Kibbey, and K. S. Warren, "Dissolution of Insolubles from Darex and Sulfex Processes," ORNL CF 60-6-70 (June 14, 1960).

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