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QUARTERLY TECHNICAL PROGRESS REPORT
FUELS, MATERIALS, AND COOLANT
CHEMISTRY PROGRAMS
JANUARY-MARCH 1970

MASTER

AEC Research and Development Report

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Printed in the United States of America
Available from

Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

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The preceding Quarterly Technical Progress Report
was AI-AEC-12913

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CONTRACT: AT(04-3)-701
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CONTENTS

<u>AEC Task No.</u>	<u>Project Title</u>	<u>Page</u>
10-A	Fuel Cladding and Structural Materials	5
10-C	Mechanical Properties, Static Sodium	11
10-D	Dynamic Loop Testing	23
2-A	Irradiation Damage in Cladding and Core Structural Materials	57
2-B	Integrated (n, α) Cross-Sections by Helium Determinations	67
11	Mixed Carbide Fuels	75
12-A	Sodium Chemistry	85
12-B	Fission-Product Contamination	89

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Program:	Fuel Cladding and Structural Materials		
AEC Task:	10-A, Materials Management		
Project Manager:	J. J. Gill		
Reporting Period:	January-March 1970		
General Order:	6507	Subaccount:	29511-2
		AEC Category:	04-01-61-03.1

Principal Investigators: E. W. Murbach and D. F. Atkins

I. PROJECT OBJECTIVES

The objectives of this project are to: (1) manage a source of stock "systems-quality" sodium for the program; (2) monitor the compositions of sodium and of gas atmospheres in use on all the subtasks in the program, and provide a management system for ensuring the use of proper procedures in sampling, transferring, and analyzing of sodium; and (3) procure, characterize, and manage a supply of cladding materials for the program.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

To date, 20 retorts have been loaded with sodium for Subtask 10-C.

The supply loop was operated with the cold-trap isolated for 23 days. The oxygen meter was quite stable during this period, showing that there was no leakage of oxygen into the loop.

A few ppm of hydrogen raised the plugging temperature of the supply loop to approximately 500°F. This amount of hydrogen did not interfere with oxygen determination by mercury amalgamation.

A new lot of 316 stainless-steel tubing (0.250-in. OD by 0.010-in. wall) was purchased to replenish the existing stock. This tubing was fabricated from the reference heat of alloy.

Tubing from three heats of 316 stainless steel was solution-annealed to provide test material for comparison with the as-received cold-worked condition.

A small quantity (120 ft) of FFTF preproduction cladding (316 stainless steel, 20% cold-work) was purchased for mechanical-property studies on other tasks of the program.

III. PROGRESS DURING REPORT PERIOD

During most of this quarter the sodium supply loop has been operated as an experimental facility. In the first experiment the cold-trap was operated between 235 and 280°F (loop temperature 500°F) to check the oxygen-meter response. The results are shown in Table 1.

TABLE 1
OXYGEN-METER READING vs
COLD-TRAP TEMPERATURE

Cold-Trap (°F)	Oxygen Meter (volts)
235	1.166
250	1.162
260	1.159
270	1.155
280	1.151

Assuming the oxygen concentration is equivalent to the cold-trap temperature and taking the resultant oxygen values from AI-AEC-12685, the equation for $\log C$ vs E (where C is ppm oxygen and E is the emf in volts) has a slope of -0.038. The theoretical slope of the oxygen meter at 500°F is -0.053.

An experiment was carried out to determine the effect of added hydrogen on the plugging temperature of the loop. There have been reports from other sites in which two plugging breaks have been observed in sodium systems. It has been postulated that the upper break is due to hydrogen. The oxygen content of the supply loop is normally quite low, probably about 1 ppm, so any change in plugging temperature should be due to the added hydrogen.

With the loop operating at 500°F the cold-trap was valved out of the system and hydrogen equivalent to about 10 wppm was added to the cover-gas over the surge tank. Pressure in the surge tank rose to 3 psi but slowly decreased as the hydrogen dissolved. A sharp plugging break was observed at 465°F. The temperature of the loop was increased to 600°F and the plugging temperature increased to 525°F as more hydrogen apparently dissolved.

Removal of the hydrogen was started by pumping on the gas space over the surge tank. At the start of the removal process, pressure at the pump was 100 microns. After several days of pumping, the pressure at the pump decreased to less than 1μ . A gradual decrease in plugging temperature to 460°F was noted. At this time the rate of hydrogen-removal had become quite low so the cold-trap was returned to service. After cold-trapping for 24 hr at 250°F the plugging temperature had decreased to 208°F .

This experiment has demonstrated that H_2 in Na does show a distinct plugging break. In fact large amounts of H_2 would completely mask an oxide-plugging break. In some of the plugging runs the orifice was completely plugged at 450 to 500°F .

The loop was sampled twice during the experiment, once the day of the initial addition and once during the H_2 -removal process. During the period of 600°F operation the reading of the oxygen meter decreased nearly 20 millivolts. The second sample did show an increase in O_2 level of about 4 ppm. The source of this oxygen is not clear; however, it is probably due to the release by the stainless steel of previously gettered oxygen.

The O_2 level of the first sample was normal, less than 5 ppm as determined by mercury amalgamation, showing that hydrogen does not interfere in the analysis as any H_2 remaining as sodium hydride would indicate a large increase in O_2 . For example, 1 ppm of H_2 as NaH would give an apparent O_2 content of 8 ppm. The available information gives the heat of formation of NaH as ~ 13 kcal and that of NaHg_4 as ~ 20 kcal. Although the free energies are not available they should not be much different from the heats of formation; therefore, it is logical that Hg would decompose sodium hydride with the liberation of H_2 gas so that no interference in the analysis would result.

An experiment is in progress to study the transfer of impurities from the flowthrough cold-trap to a diffusion cold-trap. A vessel of about one-liter capacity was attached to a sample station, the flow started, and the vessel lower end cooled to about 210°F . The temperature of the cold-trap was raised to 300°F and then the cold-trap was valved out of the system. The reading of the oxygen meter dropped about 15 millivolts and then slowly increased about 4 mv in a week. After the cold trap was valved into the system, its temperature

was raised to 325°F which resulted in another drop in the emf of the oxygen meter. With the cold-trap isolated, the oxygen-meter reading is slowly increasing which indicates that impurities are slowly being gettered by the diffusion cold-trap. At a cold-trap temperature of 325°F the amount of oxygen added to the loop is about 5 ppm. The flow through the diffusion cold-trap is less than 0.5 gpm so that the combination of low O₂ and low flowrate would result in slow removal of O₂.

During the quarter four retorts were loaded with Na for Subtask 10-C. In addition, two vessels were loaded with Na for the reactor safety program.

Sampling of the loops and retorts was continued on a routine basis. With the exception of one retort sample which analyzed 30 ppm of oxygen (for no apparent reason) all analyses were normal.

Inspection and characterization steps were completed on a recent shipment of 488 ft of 316 stainless-steel tubing (0.250-in. OD by 0.010-in. wall). The tubing was found to be of the same high quality as previous lots fabricated from the same heat.

A shipment of 120 ft of FFTF preproduction cladding (316 stainless steel, 20% cold-work) was received for mechanical-property studies in high-temperature sodium. Following inspection, 20 ft were issued for helium-injection studies and 10 ft supplied for biaxial creep tests in static sodium. The remainder was placed in controlled storage.

IV. EVALUATION OF EFFORT TO DATE

Due to the relatively light schedule on sodium loading, some interesting experiments with the supply loop were possible. The behavior of hydrogen in an otherwise clean sodium system is pertinent.

The oxygen-meter electrode is still functioning well after about 22 months of operation which is encouraging from the standpoint of reactor operation.

V. NEXT REPORT PERIOD ACTIVITIES

Routine operation of the supply loop will continue and sodium will be dispersed as needed.

It is expected that the program will be reoriented, so other effort will depend on new planning and scheduling.

Routine control, release, and inventory of characterized tubing will be carried out as needed.

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Program: Fuel Cladding and Structural Materials

AEC Task: 10-C — Mechanical Properties, Static Sodium

Project Manager: J. J. Gill

Reporting Period: January-March 1970

General Order: 06507

Subaccount: 29530

AEC Category: 04-01-61-03.1

Principal Investigator: D. F. Atkins

I. PROJECT OBJECTIVES

Objectives of this project are to determine the individual and combined effects of environment (sodium and helium), strain rate (10^{-7} to 10^{-3} /hr), and stress condition (uniaxial and biaxial) on the creep and stress-rupture characteristics of Type-304 stainless steel and Type-316 stainless-steel cladding alloys.

II. MAJOR ACCOMPLISHMENTS DURING FY 1970

Studies of the influence of heat-to-heat behavior on the biaxial stress-rupture of 316 stainless-steel tubing were started. Twenty-four specimens from two new heats of tubing were tested in 1200°F sodium and the results compared with the reference heat of tubing.

State-of-stress studies were completed on 316 stainless-steel tubing in 1200°F sodium. Under the stress modes of uniaxial, 2:1 biaxial, and 1:1 biaxial the stress-rupture life was found to correlate best with the maximum principal stress. Similar studies in 1000°F sodium are in progress.

Studies on the effect of sodium cleaning and subsequent storage on the stress-rupture behavior of 316 stainless-steel tubing were begun. Four 12-specimen retorts containing cold-worked 316 stainless-steel tubing were placed on test at 1200 and 1000°F. The specimens will be soaked in high-temperature sodium for 2000 hours before the cleaning and storage operations.

Scoping studies were completed on the biaxial stress-rupture performance of Sandvik 12R72HV stainless-steel tubing in high-temperature sodium.

III. PROGRESS DURING REPORT PERIOD

Biaxial stress-rupture tests on 304 and 316 stainless-steel tubing continue to provide data on the long-term performance of these alloys in high-temperature sodium. Thirteen retorts, each containing twelve tubular specimens, are on test at 900, 1000, and 1200°F. The status and test conditions are given in Table 1. In addition to the multiple specimen retorts which impose a 2:1 biaxial-stress ratio on the specimens, single-specimen sodium retorts are used to study alloy performance under uniaxial and 1:1 biaxial-stress modes. Table 2 lists the details of the more recent tests.

Four 12-specimen retorts were assembled, loaded with sodium, and placed on test. The specimens (cold-worked 316 stainless-steel tubing) will be soaked in sodium at 1000 and 1200°F for 2000 hr. Four of the twelve specimens in each retort are under a moderate hoop stress (50% of the stress required for 1% strain in 250,000 hr). The stressed specimens will show if there is a stress-induced effect during sodium exposure on subsequent cleaning and storage operations. After sodium exposure the specimens will be removed and subjected to a variety of cleaning, handling, and storage treatments. The specimens will then be returned to static sodium for biaxial stress-rupture testing. The use of thin-walled tubing (0.010-in. wall) will provide a high sensitivity for detecting subtle effects which may be hidden by other more conventional methods of evaluation. A comparison of the stress-rupture properties of the sodium-cleaned specimens with extensive base-line data already in existence will give an evaluation, in practical terms, of the effectiveness of the cleaning and storage procedures.

A retort containing twelve specimens of Sandvik 12R72HV tubing (0.015-in. wall by 0.240-in. OD) completed biaxial stress-rupture test in 1400°F static sodium. Diametral strain measurements were calculated and are listed in Table 3. Four of the specimens were in the solution-annealed condition (1 hr at 1100°C) while the remaining eight contained about 15% cold-work (as-received condition). The large reduction in rupture strain due to cold-work is apparent in Table 3. The strength and rupture strain for the annealed material is about the same as for annealed 316 stainless steel at the same temperature. In the cold-worked condition the Sandvik alloy exhibits greater strength but considerably less rupture strain than 10 to 15% cold-worked 316 stainless-steel tubing under

TABLE 1
BIAXIAL STRESS-RUPTURE TESTS, 12 SPECIMENS PER RETORT

Retort Number	Test Temperature (°F)	Alloy	Clad Condition*	Environment	Life Span (hr)	Status		Remarks	Dates Start/Completed
						Test Complete	Examination Complete		
17	1200	304 SS	Ann	Sodium	5000 to 10,000	3/4 Complete		Extension of data	2-22-68
18	1200		Ann		100 to 5000	x	x	Effect of cold-work	1-25-68/8-18-68
19	1000		AR		5000 to 10,000	x		Extension of data	4-2-68/5-12-69
20	900		AR + Ann		100 to 7100	x		Scoping test	12-1-67/9-22-68
21	1000		Ann		100 to 2300	x	x	Effect of cold-work	2-15-68/5-20-68
22	1000		Ann		5000 to 10,000	3/4 Complete		Extension of data	2-29-68
23	1200		AR + Na Exp		100 to 5000	x	x	Effect of flowing sodium	8-22-68/1-6-69
24	1300		AR	Na + Cu	100 to 1000	x	x	Effect of copper	3-26-68/5-15-68
25	900		Ann	Na	1000 to 10,000	3/4 Complete		Extension of data	10-8-68
26	900		AR	Na	1000 to 10,000	1/2 Complete		Extension of data	3-27-69
27	1000		AR	He	100 to 3000	x	x	Effect of sodium	3-4-69/6-29-69
110	1000	316 SS	AR	Sodium	1000 to 10,000	3/4 Complete		Extension of data	4-23-68
111	1000		Ann		1000 to 10,000	Discontinued		Extension of data	5-10-68/6-4-68
112	900		Ann		100 to 5000	x	x	Extension of data	6-20-68/3-12-69
113	900		AR		100 to 10,000	In Progress		Extension of data	8-16-68
114	1200		AR + Aged		100 to 3400	x	x	Effect of thermal aging	10-29-68/6-21-69
115	1000		AR		100 to 5000	3/4 Complete		Effect of sodium	10-18-68
116	1000		AR + Aged		100 to 5000	3/4 Complete		Effect of thermal aging	11-5-68
117	1200		Ann		1000 to 5700	x		Extension of data	12-4-68/8-1-69
119	1000		Ann		1000 to 3000	x	x	Extension of data	12-11-68/4-14-69
120	900		Ann		1000 to 10,000	In Progress		Extension of data	12-20-68
121	1000		Ann		100 to 5000	x	x	Effect of sodium	6-5-69/12-5-69
122	1200		AR		100 to 1000	x		Heat-to-heat study	8-29-69/1-6-70
123	1200		AR		100 to 1000	x		Heat-to-heat study	8-29-69/11-28-69
SV-2	1400	Sandvik 12R-72	AR + Ann		100 to 2000	x	x	New-alloy scoping	11-30-69/1-9-70
126	1200	316 SS	AR	Sodium	2000 Soak	In Progress		Cleaning study	12-29-69
127	1000		AR						1-8-70
128	1200		AR						1-15-70
129	1000		AR						2-19-70

*AR = as received tubing containing 10 to 15% cold-work.
Ann = annealed tubing.

AI-AEC-12948
13

TABLE 2
SINGLE-SPECIMEN STATIC RETORT TESTS

Test No.	Test Temperature (°F)	Alloy	Clad Condition*	Environment	Stress (psi)	Rupture Time (hr)	Dates
							Started/Completed
1:1 Biaxial Stress Rupture							
619B	1200	316 SS	Ann	Sodium	27,000	76	6-4-69/6-7-69
620B	1200	↓	AR	↓	27,000	657	6-11-69/7-9-69
628B	1000	↓	AR	↓	48,000	1364	9-19-69/11-15-69
629B	↓	↓	AR	↓	52,000	661	9-23-69/10-21-69
630B	↓	↓	AR	↓	50,000	685	9-30-69/10-29-69
631B	↓	↓	Ann	↓	48,000	376	11-12-69/11-28-69
Uniaxial Stress Rupture							
621U	1200	316 SS	AR	Na	27,000	705	6-11-69/7-11-69
622U	↓	↓	AR	Na	30,000	361	6-12-69/6-27-69
623U	↓	↓	AR	Na	29,000	442	6-20-69/7-15-69
625U	↓	↓	AR	He	27,000	738	7-21-69/8-21-69
626U	1000	↓	AR	Na	50,000	1162	9-30-69/11-18-69
627U	↓	↓	AR	Na	48,000	748	9-17-69/10-20-69
632U	↓	↓	Ann	Na	48,000	567	11-26-69/12-20-69
633U	↓	↓	AR	Na	54,000	664	12-30-69/1-27-70
634U	↓	↓	Ann	Na	54,000	-	2-27-70
635U	↓	↓	Ann	Na	42,000	412	2-27-70/3-17-70

*AR = as-received tubing containing 10 to 15% cold-work.
Ann = annealed tubing.

the same test conditions. Over the same range of rupture life the 316 stainless-steel tubing exhibits 5 to 10% strain. Average diametral strain rates were calculated by dividing strain at rupture by rupture life. The values are listed in Table 3 and plotted as a function of stress level in Figure 1.

Metallographic studies were made on the Sandvik 12R72HV specimens after stress-rupture test. A thin layer of fine-grain structure about 0.001 in. in depth was observed on the ID of the tubing. This structure is shown in Figure 2. The same duplex structure was also observed on the as-received tubing. Because of the location and small amount of this fine structure it probably had little influence on the stress-rupture properties of the tubing. The fine-grained subsurface structure is believed to be caused by surface contamination, possibly from a carbonaceous lubricant or solvent, during the tubing fabrication.

The microstructure of the annealed Sandvik 12R72HV tubing is shown after stress-rupture test in Figure 3. The grain size of the alloy was somewhat greater after the annealing treatment (1 hr at 1100°C). The elongated grain structure and thinner-wall cross-section is a reflection of the much larger strain observed on the annealed specimens.

Rupture of both the cold-worked and annealed tubing occurred by intergranular cracking. Extensive grain-boundary voids, particularly in the cold-worked tubing, developed in the vicinity of the ruptures. Figure 4 shows a typical rupture of the tubing wall.

Diametral strain measurements were obtained after rupture on 24 specimens from the two new heats of 316 stainless-steel cladding. As shown in Tables 4 and 5 the rupture strain on Heat 20303, the stronger of the two, was considerably lower than on 03574. This behavior suggests that the cladding from Heat 20303 contains a greater amount of cold-work. Since both heats were purchased from the same tube mill to the same tube-fabrication specification the strain results reflect the variation in cold-work which can be obtained when a 1/8 hard condition is specified. Average strain-rate values were calculated and plotted against stress level as shown in Figure 5. The curve for the reference heat of cladding is also shown for comparison. The steeper slope generated by the data from Heat 20303 is believed to be a reflection of greater cold-work. However, testing on solution-annealed material from these two heats must be done before the influences of heat chemistry and cold-work can be separated.

TABLE 3

DIAMETRAL-STRAIN MEASUREMENTS ON SANDVIK 12R72HV TUBING
STRESS-RUPTURE TESTED IN 1400°F SODIUM

Specimen No.	Hoop Stress (psi)	Rupture Life (hr)	Diametral Strain (%)	Diametral Strain Rate (%/hr)
SV-1*	20,000	<8	22.13	2.76
SV-2*	18,000	8	24.22	3.03
SV-3*	17,000	18	19.48	1.08
SV-4*	16,000	96	24.90	2.59×10^{-1}
SV-5	16,000	777	1.18	1.51×10^{-3}
SV-6	16,000	690	1.14	1.65×10^{-3}
SV-7	17,000	672	1.39	2.07×10^{-3}
SV-8	17,000	577	1.73	3.00×10^{-3}
SV-9	18,000	307	1.90	6.19×10^{-3}
SV-10	18,000	417	0.88	2.11×10^{-3}
SV-11	20,000	288	0.55	1.91×10^{-3}
SV-12	20,000	310	0.63	2.03×10^{-3}

*Solution-annealed specimens; all other specimens contained ~15% cold-work.

Figure 1. Strain-Rate Sensitivity to Stress, Sandvik 12R72HV
Tubing in 1400°F Sodium

AI-AEC-12948

Marble's Etch 8644-3-3 250X

Figure 2. Cross-Section of Sandvik 12R72HV Tubing (15% cold-work) After Stress-Rupture Test in 1400°F Sodium (Note fine-grain structure on ID. Black areas are grain-boundary voids. Hoop stress, 17,000 psi; rupture life, 577 hr)

Marble's Etch 8644-1-2 250X

Figure 3. Annealed Sandvik 12R72HV Tubing After Stress-Rupture Testing in 1400°F Sodium (hoop stress, 16,000 psi; rupture life, 96 hr)

Marble's Etch

8644-3-4

250X

Figure 4. Stress-Rupture Failure in Wall of Sandvik 12R72HV Tubing (15% cold-work). Tested in 1400°F Sodium Under 17,000-psi Hoop Stress

Figure 5. Heat-to-Heat Comparison, Strain-Rate Sensitivity to Biaxial Stress

During stress-rupture testing in static-sodium retorts sodium samples are taken during loading, at the midpoint of the test, and after all specimens have ruptured. The oxygen content in the sodium was compiled for all retorts used since the start of the program. The values are plotted as a function of test time at test temperature in Figure 6. No distinction was made as to the sodium temperature. The sodium temperatures were 900, 1000, 1100, 1200 and 1400°F. A computer analysis of the data points was made using linear-regression techniques. The least-squares fit, using the linear transform of an exponential function, yields a horizontal line about 9-ppm oxygen (Figure 6).

Figure 6. Static Sodium Retort Oxygen Level as Function of Test Hours

TABLE 4
DIAMETRAL-STRAIN MEASUREMENTS ON 316 STAINLESS-STEEL
TUBING (10 to 15% COLD-WORK), HEAT 20303, STRESS-
RUPTURE TESTED IN 1200°F SODIUM

Specimen No.	Hoop Stress (psi)	Rupture Life (%)	Diametral Strain (%)	Diametral Strain Rate (%/hr)
T6-1	27,000	2158	1.64	7.60×10^{-4}
-2	↓	2119	1.43	6.75×10^{-4}
-3	↓	2119	1.47	6.94×10^{-4}
-4	30,000	1531	1.90	1.24×10^{-3}
-5	↓	1484	1.57	1.06×10^{-3}
-6	↓	1484	1.74	1.17×10^{-3}
-7	32,000	1248	1.84	1.47×10^{-3}
-8	↓	1230	1.94	1.58×10^{-3}
-9	↓	1202	2.24	1.86×10^{-3}
-10	35,000	720	2.07	2.87×10^{-3}
-11	↓	549	1.37	2.49×10^{-3}
-12	↓	776	2.24	2.86×10^{-3}

To provide information on the influence of stress mode on the stress-rupture behavior of 316 stainless-steel tubing, three uniaxial tests in 1000°F sodium were started. One specimen of cold-worked (10 to 15%) tubing failed after 664 hr under a stress of 54,000 psi. An annealed specimen under the same stress ruptured after 412 hr. Both specimens exhibited very low strain (1.5 to 2.0%). The third test with annealed tubing continues at a stress of 42,000 psi. A 900-hr rupture life is predicted for this specimen.

The fabrication of 24 specimens of FFTF 316 stainless-steel preproduction cladding (20% cold-work) was completed. The OD of each specimen was measured with a precision profilometer. Twelve of the specimens will be assembled in a sodium retort for creep studies under conditions of incremental stress buildup on the cladding wall. This work will supply support information for the strain model of the FFTF fission-gas plenum.

TABLE 5
DIAMETRAL-STRAIN MEASUREMENTS ON 316 STAINLESS-STEEL
TUBING (10 to 15% COLD-WORK), HEAT 03574, STRESS-
RUPTURE TESTED IN 1200°F SODIUM

Specimen No.	Hoop Stress (psi)	Rupture Life (hr)	Diametral Strain (%)	Diametral Strain Rate (%/hr)
K6-1	27,000	3080	5.47	1.77×10^{-3}
-2	↓	2997	3.39	1.13×10^{-3}
-3	↓	3069	4.73	1.54×10^{-3}
-4	30,000	1187	7.42	6.25×10^{-3}
-5	↓	1051	6.58	6.26×10^{-3}
-6	↓	1329	6.98	5.25×10^{-3}
-7	32,000	559	11.45	2.05×10^{-2}
-8	↓	601	9.77	1.62×10^{-2}
-9	↓	635	7.32	1.15×10^{-2}
-10	35,000	215	6.58	3.06×10^{-2}
-11	↓	242	7.08	2.92×10^{-2}
-12	↓	233	9.16	3.93×10^{-2}

IV. EVALUATION OF EFFORT TO DATE

The experimental program in static-sodium retorts continues to provide important information on the stress-rupture behavior of 316 stainless steel. Recent studies on cold-worked tubing in 1000°F sodium have shown that the alloy has quite low ductility at this temperature. Only 1 to 2% strain has been observed on the uniaxial and 1:1-biaxial stress-rupture tests. Very little tertiary-creep strain occurs at 1000°F whereas at 1200°F tertiary creep contributed to a major portion of the total rupture strain. These observations are very significant to design personnel who often must use data generated at higher temperatures to design of components for service in the 800 to 1000°F range.

V. NEXT REPORT PERIOD ACTIVITIES

Twelve specimens of FFTF 316 stainless-steel tubing will begin test in 1200°F sodium to study strain behavior with incremental increases in hoop stress.

Metallographic examinations will be made on recently tested stress-rupture specimens to evaluate structure in relation to stress level, rupture life, and fracture mode.

Additional single-specimen sodium retorts will be assembled to study the uniaxial and 1:1-biaxial stress-rupture behavior of 316 stainless-steel cladding at 1000°F.

Program: Fuel Cladding and Structural Materials

AEC Task: 10-D, Dynamic Loop Testing

Project Manager: J. J. Gill

Reporting Period: January-March 1970

General Order: 6507

Subaccount: 29540

AEC Category: 04-01-61-03.1

Principal Investigator: J. H. Shively

I. PROJECT OBJECTIVES

Project objectives are to (1) study, evaluate, and analyze the effects of flowing-sodium environment on the mechanical behavior of stainless steel, and (2) provide a design basis for the combined effects of mass transfer, temperature, heat flux, biaxial stresses, and load cycling on the creep-rupture behavior of fuel cladding in a flowing-sodium system.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

A 19-pin hexagonal array of wire-wrapped 0.250-in. cladding was constructed and tested for 2613 hr in 1200°F flowing sodium this year. A ΔT leg modification was built and installed on the 2-in. isothermal loop to accommodate the 19-pin array. Each tube in the array was individually pressurized. The combined effects of mass transfer and contact forces from wire wrap on biaxial creep and stress-rupture behavior of cold-worked Type-316 stainless steel were studied.

Two 12-pin static-sodium retorts containing individually pressurized tubes of cold-worked Type-316 stainless steel were built and put on test. One test on 0.295-in.-OD cladding has been completed. The other on 0.250-in.-OD cladding continues. The purpose of the tests is to provide a base-line to which the data obtained in the ΔT -leg experiment can be compared directly. In addition biaxial creep data on Type-316 stainless steel is needed to analyze the effects of a cyclic heat flux on stress-rupture behavior.

Construction of specimens for a second multipin test in flowing sodium has been completed. The test involves biaxial creep of five heats of austenitic stainless steel, stress-rupture of tubes used in the first multipin test, corrosion

rings, and post-exposure stress-rupture specimens. None of the tubes is wire-wrapped. The assembly is scheduled for completion by the end of next month.

A multipin retort was constructed and filled with sodium for inducing case-carburization in Type 316 stainless steel by methane-gas reaction with liquid sodium. The surface of the retort was aluminized to prevent carbon pickup by the retort. Post-exposure, tensile, fatigue, and biaxial stress-rupture tests will be made.

III. PROGRESS DURING REPORT PERIOD

A. SUMMARY

The ΔT leg experiment in the 2-in. flowing-sodium loop was terminated after 2613 hr at test conditions listed in Table 1. The stresses and rupture data from the test are given in Table 2 and Figure 1. For comparison, stress-rupture data in static sodium are also given in Figure 1. The intent of the test was to carry 12 specimens to failure and determine creep behavior from the other seven. It appears that neither the wires nor the flowing sodium produced any large effect on stress-rupture life.

TABLE 1
TEST CONDITIONS FOR ΔT LEG

Test Temperature	1200°F
Sodium Velocity	6 ft/sec
Leg ΔT	50°F
Bulk-Sodium Temperature	1150°F
Oxygen Content	5 ppm
Carbon Content	12 ppm

After cleaning and a preliminary examination of the integral array, the tubes were individually removed and the examination continued. Twelve failures were noted, one of the creep tubes failed unexpectedly, and one stress-rupture tube did not fail at the time the test was ended. Nine of the failures appear to be associated with the presence of wires. They were located either

TABLE 2
SUMMARY OF DATA AND TEST CONDITIONS ON TUBES
IN 1200°F FLOWING SODIUM

Pin No.	Hoop Stress (psi)	Type of Test	Time at Pressure (hr)	Rupture or Average Strain (%)	Average Strain Rate (%/hr)		
16	25,148	Stress-Rupture	2290	2.5	1.1×10^{-3}		
15			2613*	3.6	1.4×10^{-3}		
14			2418	3.1	1.3×10^{-3}		
5			809†	0.6	-		
3			2100	2.9	1.4×10^{-3}		
9		Creep	2369	3.2	1.4×10^{-3}		
10			1009†	1.0	DNA§		
2			2009	2.0	↓		
13			1391	0.8			
20			885	0.6			
12	500	0.4					
11	306	0.4					
25	27,939	Stress-Rupture	100	0.2	↓		
18			1242	2.6		2.1×10^{-3}	
17			498†	0.7		-	
6			1242	3.4		2.7×10^{-3}	
7			29,804	754		4.8	6.4×10^{-3}
19				706		3.0	4.2×10^{-3}
21	987	DNA		-			

*Test was ended before rupture.

†Denotes suspected premature ruptures.

§DNA = does not apply to creep test.

3-17-70 UNCL

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Figure 1. Stress-Rupture of Cold-Worked Type 316 Stainless Steel in Static and Dynamic Sodium at 1200°F

under wires on the tube or near polished areas from loose wires on neighboring tubes. Three failures (designated premature) were not associated with wires; rather they appear to have been caused by undetected flaws in the tubing. Preliminary examination has revealed several instances of point-contact adhesion (self-weld) of the wires to their tubes and of loose wires from neighboring tubes to cladding.

Four high-flux heater assemblies were built and tested in the 2-in. loop. The cladding was annealed Type 316 stainless steel and exhibited a large amount of strain before rupture. The test conditions and preliminary results are shown in Table 3.

TABLE 3
TEST CONDITIONS AND RESULTS FOR HIGH-FLUX HEATERS

Heater No.	Material	Hoop Stress (psi)	Heat Flux $\times 10^6$ Btu/ft ² -hr	Cycle Mode	Time-to-Failure of Cladding (hr)	% $\Delta D/D$
305	316 SS (Annealed)	21,000	0.8	Steady	109	10.7
306	↓	↓	↓	Steady	17	-
307	↓	↓	↓	9 min on, 3 min off	114	>14.1
308	↓	↓	↓	↓	120	>14.1

The static-sodium retort with associated specimen holders for producing a carbon case on the surface of stainless-steel specimens was built and coated with aluminum to reduce carbon pickup by the retort and specimen holders. The aluminum was subsequently oxidized at 1800°F for 1 hr to prevent contamination of the Na with Al. The retort assembly is shown in Figure 2.

Four additional high-flux heater assemblies were built and are currently on test. The test conditions are given in Table 3. One heater is instrumented with thermocouples to obtain confirmation of cladding temperatures calculated using heat-transfer methods. Two of the heaters are an advanced design electrical heater which replaces the graphite heating element with a hollow metal tube.

Figure 2. Retort Assembly for Surface Carburization Studies on Type 316 Stainless Steel

B. TECHNICAL PROGRESS

1. Mechanical Properties

a. ΔT -Leg Experiment

As part of this program biaxial stress-rupture and creep tests were made on cold-worked Type 316 stainless-steel cladding in flowing sodium and under a loop ΔT . The cladding was wire-wrapped. The purpose of the test was to study the combined effects of mass transfer from flowing sodium and wire-wrap spacers on the mechanical behavior of the cladding. Suitable control tests were also made in static sodium but without wires.

The 2-in. flowing-sodium loop is shown schematically in Figure 3. As previously reported, to accommodate the test assembly and create the conditions which favor mass transfer a ΔT leg was added to the loop (see Figure 4). The horizontal section is used to heat the flowing sodium before entering the test section. The leg was designed to generate a 50°F ΔT in sodium just before entering the test column. The temperature at the inlet to the test column was fixed at 1200°F . Heat losses along the piping are sufficient to restore to 1150°F the temperature at the inlet to the leg. The conditions of the test are given in Table 1.

(1) The 19-Pin Assembly

The test assembly consists of 19 individually pressurized tubes of Type 316 stainless steel in the cold-worked condition. The tubes are 2-in. long by 0.250-in. OD by 10-mil wall. The tubes were wrapped with 70-mil wires of annealed Type 316 stainless steel. During assembly the tubes were arranged in a hexagonal array and a shroud tube was placed around them. The completed assembly was placed in the loop as shown in Figure 5. Pertinent data about the assembly are given in Table 4.

The pressure in the tubes was programmed for stress-to-rupture and creep testing. In the stress-rupture test the internal pressure was held constant until the tube failed. In the creep test seven identical tubes were pressurized to the same level and held constant. At prescribed time interval the pressure on a given tube was removed. This was repeated on all seven tubes for times ranging from 100 to 2613 hr. After completion of the test the diameter of each tube

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Figure 3. Diagram of the 2-in. Flowing-Sodium Loop with ΔT Leg Modification

Figure 4. The ΔT Leg

Figure 5. The 19-Pin Test Assembly with Housing

TABLE 4
TEST-ASSEMBLY INFORMATION

Cladding: Material	10 to 15% CW Type 316 stainless steel
Dimensions, in.	0.250 OD by 0.010 wall by 20 long
Wire-wrap: Material	Annealed Type 316 stainless steel
Diameter, in.	0.070
Pitch, in.	12
Tension, lb	40
Strain allowance in bundle	8% $\Delta D/D$
Shroud material	Annealed Type 316 stainless steel
Pressurization gas	Helium
Pellet dimensions, in.	0.220 OD by 0.5 long
Pellet material	Annealed Type 316 stainless steel
Weight of a pin, lb	1
End-cap: Weld type	Electron-beam lap
Material	Annealed Type 316 stainless steel
Type	Solid plug

was measured to give diametral strain as a function of time at pressure. The strains of seven tubes then comprise a composite creep curve. The test conditions for each tube in the array are given in Table 2 and their position is shown in Figure 6.

It is possible to compare the results of the biaxial-creep and stress-rupture tests in flowing sodium to those in static sodium. Unfortunately most of the base-line in static sodium is for a different lot (same heat) of cladding with a 0.295-in. OD and 10-mil wall. (The tubing tested in flowing sodium was 0.250-in. OD and 10-mil wall.) The smaller diameter was selected because it was closer to FFTF reference diameter. The tubing was procured to the same specification for both lots (except diameter) and contains 10 to 15% cold-work in the "as-received" condition. It is possible that the two lots do not contain exactly the same degree of cold-work.

A comparison of the results of stress-rupture tests in flowing (ΔT leg) and in static sodium is given in Figure 1. As can be seen the stress-rupture data

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Figure 6. The Test Configuration as Viewed from the Inlet

and the associated statistical band for the wire-wrapped tubes tested in flowing sodium overlap the data and band for unwrapped tubes in static sodium. Stress-rupture tests in static sodium on the same lot as used in the ΔT leg are still in progress. Ruptures in static sodium for this lot were obtained at the highest stress level (Retort 125 in Table 5). At the same stress level (29,804 psi) the time-to-failure in this retort agreed with that in flowing sodium but not with that in static sodium on another lot (0.295-in. OD, Retort 124). From these data the stress-rupture life of the 0.250-in.-OD tubing is longer than that of 0.295-in.-OD at the same hoop stress at 1200°F. The exact reason is not known but the difference may be due to differences in cold-work.

The slope of the flowing-sodium stress-rupture curve is steeper than for the static-sodium curve (Figure 5). The reason for this steeper slope is not known; but there are several possibilities. A steeper slope could arise from the presence of more cold-work in 0.250-in.-diam tubing (flowing sodium) than the 0.295-in. Previous work in Task C of this program has shown that the slope of stress-rupture curve increases with the degree of cold-work.⁽¹⁾ Another possibility is that mass transfer of carbon out of the cladding resulted in a weakening. This effect has been observed in zirconium-gettered static sodium by Atkins and Lee⁽²⁾ and more recently by General Electric in flowing sodium.⁽³⁾ Still another possibility is that wire-cladding interactions which occurred during wire attachment and/or during the test reduced the life under stress. Further insight into these mechanisms must await completion of the control tests on the 0.250-in.-OD lot and further evaluation of the results.

The rupture ductility of the wire-wrapped tubes tested in flowing sodium is less than that shown by unwrapped tubes in static sodium. This can be seen by comparison of strain at failures for the two tests in Tables 2 and 5. It can also be seen in Figure 7 in which the strain at rupture is plotted vs the average strain rate. The average strain rate is simply the rupture strain divided by the time-to-failure. The flowing-sodium data are displaced from the static-sodium data and lie below. Again, these data are for two different lots.

The rupture-strain data may provide additional insight into the stress-rupture mechanisms. The lower rupture strain for the smaller-diameter lot of tubing is commensurate with the presence of more cold-work in the tubing.⁽¹⁾

TABLE 5
SUMMARY OF DATA AND TEST CONDITIONS ON TUBES
IN 1200°F STATIC SODIUM

Retort No.	Hoop Stress (psi)	Type of Test	Tube Diameter (in.)	Time at Pressure (hr)	Rupture or Average Strain (%)	Average Strain Rate (%/hr x 10 ³)
102	22,000	Stress-Rupture	0.295	6770	4.3	0.63
				7152	5.8	0.81
				7046	4.8	0.67
	25,000			3412	6.7	2.0
				3623	9.8	2.7
				3396	5.1	1.5
	30,000			752	7.0	9.2
	30,000			790	6.3	8.0
	35,000		105	5.5	52.4	
109	27,000	Stress-Rupture	0.295	1189	8.3	7.0
				1354	8.7	6.4
				1822	12.2	6.7
	32,000			276	5.3	19.2
				292	6.4	22.0
				206	2.5	12.0
				129	4.0	30.6
	34,000					
124	25,148	Creep	0.295	97	0.25	DNA*
				306	0.42	
				502	0.50	
				866	0.84	
				1433	1.3	
				1994	3.1	
		Stress-Rupture		2474	9.1	3.6
				2522	8.6	3.4
				2266	8.2	3.6
	29,004			489	4.9	10.0
				489	5.7	11.6
				513	7.4	14.4
125	25,148	Creep	0.250	100	NA†	NA†
				300		
				500		
				885		
				1400		
				2000		
		Stress-Rupture		NA		
				NA		
				NA		
	29,804			776		
				814		
				824		

*DNA = Does not apply to creep test.

†Not available because test is still underway.

3-27-70 UNCL

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Figure 7. Rupture Strain of Cold-Worked Type 316 Stainless Steel as a Function of Average Strainrate at 1200°F

Unfortunately, it is also compatible with the decarburization mechanism⁽²⁾ and the wire-interaction mechanism mentioned earlier.

It should be mentioned that the average strain rate is not an accurate prediction of the secondary creep rate in cold-worked stainless steel. This can be seen in Figure 8 which shows the composite biaxial-creep curves for the two lots of stainless steel. As can be seen the tubes exhibited a large amount of third-stage creep. The average creep rate, represented by a line joining the origin to the rupture strain clearly overestimates the secondary-creep rate by at least a factor of 5 in static sodium and 3 in flowing sodium. The creep data in flowing sodium shown in Figure 8 are not identical to those in static sodium. At least part of this difference may be explained by the presence of slightly more cold-work in the 0.250-in.-OD tubes than in the 0.295-in.-OD tubes, i. e. slightly lower secondary-creep rate and lower rupture strain. A biaxial-creep test is currently under way in static sodium on the 0.250-in.-OD tubing at 1200°F.

Premature failures were observed in three tubes presumably due to undetected defects in cladding. These failures fell outside the 2σ scatter band in the stress-rupture curve for the 0.250-in.OD in flowing sodium (see Figure 1) but fell on the creep curve (see Figure 8). The defects may be located on the ID of the tubing. The strain-to-failure on these tubes was also less than companion specimens. In fact they lie on the creep curve. If these failures are due to cladding defects, then defects constituted a more serious life-limiting factor than either flowing sodium or wire interactions.

The wires on all tubes tested in 19-pin array in flowing sodium slackened. The first evidence of this was seen when pin No. 21 (corner) was removed from the array with the shroud tube still in place. The force to remove this pin was approximately 6 lb; the weight of the pin outside the assembly was approximately 1 lb. After removal of the shroud tube from array, the reason for this interference was revealed. Most of the wires were loose and no longer in contact with the tubes, as can be seen in Figure 9. A differential-strain allowance of 8% (20 mils) between the wire-wrapped tubes and housing, or between tubes, was built into the assembly. Most of the wires moved away from the tubes by more than this amount, which produced an interference between loose wires and the shroud or neighboring tubes. The loose wires may be explained by tube shrinkage during creep.

3-17-70 UNCL

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Figure 8. Biaxial Creep and Stress-Rupture Strain of Cold-Worked Type 316 Stainless Steel as a Function of Time at Pressure in 1200°F Static and Dynamic Sodium

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40

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Figure 9. Loose Wire on Pin No. 21

Unfortunately the length of the tubes was not measured accurately before the test. Previous work on short tubes had failed to show evidence of length changes of tubes. Therefore it is only possible to compare the length of untested tubes with those tested in flowing sodium. The distances between the end-cap welds and between shoulders on the weld preps were used as an index (see Table 6). All tested tubes were shorter than the untested tubes.

The shortening is a function of time under stress and stress level (Figure 10). Clearly all tubes became shorter during the test. The length decrease vs diametral strain for the tubes tested is shown in Figure 11. The degree of shortening is clearly proportional to the creep strain within the uncertainty in the length.

Tube shrinkage is believed to be largely due to mechanical anisotropy. For a perfectly isotropic material under a 2:1 stress-state the length of the tube remains fixed. However, if the creep strength in the longitudinal direction is greater than the hoop direction this assertion is not valid. In fact in this case the tubes would shrink. Previous work on cold-worked Type 316 steel under the 1:0, 2:1, and 1:1 stress-state showed that the rupture time obeys the largest principal stress-failure criterion. Thus the cold-worked Type 316 stainless steel does not exhibit isotropy with respect to failure. It seems likely that it also does not exhibit isotropy with respect to creep-strain.

The shortening of tubes can completely explain the looseness of wires in the 19-tube test. A length decrease of 20 mils (0.1%) produces a situation in which the wires appear longer. Since the ends of the wires are fixed the wires bow outward away from the cladding. The amount of deflection can be calculated by assuming the tube is a line cutting through an obtuse angle formed by the wire at the midpoint. The distance between the wire and the tube can be calculated by the following formula:

$$h \cong 2L \Delta L,$$

where

h = height of wire above cladding,

L = length of tube, and

ΔL = tube shrinkage.

TABLE 6
SUMMARY OF TUBE-LENGTH
MEASUREMENTS

Tube No.	Tube Lengths		Time Under Stress (hr)
	X (in.)	Y (in.)	
2	20.159	20.023	2009
3	20.152	20.019	2100
5	20.171	20.033	809
6	20.121	19.992	1242
7	20.115	19.991	754
9	20.150	20.010	2369
10	20.165	20.026	1009
11	20.155	20.021	306
12	20.169	20.025	500
13	20.164	20.019	1391
14	20.110	19.969	2418
15	20.141	20.007	2613
16	20.148	20.021	2290
17	20.156	20.023	498
18	20.139	19.999	1242
19	20.145	20.002	706
20	20.174	20.027	885
21	20.152	20.007	987
25	20.165	20.026	100
1	20.179	20.042	0
4	20.191	20.056	0
22	20.181	20.041	0
23	20.176	20.049	0
8	20.171	20.041	0

X = Shoulder-to-shoulder distance.
Y = Weld-to-weld distance.

3-17-70 UNCL

6507-4704

Figure 10. Tube Shrinkage (weld-to-weld distance) as a Function of Time Under Pressure and Initial Hoop Stress

AI-AEC-12948
44

3-16-70 UNCL

6507-4705

Figure 11. Tube Shrinkage (weld-to-weld distance) in Relation to Diametral Strain

Using 20 mils for ΔL , the distance between the wire and the tube is about 0.4 in. This value perhaps represents an upper limit. It exceeds by a factor of ten the space allowed between adjacent tubes.

(2) Self-Welding

Adhesion of wires to cladding was observed at two specific locations along the length of a tube: (1) at the ends of the tube where the wire remains tightly compressed against the cladding, and (2) at the center of tube where a loose wire touched a neighboring tube. Near the ends of the tube the adhesion remained intact after the test. Welds of this type are shown in Figures 12 and 13. After the wire was removed from the tube the adhesion was essentially between high points on the surface. This suggests that the contact pressure was not very large. The contact was essentially along a line as shown in Figure 14.

In contrast the contact adhesion of loose wires to neighboring tubes was broken after the test. There is even evidence that these welds were formed and broken during this test, as illustrated in Figure 15. As can be seen in the figures, sizeable nuggets were removed from the wire and deposited on the cladding. In one case the welding appeared to progress along the tube as the wire became looser. The size of weld-deposit increased progressively indicating that the force producing the welds increased with time. This is understandable because the wire-cladding interference increases with creep strain (tube shrinkage).

(3) Failure Location and Wire Interaction

Failures were observed in 12 of the 19 tubes tested. Actually one creep specimen not programmed for failure did rupture prematurely, and one stress-rupture specimen did not fail before the test was terminated after 2613 hr. The location of all failures has been determined. Seven cladding failures occurred near the inlet to the bundle within 1 in. of the end-cap weld and very near the end-cap plug. Two failures occurred near the middle of the bundle. There was evidence of a loose-wire interaction with the cladding at this site of the failure. Three of the failures were not associated with wires in any way. These failures were all premature and are believed to be due to undetected cladding defects.

Figure 12. Self-Weld (Adhesion) of Loose Wire to Cladding on a Neighboring Tube

Figure 13. Self-Weld (Adhesion) of Wire to Cladding

Figure 14. Several Self-Welds (Adhesions) of Loose Wire to Neighboring Tube

Figure 15. Wire-Cladding Interaction on a Tube Tested in Flowing Sodium

Figure 16. Wire-Cladding Interaction on an Untested Tube

The failures under wires are probably associated with wire-cladding interactions produced during the wire-wrapping process. A flat spot on the cladding and wire was observed near the end-cap weld, shown in Figures 16 and 17 for tested and untested pins respectively. A triangular zone of interaction on the surface of the cladding and of the wire results when the clamping force is used to hold the wires in place on the tubes during welding of the wire to the end-cap. The zone is triangular because the end-cap plug is tapered.

The seven failures which occurred under wires were located just behind these zones of interaction. The failure extended into the zone in some cases. The failure region was one major region of self-welding of wires to cladding. The analysis of the causes of the location of failures is, therefore, preliminary and inconclusive. An important point worth mentioning is that undetected cladding defects had a stronger impact on rupture life and ductility than wire interactions produced during fabrication. In fact it remains to be proven that either the wires or the flowing sodium produced any effect on mechanical behavior. This information must await completion of the control tests in static sodium on the same lot of tubing.

In summary seven failures were found under wires. The presence of a cladding-wire interaction was detected on both tested and untested tubes. The interaction was introduced by clamping forces during wire attachment to the cladding. The effect of the interaction is unknown.

(4) Mass Transfer

Surface replication of tubes exposed to flowing sodium has been used successfully to observe the effects of mass transfer. Replicas were made of the surfaces of tubes tested in flowing sodium in the ΔT leg. The appearances of the surfaces before and after exposure are shown in Figure 18. The extent of mass transfer cannot be determined from these replicas. Comparison of the surface from tests in flowing sodium under high heat flux to the surface shown in Figure 10 suggests that less corrosion has taken place.⁽⁴⁾ This is not surprising because the surface temperature of the cladding is 50°F higher under a heat flux and because of a lower velocity in the ΔT leg (20 ft vs 6 ft/sec).

In addition the driving force for corrosion provided by the undersaturation of the sodium with the corroding species is less in the ΔT leg than in the heater

Figure 17. Photomicrograph of a Replica on an Untested Tube Surface

a. Outlet

b. Inlet

Figure 18. Photomicrographs of Replicas at the Inlet and Outlet Surfaces of a Tube Tested in Flowing Sodium

test. The smaller amount of undersaturation is a result of preheating the sodium in the ΔT leg before its entrance to the test column. During the preheating the surface of preheaters experiences selective removal of the components in the stainless steel. This reduces the degree of undersaturation and hence the driving force for mass transfer.

Preliminary metallographic studies have revealed the presence of a ferrite layer on the surface of the cladding exposed to flowing sodium. No ferrite layer was observed in regions shadowed by the wires. The presence of a ferrite layer (Figure 19) is indicative of selective removal of chromium and nickel from the surface of the steel. The metastable austenite then transforms to ferrite.

The mass transfer is not the same at all locations on the surface of the tubes tested in flowing sodium. As expected the surface polishing is less at the outlet to the assembly than at the inlet. This is a result of the corrosion along the bundle decreasing the driving force for mass transfer by reducing the degree of undersaturation. In addition, the surface of the tubes shadowed by the wires does not exhibit strong evidence of mass transfer.

Figure 19. Ferrite Layer on the Surface
Near the Inlet of a Tube Tested
in Flowing Sodium

Taken together the metallography and surface impressions revealed evidence of mass transfer on the cladding. It is not known whether there was any migration of carbon from or into the cladding.

The shroud tube made of annealed Type 316 stainless steel did not observably distort during the test. There was no evidence of wire interaction with the shroud. Based on this, the strain allowance in the array, and the strain measurements on the tubes, no mutual interaction occurred between the wires, tubes, and shroud during the test in the region of failures under the wires. Mutual interaction did occur in the center of the bundle between loose wires and the shroud or cladding.

b. Interstitial Effects on Stainless Steels

An investigation has been started to determine the effects of changing carbon content in liquid sodium on the engineering-design properties of cold-worked 316 stainless-steel tubing. The strength and ductility of the material will be determined as a function of case-carbon content for creep-rupture, short-term tensile, and low-cycle fatigue. Carbon surface concentrations of about 1.50, 1.75, and 2.00% will be created by adding methane-gas to a retort containing the specimens immersed in liquid sodium. The gas will be added incrementally from a reservoir. After the desired carbon case concentrations have been attained (~167 hr) the specimens will be removed from the retort and tested by straight tensile, stress-rupture, and low-cycle fatigue in helium. Such information will enable a correlation between carbon-case concentrations and mechanical properties. The exposure parameters in the case-carburization phase of the program are given in Table 7.

The carburization retort is complete and loaded with sodium. The retort assembly for introducing carbon into stainless steel through the sodium is shown in Figure 2. A key feature of this retort is that the internal surfaces are aluminized, followed by an oxidation treatment. The aluminum acts as a barrier to carbon pickup by the surface and prevents carburization of the stainless-steel retort. The aluminum oxide acts as a barrier to migration of aluminum into the sodium. The exposure is scheduled to begin immediately.

TABLE 7
EXPOSURE CONDITIONS FOR CARBON-LIMITS STUDY

1. Sample configuration	Tubing, 0.275-in. ID by 0.010-in. wall by 4 in. long, 6.8-in. long
2. Temperature	1200°F
3. Stress	Filled with helium (1 atm at 1200°F)
4. External environment	Liquid sodium
5. Material	Type 316 stainless steel as-received (10 to 15% C. W.)
6. C source	CH ₄ gas
7. Circulation (of liquid Na)	Revolving retort at 30° angle

Electron-microprobe analysis is expected to be used in the carbon-limits work. In preparation for these studies several improvements in the electron-microprobe were made, to eliminate the carbonaceous deposit formed during an analysis by the interaction of the electron-beam with oil vapors from the diffusion-pump at the point on the specimen surface being analyzed. A combination cold-finger/gas-jet assembly has been designed and assembled which should satisfy the requirement of minimal carbon contamination during analysis. The cold-finger will condense the oil vapors in the proximity of the specimen while the gas-jet (using nitrogen) will "scrub" the surface clean. In addition a nitrogen-diffusion pump baffle will be used to reduce the amount of oil vapors in the electron-microprobe vacuum tank. As part of the overall setup a number of needle, leak, and relief valves and pressure gauges (standard and compound) have been gathered. The cold-finger gas-jet and nitrogen-diffusion baffle are currently being installed on the probe.

c. High-Flux Heaters

Five high-flux assemblies were tested during the quarter. One test consisted of a proof-of-principle test on an AI design built by Watlow. This heater failed by open-circuit during start for unknown reasons. The four other tests with heaters involved tests on annealed Type 316 stainless-steel cladding. The test conditions and preliminary results from these tests are given in Table 3.

The most surprising result of this test was the large amount of strain exhibited in the cyclic specimens. The exact strain at rupture could not be measured because of the nature of the failure. The strain was so excessive that it partially blocked the coolant channel and caused overheating of the cladding.

Four additional heaters were built and are now on test in flowing sodium. The test conditions are the same as summarized in Table 3. One heater contains thermocouples at the surface of the cladding and in the NaK annulus between the cladding and the sheath. This heater will be used to provide temperature data under high flux. Two developmental heaters are also included. One test involves the effects of a steady-state heat flux on biaxial-creep rupture.

IV. EVALUATION OF EFFORT TO DATE

The program for the subtask continues to provide data on the effects of flowing sodium on mechanical behavior. The most significant observations this quarter are connected with the ΔT -leg test recently completed. It was surprising that as a result of tube shrinkage, wires were slacked. Also, evidence of self-welding between wires and cladding was not anticipated.

V. NEXT REPORT PERIOD ACTIVITIES

The following major activities are anticipated for the next report period:

- 1) Completion of three 1000-hr steady-state heater tests on cladding in flowing sodium.
- 2) Complete fabrication of second Δ -T leg multipin test assembly and start of testing in flowing sodium.
- 3) Issue interim topical report on the results of the first ΔT -leg experiment.
- 4) Complete metallography and carbon analysis of cladding used in the first ΔT -leg test.

REFERENCES FOR TASK 10-D

1. J. H. Shively, D. F. Atkins, and W. T. Lee, "Creep Under High Heat Flux and Stress-Rupture Properties of Austenitic Stainless Steels in Elevated-Temperature Sodium," ANL 7520, Part I, November 9, 1968
2. W. T. Lee, "Biaxial Stress-Rupture Properties of Austenitic Stainless Steels in Zirconium-Gettered Sodium," NAA-SR-12353, October 1, 1967
3. Quarterly Progress Report No. 11, GEAP 13539-11, November 1969
4. J. Hopfenfeld, "Corrosion of Type 316 Stainless Steel with Surface Heat Flux in 1200°F Flowing Sodium," AI-AEC-12898, November 30, 1969

Program: Fuel Cladding and Structural Materials

AEC Task: *Task 2-A, Irradiation Damage in Cladding and Core Structural Materials

Project Manager: J. J. Gill

Reporting Period: January-March 1970

General Order: 6506

Subaccount: 29621 & 29622 AEC Category: 04-40-02-01.1

Principal Investigator: D. Kramer

I. PROJECT OBJECTIVES

The objectives of this task are to:

- 1) Study the process of void formation and helium embrittlement in fuel cladding and core structural alloys by means of accelerator irradiations and reactor irradiations.
- 2) Determine the effects of flux, fluence, temperature, helium concentration, and microstructure on void formation and helium embrittlement.
- 3) Make preliminary evaluations of modified and new alloys developed by other laboratories.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

The production of voids in Type 316 stainless steel by present proton-irradiation techniques was demonstrated. Voids were observed in a sample containing 2×10^{-6} atom fraction helium irradiated with 1.2-Mev protons to a fluence of 6×10^{18} p/cm² at 500°C.

Stress-rupture tests of Type 316 stainless steel at 700°C were completed for helium concentrations of 1.5×10^{-6} and 4×10^{-5} atom fraction at stresses between 10,000 and 30,000 psi.

*Task 2-A [Contract AT(04-3)-824] was previously Task 10-B-1 under Contract AT(04-3)-701.

III. PROGRESS DURING REPORT PERIOD

A. VOID FORMATION IN TYPE 316 STAINLESS STEEL

Additional electron microscopy was done on Sample 6 at proton-penetration depths of 0.0003 and 0.0004 in. as measured by x-ray techniques. The foil previously reported to be at the 0.0003-in. depth (October-December 1969 Quarterly Technical Progress Report, AI-AEC-12913) without the benefit of x-ray measurements is now known to be at a depth between 0.0003 and 0.0004 in. The distribution of voids at the true 0.0003-in. proton-penetration depth is given in Table 1 and their appearance is shown in Figure 1.

Some regions with voids were observed at the 0.0004-in. depth although most of the foil had no voids. Since 0.0004 in. is beyond the range of the 1.2-Mev protons used (0.00033 in. is the end-of-range) it is likely that surface irregularities which provide deeper-than-average penetrations or uneven etching during the thinning process are responsible for this effect.

Tables 2 and 3 summarize the irradiations to date and the recent results of electron microscopy. The irradiation temperature for the second series of samples is now shown as approximately 550°C instead of 500 as previously reported. A temperature difference of about 50°C that arose during irradiation between a thermocouple and the infrared detector was resolved in favor of the latter. This problem will be corrected by relocation of thermocouples.

Voids were present at the 0.0004-in. depth of Sample 35 (Figure 2) but not in Sample 46, thus emphasizing the importance of helium since the latter did not contain any. Owing to the higher incident energy of the proton beam (the end-of-range of a 1.4-Mev proton is 0.00041 in.), the 0.0004-in. depth of Samples 46 and 35 is equivalent to the 0.0003-in. depth of Sample 6 for proton energy in those locations. Dislocation loops and irradiation-induced precipitates which had grown on pre-existing dislocations were also observed in Samples 46 and 35 (Figure 3).

A contribution to the 1970 AEC Report on Fundamental Nuclear Energy Research was completed on the subject of void formation in stainless steel by proton irradiation.

TABLE 1
VOID FORMATION AT A PROTON-PENETRATION DEPTH OF
0.0003 in. IN SAMPLE 6

Void Diameter (Å)	% of Total	Void Density ($\times 10^{14} \text{ cm}^{-3}$)	Volume Increase (%)
0 to 25	-	-	-
25 to 50	3.3	1.5	0.00
50 to 75	9.5	4.3	0.01
75 to 100	5.5	2.5	0.01
100 to 125	11.1	5.0	0.04
125 to 150	17.9	8.1	0.11
150 to 175	24.3	10.9	0.24
175 to 200	16.1	7.3	0.25
200 to 225	7.2	3.3	0.16
225 to 250	4.1	1.9	0.13
250 to 275	0.8	0.3	0.03
275 to 300	0.2	0.1	0.01
TOTALS		$4.5 \times 10^{15} \text{ cm}^{-3}$	1.0%

Figure 1. Transmission-Electron Micrograph of Type 316
Stainless Steel Containing 2×10^{-6} Atom-Fraction
Helium and Irradiated with Protons at 500°C.
Proton-Penetration Depth = 0.0003 in.

TABLE 2
PROTON-IRRADIATED SAMPLES OF TYPE 316 STAINLESS STEEL

Irradiation No.	Sample No.	Atom Fraction Helium ($\times 10^{-6}$)	Temperature ($^{\circ}\text{C}$)	Energy (Mev)	Flux ($\text{p}/\text{cm}^2/\text{sec}$)	Fluence (p/cm^2)
1-1	6	2	500	1.2	1×10^{15}	6×10^{18}
2-1	46	~ 0	~ 550	1.4	4.4×10^{14}	6×10^{18}
2-2	35	~ 10	~ 550	1.4	4.4×10^{14}	6×10^{18}
2-5	3	0.09	~ 550	1.4	4.4×10^{14}	6×10^{18}

TABLE 3
ELECTRON-MICROSCOPE RESULTS

Sample No.	Proton Penetration Depth (in.)	Void Density (cm^{-3})	Void Diameter ($\text{\AA}$)	Volume Increase (%)
6	0.0001	3×10^{15}	110	0.2
	0.0002	1×10^{16}	70	0.2
	0.0003	5×10^{15}	150	1
	0.0003X	3×10^{15}	300	5
	0.0004	Few	-	-
46	0.0004	None	-	-
35	0.0003	None	-	-
	0.0004	Low	150 to 200	-
3	Not examined	-	-	-

Figure 2. Transmission-Electron Micrograph of Type 316 Stainless Steel Containing a Nominal 1×10^{-5} Atom-Fraction Helium and Irradiated With Protons at About 550°C. Proton-Penetration Depth = 0.0004 in.

Figure 3. Transmission-Electron Micrograph of Type 316 Stainless Steel Containing a Nominal 1×10^{-5} Atom-Fraction Helium and Irradiated With Protons at About 550°C. Irradiation-Induced Precipitates are Seen on Dislocations. Proton-Penetration Depth = 0.0003 in.

B. HELIUM EMBRITTLEMENT

1. Creep of Type 316 Stainless Steel

Five creep tests were completed and two others are in progress on samples without helium at 700°C. Test results are given in Table 4 and average curves are plotted in Figure 4. The remaining two samples will complete the testing of samples without helium at 700°C.

2. Grain-Boundary Sliding

A sample of Sandvik 12R72HV containing 5×10^{-5} atom fraction helium was elongated 12.1% at 800°C. Subsequent measurements to determine the amount that grain-boundary sliding contributed to the total elongation were made and the results are given in Table 5 along with two previous results. It is significant that the presence of helium caused the grain-boundary sliding contribution (E_{gb}) to the total elongation to double in Sandvik 12R72HV.

TABLE 4
CREEP-TEST DATA OF TYPE 316 STAINLESS STEEL AT 700°C

Stress		Minimum Creep rate (hr ⁻¹)	Failure Time (hr)	Total Elongation (%)
(kg/mm ²)	(kpsi)			
11.2	16.0	7.48×10^{-4}	392	51
13.0	18.5	2.59×10^{-3}	129	53
13.0	18.5	2.51×10^{-3}	115	47
10.0	14.2	4.36×10^{-4}	645	52
10.0	14.2	4.15×10^{-4}	561	31
8.0	11.4	In progress	-	-
8.0	11.4	In progress	-	-

Treatment 6 = 1 hr 982°C + 8 hr 760°C

70-M18-021-12

Figure 4. Creep of Type 316 Stainless Steel at 700°C in Vacuum
(Heat treatment: 1 hr 982°C + 8 hr 760°C)

TABLE 5
GRAIN-BOUNDARY SLIDING AT 800°C

Alloy	Treatment	Atom Fraction Helium ($\times 10^{-6}$)	Total Elongation (%)	E_{gb} (%)	E_{gb}/Total Elongation (%)
316	1 hr 982°C + 8 hr 760°C	0	9.1	0.9	10
12R72HV	1 hr 1100°C + 8 hr 825°C	0	15	0.6	4
12R72HV	1 hr 1100°C + 8 hr 825°C	50	12	1.1	9

3. Sixth Helium Injection

Preparations were completed for the sixth helium injection on the cyclotron. Approximately 110 small samples of 316 SS in the annealed and 25% cold-work condition will receive 3×10^{-5} atom fraction helium and will be used for creep-testing. In addition a small number of samples of 19-9DL, 12R72HV, Vanstar, and 430 SS will also be included in the injection.

Fifteen samples of 316 SS for proton irradiation will be injected with helium to a concentration of 1×10^{-5} atom fraction. Of these, five will be in the annealed condition (Treatment 6), five will have 25% cold-work, and five will have 50% cold-work.

4. Topical Report

A final draft of a report on the effect of helium on the stress-rupture behavior of Types 304 and 316 stainless steel was completed.

C. EVALUATION OF ALLOYS FROM OTHER LABORATORIES

Alloys have been received from several laboratories, most of which will be fabricated into samples for accelerator irradiations. Table 6 gives the status of these alloys to date.

D. PUBLICATION

"Helium Embrittlement of Fuel-Cladding Alloys for Potential Service in Fast-Breeder Reactors," D. Kramer, K. R. Garr, A. G. Pard, and C. G. Rhodes, Proc. IAEA Symposium on Radiation Damage in Reactor Materials, Vol. 1, p. 303 (1969) Vienna.

TABLE 6
STATUS OF ALLOYS FROM OTHER LABORATORIES

Laboratory	Alloy	Status	Pre-irradiation Metallography
Pacific Northwest	316 SS, low nitrogen	To be fabricated	Not started
Battelle	316 SS, shock-hardened	Small sample received, strip coming	In progress
Oak Ridge	304 SS + 0.2 titanium	To be fabricated	Not started
Brookhaven	316 SS, fine-grained	To be fabricated	In progress
Brookhaven	Cobalt alloy	To be fabricated	Not started
Idaho Nuclear	19 Fe-44 Ni-37 Cr, micro-duplex	Small sample received	In progress

IV. EVALUATION OF EFFORT TO DATE

The demonstration of void formation by proton irradiation was an outstanding achievement. The experimental apparatus worked well and only some minor modifications remain to be done.

The extraction of thin foils for electron microscopy within the irradiated samples was done with confidence through the use of x-ray absorption techniques for thickness measurements. Calculations of volume increase by quantitative electron microscopy underestimate the actual volume increase because not all the voids present can be imaged. The best method of determining volume changes is by a wet-density technique. Although our samples are too small for the usual wet-density procedures a novel method using a Cartesian diver is being considered which shows promise of giving satisfactory results.

The creep-testing of 316 SS samples at 700°C is proceeding satisfactorily. The two creep machines are performing well and progress is being made on the completion of two more.

V. NEXT REPORT PERIOD ACTIVITIES

- 1) Proton irradiations of 316 SS samples will be carried out.
- 2) The sixth helium injection will be done.
- 3) The topical report on stress-rupture will be issued.
- 4) Creep-testing and construction of creep machines will continue.

Program: Fuel Cladding and Structural Materials		
AEC Task: * 2-B, Integrated (n, α) Cross-Sections by Helium Determinations		
Project Manager: J. J. Gill		
Reporting Period: January-March 1970		
General Order: 06506	Subaccount: 29623	AEC Category: 04-40-02-01.1

Principal Investigator: H. Farrar, IV

I. PROJECT OBJECTIVES

The objectives of this task are to (1) determine which elements in commonly used alloys are contributing to the high production of helium in fast-neutron irradiations so that their use might be eliminated or minimized, (2) determine the spectrum integrated (n, α) cross-sections of 47 elements in five typical fast-reactor spectra, (3) make estimates of the differential (n, α) cross-sections of the same elements by correlating the integral cross-sections in five different neutron-energy spectra by using unfolding techniques, and (4) use this information to predict with some confidence what helium production might be expected from any present or new alloy contemplated for fast-reactor use.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

Preliminary values of the spectrum-integrated (n, α) cross-sections of ten pure elements irradiated in EBR-II Row 4-center have been obtained by correlating mass-spectrometric helium concentration determinations with recently obtained neutron-spectrum information. Recovery and positive identification of the remaining samples irradiated at the five EBR-II core positions is essentially complete. New measurements of the concentrations of helium in iron and nickel at all five reactor locations have confirmed earlier results where comparable and have permitted a correlation of concentration with neutron spectrum.

The mass-spectrometer system has undergone several important modifications which have resulted in an improvement in the precision of analysis. The

*Task 2-B [Contract AT(04-3)-824] was previously Task 10-B-2 under Contract AT(04-3)-701.

evident importance of nitrogen as a contributor to the production of helium is being examined with a new technique developed at Atomic International for the measurement of extremely small quantities of nitrogen in metals.

III. PROGRESS DURING REPORT PERIOD

A. SPECTRAL AVERAGED (n, α) CROSS-SECTIONS

The measured helium concentrations in ten pure elements and Type 304 stainless steel irradiated for 8.25 days during EBR-II Run 31F have been correlated with the neutron-flux spectrum obtained by PNL using multiple-foil data and 2DB reactor physics calculations.

The expression used to obtain measured values of the spectral-averaged cross-section, $\bar{\sigma}_m$ is

$$\bar{\sigma}_m = \frac{N_{mm}}{\phi t} \quad \dots(1)$$

where N_m is the measured atom fraction of helium per sample target atom, and ϕt is the multiple-foil-derived value of total fluence, which is calculated to be 1.30×10^{21} n/cm² for the Row-4 position (radial distance 13.1 cm, 1.83 cm below midplane). Values of N_m and $\bar{\sigma}_m$ are given in Table 1 along with calculated values ($\bar{\sigma}_c$) in Column 4 obtained by using available energy-dependent cross-sections⁽¹⁻⁵⁾ $\sigma(E)$, and the expression:

$$\bar{\sigma}_c = \frac{\int_0^\infty \sigma(E) \phi(E) dE}{\int_0^\infty \phi(E) dE} \quad \dots(2)$$

where $\phi(E)$ is the multiple-foil-derived differential flux spectrum.

Comparison of the measured helium concentrations in the pure Fe, Ni, Cr, and B with the 0.32 ppma He measured in the Type 304 stainless steel suggests that as much as 38% of the total He in the stainless steel is probably due to the N. The particularly low neutron energy (1.6 Mev) at which the N (n, α) reaction will take place (compared with ~ 5 Mev for most major stainless-steel components) suggests that N could play a proportionally greater role in the production of He in materials irradiated in softer LMFBR neutron spectra.

TABLE 1
SPECTRAL-AVERAGED CROSS-SECTIONS OF MATERIAL IRRADIATED
IN EBR-II ROW-4 CENTER

Reaction	Helium Concentration and Estimated 2σ Standard Deviation (atom fraction $\times 10^6$)	Spectral Average Cross-Section (mb)	
		$\bar{\sigma}_m$ (measured)*	$\bar{\sigma}_c$ (calculated)
B (n, α)	220 ± 10	170	190
Ti (n, α)	<0.077	<0.059	-
V (n, α)	0.021 ± 0.003	0.016	-
Cr (n, α)	0.13 ± 0.03	0.10	0.061
Mn (n, α)	0.11 ± 0.01	0.085	-
Fe (n, α)	$0.097 \pm 0.004^\dagger$	0.075	0.051
Co (n, α)	0.050 ± 0.003	0.038	0.032
Ni (n, α)	$1.24 \pm 0.06^\dagger$	0.95	0.053
Cu (n, α)	0.090 ± 0.005	0.069	$0.053^\S$
Zr (n, α)	0.016 ± 0.001	0.012	-
304 SS (n, α)	0.32 ± 0.01	0.25	0.19

*Calculated from Column 2 using Equation 1 and $\phi t = 1.30 \times 10^{21} \text{ n/cm}^2$.
An estimated 10% (1σ) absolute uncertainty is assigned to these measured cross-sections, which is due mainly to the uncertainty in the absolute value of ϕt . The relative uncertainties between $\bar{\sigma}_m$ values for the different elements is limited only by the uncertainty of the measured helium concentration N_m , Column 2.

† The measured N contents of the Ni before and the Fe after the irradiation were <14 ppma and ~ 22 ppma, respectively; consequently, N appears to contribute less than 0.001 ppma to the total He content of the Fe and Ni.

§ Cross-section only for the contribution from ^{63}Cu .

B. SAMPLE PREPARATION

The five capsules and 35 subcapsules irradiated at the five EBR-II reactor locations have been opened and 500 gold-wrapped samples removed. Only sixteen samples remain unidentified; because all are known to be crystalline compounds not needed for immediate analysis they have been set aside. All the important elements found in various stainless steel and other alloys are represented at least once in each capsule. The disappearance of all metals and crystals in the Row-4 center capsule with melting points below 856°C indicates that local temperatures reached at least 856°C but not as high as 961°C, the melting point of Ag which was present. The PNL capsule irradiated adjacent to this capsule however, contained unmelted Al wire (melting point 660°C) which will be used for the determination of the He in Al. Similar Au foil was used in the PNL capsules to conduct heat from the samples but the capsules were far less densely packed. The other four Al capsules did not attain the minimum crystal melting point of 630°C.

Specimens of Fe, Ni, and Type 304 stainless steel from all five irradiation locations were prepared for He and N analysis by individual etching in concentrated HCl. Removing the surfaces of all samples permits a determination of the He or N in the bulk of the samples and minimizes the contribution of He formed at the surface as a result of any N on or near the surface during the irradiation.

A group of the next most important elements, including duplicate specimens of Fe, Ni, and stainless steel, has been selected from the five EBR-II locations for the next series of He determinations.

C. NITROGEN DETERMINATIONS

Four milligram-sized Fe samples, two irradiated and two unirradiated, after being etched in HCl were analyzed for N content using a high-sensitivity detector with a gas chromatograph. The results are compared in Table 2 with similar determinations earlier⁽¹⁾ which were made with unetched samples.

TABLE 2
NITROGEN CONCENTRATIONS IN IRON

EBR-II Irradiation Location	Nitrogen Concentration (atom fraction $\times 10^6$)	
	Etched in HCl	Unetched
Unirradiated	690	1120
Row-4 Center	6.6	22
Row-6 Center	3.6	-

It appears from these data that much of the N is removed by etching, but it is not yet known what fraction still remains on or near the surface. Further experiments must be done to determine the effects of etching and find the N contents of Fe and other elements irradiated at all five reactor locations. The important observation is the confirmation of the disappearance of N from the Fe as a result of the irradiation. The fact that the level dropped to almost zero suggests that this occurred early in the irradiation, and that therefore the contribution of He in the Fe from the N is expected to be negligible.

D. HELIUM-CONCENTRATION MEASUREMENTS

Samples of Fe and Ni wire which were originally cut from the same pieces of wire but were irradiated separately in the five EBR-II core locations were prepared by etching in HCl for He analysis by mass spectrometer. The concentrations, which ranged down to 0.47 parts per billion (atomic), are shown in Table 3 and have an absolute uncertainty of 3%. The relative uncertainties in all but the lowest concentration samples are considerably less than this. The data, which are compared in Table 3 with two values obtained earlier⁽¹⁾ using samples of the same wire materials, agree very well. The earlier measurements also included the analysis of He in Ni obtained from a different source which gave a concentration value of 1.20×10^{-6} atom fraction used in Table 1. No explanation is being offered at this time for the 11% difference. It should be noted, however, that the N concentration of the wire material was found earlier⁽¹⁾ to be less than 1.4×10^{-5} atom fraction before irradiation; hence

TABLE 3
HELIUM CONCENTRATIONS IN IRRADIATED IRON AND NICKEL WIRE

Sample Number	EBR-II Capsule Location			Neutron Flux Above E_{min}^* (n/cm ² -sec)	Helium Concentrations (Atom Fraction x 10 ⁹)	
	Row	Radius From Core Center (cm)	Distance Above Midplane (cm)		Reported Earlier [†]	New Data [§]
Fe-1	4	13.1	-1.83	1.69×10^{13}	97 ± 4	100.9 ± 1.3
Fe-2	6	24.7	-1.83	9.60×10^{12}		63.3 ± 0.8
Fe-3	7	33.6	-1.86	3.34×10^{12}		23.8 ± 0.3
Fe-4	7	33.6	37.39	2.80×10^{11}		2.54 ± 0.10
Fe-5	7	33.6	55.09	6.10×10^{10}		0.47 ± 0.05
Ni-1	4	13.1	-1.83	1.36×10^{13}	1340 ± 80	1370 ± 15
Ni-2	6	24.7	-1.83	7.75×10^{12}		790 ± 10
Ni-3	7	33.6	-1.86	2.70×10^{12}		283 ± 3
Ni-4	7	33.6	37.39	2.23×10^{11}		26.6 ± 0.4
Ni-5	7	33.6	55.09	4.95×10^{10}		4.26 ± 0.14

* E_{min} is 5.2 Mev and 5.5 Mev for iron and nickel, respectively.

†Data reported in Reference 1 were obtained from the same wire but from different positions within the capsule.

§Reported uncertainty is the 2σ standard deviation of samples within this group. Absolute uncertainty of the group as a whole is $\pm 3\%$ of the concentration value.

that N is not contributing significantly to the higher of the two He concentrations. The flux gradient across this capsule was relatively small. Duplicate He analyses and more N analyses will help explain this small problem. The data in the table indicate no apparent loss of He as a result of the recently analyzed samples having been etched in HCl.

Also shown in Table 3 are neutron fluxes above 5.2 and 5.5 Mev, the energies E_{min} , above which the (n, α) reactions with Fe and Ni respectively become significant.⁽²⁾ These preliminary values are calculated for the center position of each capsule by interpolation of dosimetry data recently obtained by PNL.⁽³⁾ Final correlation of these data, with suitable corrections for the very small effects of N and for the distance of the sample from the capsule center, must

await the confirmation of the above results by the forthcoming analysis of duplicate specimens. Nevertheless it is interesting to note that the ratio of the He concentration in Ni to that in Fe decreases as the neutron spectrum softens. The measured He concentrations follow the calculated neutron-flux values above $E_{\min}$ very well, the Ni values doing so more closely than the Fe. As expected, the values of the spectrum-averaged neutron cross-sections $\bar{\sigma}_m$ (Equation 1) decrease by more than an order of magnitude for the traverse from the first to the fifth capsule, but the results are not yet ready to be reported.

E. MASS-SPECTROMETER SYSTEM OPERATION AND MODIFICATION

A series of breakdowns and persistent vacuum leaks in the mass-spectrometer system required the venting of the mass spectrometer four separate times. Each venting in turn required two days of bake-out to obtain a sufficiently good vacuum that the mass spectrometer could be operated for short periods without the aid of vacuum pumps (static mode operation). The electromultiplier, which had lost much of its sensitivity, was replaced and a titanium "flash-getter" assembly was made and attached to the mass spectrometer through a large all-metal valve. By periodically spreading a thin layer of titanium on the liquid-nitrogen-cooled inner surfaces of the getter chamber, it is hoped that the hydrogen background level in the mass spectrometer will be at last reduced. A platinum-black getter that had no obvious effect on the hydrogen level was replaced with copper oxide.

F. COMPUTER CODE

A mathematical procedure to handle the data from the mass-spectrometer automatic-readout equipment has been worked out. Mathematical models to represent the various known contributions to the data from helium permeation and background changes are now being formulated.

IV. EVALUATION OF EFFORT TO DATE

The spectral-averaged cross-sections thus far obtained are as expected considerably different from those calculated. It is evident that N plays an important role in the production of He, and steps are being taken to assess this contribution by making low-level N determinations.

A series of analyses of Fe and Ni samples from all five reactor locations has given excellent results which agree very well with earlier data where it exists. An initial comparison of these results with fluence data obtained using dosimetry foils has proven very interesting. In doing these samples the mass-spectrometer system operated very well, displaying a detection limit of less than 2×10^{-11} atom fraction. Maintaining the mass-spectrometer system, however, is taking a larger fraction of the total effort than anticipated. Routine analyses (generally between 0.1 and 100 ppma) can be done at any time, but the precise determination of concentrations in the parts-per-billion range requires the instrument to be operating well and without vacuum leaks.

V. NEXT REPORT PERIOD ACTIVITIES

1. Mass spectrometric analyses of the He content of the most important elements found in LMFBR candidate cladding and structural materials will be continued.
2. Helium-concentration data will be correlated with fluence and neutron-spectrum information.
3. A number of low-level nitrogen determinations will be made.
4. A technical report describing the mass-spectrometer system and its method of operation will be written.

REFERENCES FOR TASK 2-B

1. H. Farrar IV in "Quarterly Technical Progress Report, Fuels Materials and Coolant Chemistry Programs, Oct-Dec 1969," AI-AEC-12913, Atomics International (1970)
2. W. N. McElroy, H. Farrar IV and C. H. Knox, "Helium Production Cross-Sections for Elements Irradiated in EBR-II," Submitted for Presentation to the American Nuclear Society 16th Annual Meeting, Los Angeles (1970)
3. W. N. McElroy, Private Communication (1970)

Program: Fuels and Cladding

AEC Task: 11, Mixed Carbide Fuels (Basic Properties)

Project Manager: J. L. Ballif

Reporting Period: January-March 1970

General Order: 7707

Subaccount: 28210

AEC Category: 04-40-02-01

Principal Investigator: G. Ervin, Jr.

I. PROJECT OBJECTIVES

- 1) To determine selected microstructural attributes and mechanical and physical properties of mixed carbide fuels both to establish the potential for these materials as nuclear fuels and provide a basis for interpreting and understanding irradiation behavior in terms of atomistic transport phenomena.
- 2) To develop, where necessary, preparative techniques for polycrystalline mixed carbide materials of controlled structure and stoichiometry.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

All planned experimental work on the NRX-9-101 samples was completed. Surface replicas of seven fuel samples were prepared at Los Alamos, and these were studied by electron micrography at Atomics International. Two additional measures of burnup were obtained by analyses of flux-monitor wires and of fuel samples, each agreeing reasonably well with that calculated previously from in-pile temperature data. A comprehensive report on the experiment has been published as topical report AI-AEC-12928. Also a paper on the subject was presented at a meeting of the Nuclear Division of the American Ceramic Society in Seattle in October, and a condensed version of the report sent for publication in the Journal of the American Ceramic Society.

Sintered pellets of $U_{0.8}Pu_{0.2}C - 1.6\% W$ were prepared and examined chemically and metallographically. Only limited solution of tungsten in the carbide was achieved, and surface replicas indicate that no fine particles were present in the trials made thus far.

In the helium-injection study it has been shown that two-thirds of the He bubbles produced by uniform temperature annealing adhere to the fine W particles in the UC-W alloys.

III. PROGRESS DURING REPORT PERIOD

A. COMPLETION OF THE NRX-9-101 EXPERIMENT

Work has been completed on two measures of burnup in addition to that already put to use, based on the in-pile temperature measurements. One of these was an isotopic analysis of flux-monitor wires and the other an analysis of irradiated-fuel samples. Difficulties were experienced with measurements in each, but both supplied some degree of confirmation of the burnup figures used.

Four flux-monitor wires were attached to the capsule during irradiation in the NRX reactor. They were 0.020-in. diam and enclosed in stainless-steel tubing. Two were aluminum with 1 wt % cobalt, one was iron and the other nickel. The reactions to be observed were $\text{Co}^{59}(n,\gamma)\text{Co}^{60}$ for the thermal-neutron flux, and $\text{Fe}^{54}(n,p)\text{Mn}^{54}$ and $\text{Ni}^{58}(n,p)\text{Co}^{58}$ for the fast flux. Short sections were cut from each for determination of specific activity of these isotopes. In transferring the specimens from Atomics International where the wires were removed from the capsule, to Los Alamos Scientific Laboratory where the analysis was made, only one of the sections examined was of the wire itself, and the other sections erroneously included the stainless-steel tubing. The only way to recover the results was to assume equal lengths of tubing and wire and thus calculate the contribution of the stainless steel to the reported activities. However, this greatly impaired the accuracy of the results which showed a great deal of scatter. The data and calculations are given in a report by F. C. Franklin (TI-707-24-008, December 1969) and the calculated burnups from this report are reproduced in Table 1. The range was from about 13,000 Mwd/MTU at the ends of the fuel string to about 20,000 Mwd/MTU at the center. This is about the same as the range calculated from temperature measurements (13,000 to 19,000) but the most probable values for the flux-monitor wire data are lower by 5 to 10% for the maximum burnup.

Three fuel specimens were selected for burnup analyses, and the work was done at Idaho Nuclear Laboratories. On this analysis also there were difficulties

TABLE 1
BURNUP CALCULATED FROM FLUX-MONITOR
WIRE DATA

Sample No.	Distance from Centerline of Fuel Column (in.)	Fuel Burnup (Mwd/MTU)
0a (top)	27.5	16,009
b	19.9	16,215
c	10.3	16,215
d	0.3	20,286
e	10.9	16,560
f	20.5	15,180
g	28.1	13,455
180a (top)	27.5	12,972
b	19.9	14,835
c	10.3	15,939
d	0.3	17,457
e	10.9	13,317
f	20.5	14,973
g	28.1	15,663

which impaired the value of the results. In the first place one of the samples was accidentally destroyed, leaving only No. 18 and 77, 21 and 9 in. away from the center of the fuel string respectively. Each sample was analyzed for uranium isotopes and for neodymium, thus obtaining two independent measures of at.% burnup. The Nd method is normally the more accurate of the two unless there is also an accurate isotopic U analysis of the unirradiated fuel, in this case not available. However, Nd analysis by the standard ASTM method proved to be unsuccessful on these samples, perhaps because the W in the UC samples interfered with the separation. A new separation method for Nd was developed at Idaho Nuclear, which appeared to be satisfactory but gave reasonable results only for Sample No. 18. The results and the conversion to Mwd/MTU units are shown in Table 2.

TABLE 2
BURNUP CALCULATED FROM FUEL ANALYSIS

Fuel Specimen No.	Analytical Method	Burnup (at.%)	Burnup (Mwd/MTU)
18	Isotopic uranium	1.50	12,900
18	Neodymium	1.69	14,600
77	Isotopic uranium	2.00	17,200
77	Neodymium	-	-

Burnups calculated from temperature data were 16,500 Mwd/MTU for Sample 18 and 18,500 for 77, higher by ~10% than those of Table 2. Considering the problems described for the fuel and flux-monitor wire analyses, the results calculated from temperature data were chosen as the most reliable.

Some additional observations were made on surface replicas of a sample which had been put into the reactor in the solution-annealed condition with no fine particles present. They showed a fine particle precipitate ranging in size from the limit of resolution, about 0.03 microns up to about 0.5, and a visible particle density of about $10^{14}/\text{cm}^3$. There may have been many more smaller particles not visible. The significant point is that numerous particles did precipitate from this solution-annealed sample during irradiation exposure, just as planned for this type of sample. Each group of solution-annealed samples showed less swelling than the corresponding group in the precipitated state, probably because of a higher density of particles in the irradiated solution-annealed samples.

A comprehensive final report on the NRX-9-101 experiment, AI-AEC-12928, has been issued and a condensed version sent to the Journal of the American Ceramic Society for publication.

B. PREPARATION OF MODIFIED URANIUM-PLUTONIUM/CARBON

The purpose of this work is to develop methods for preparing (U,Pu)C-W alloys analogous to the UC-W alloys used in the NRX-9-101 swelling experiment.

The tungsten-doped mixed-carbide fuel samples will be subjected to a comparative-swelling test in an EBR-II irradiation. Accordingly it is desired to obtain (U,Pu)C-W alloys in two forms, the tungsten being in solution in one and dispersed as very fine particles of second phase in the other.

Surface replicas have been prepared of the first lot of $U_{0.8}Pu_{0.2}C - 1.6\% W$ sintered pellets. A problem arose in getting the alpha contamination (Pu) down to levels which would permit removal of the replicas from the plutonium facility for electron-microscope examination. Suitably clean replicas were obtained by paying careful attention to cleanliness in the glovebox and in all handling procedures, plus the use of an acid rinse to dissolve some surface particles from the acetate film. The replicas were removed from the plutonium facility for shadowing and then brought back for dissolving the acetate film in order that the Pu which was trapped in the film would be kept in the plutonium glovebox rather than contaminating another area.

Replicas of good quality were obtained. Structural features which were observed include grain boundaries, large square-shaped particles of second-phase (W or U), round pores and occasional clumps of gas bubbles. Figure 1 at a magnification of 9000X shows pores, grain boundaries, and scratches due to incomplete polishing. Figure 2 at the same magnification shows second-phase particles and grain boundaries, while Figure 3 shows the same particles at higher magnification, 21,000X. The particles are 1 to 2 microns across and there is no dense dispersion of much finer particles, 0.1 to 0.3 microns, such as has been made with UC-W alloys. However, these (U,Pu)C-W samples were in the solution-annealed condition and no particles would be expected. The next step is to anneal at an intermediate temperature in an attempt to precipitate particles.

Clusters of bubbles are seen in Figure 4, the spots of about 0.1-micron diam with long shadowtails. One possible cause of these bubbles is evolution of carbon monoxide during the high-temperature firing. Oxygen is known to be present due to surface contamination of the fine powder which was the starting material for the sintered pellets.

It has been found that even a 7-hr anneal at 2050°C was not successful in dissolving all of the W in a 1.6% mixture with (U,Pu)C powder, i. e. a part of

Figure 1. Surface Replica of $U_{0.8}Pu_{0.2}C - 1.6\% W$ Sintered at $2050^{\circ}C$ Showing Grain Boundaries and Pores but Absence of Fine Particles (900X)

Figure 2. Surface Replica of Same Material as Figure 1 Showing Grain Boundaries and 1-to 2-micron Diameter Grains of Second-Phase (tungsten or uranium) and Absence of Fire Particles (9000X)

AI-AEC-12948

Figure 3. View of Area Similar to That of Figure 2 at Higher Magnification (21,000X)

Figure 4. Surface Replica of Same Material as Figure 1 Showing Clusters of Gas Bubbles (21,000X)

the W was found in the form of the original grains, 10- to 20-micron size. Another approach to getting W in solution in (U,Pu)C is in process. Rods of UC-W containing 2.5% W in solution in UC have been prepared by arc-melting. This material will be crushed and mixed with PuC powder made from PuO_2 and carbon, and the mixture will be pressed and sintered. The basis for this approach is that PuC should be more readily homogenized with UC than was the W.

C. HELIUM INJECTION

Helium injection is under development as a technique to look at the behavior of rare gas in carbide materials because of the relevance of this behavior to fuel swelling. A paper on the initial phases of this study is scheduled for presentation at the Nuclear Division session of the annual American Ceramic Society meeting in Philadelphia in May. A further examination of surface replicas of annealed samples showed a band of helium bubbles twice as wide as the injected band (40 vs 20 microns) after 10 min at 1500°C and essentially no further change after 100 min at 1500°C. Two-thirds of the observed bubbles were attached to W particles. This diffusion distance of He bubbles in UC-W will be compared with that in UC with no particles present.

IV. EVALUATION OF EFFORT TO DATE

The work on developing mixed carbide fuel with a fine-particle dispersion was held up during most of this quarter but has now been reactivated. A procedure was established for making surface replicas of mixed carbide samples with low enough contamination for electron microscopy outside the plutonium area. A preliminary design for an EBR-II experiment to test swelling of this type of fuel is in readiness and further engineering of this proposed experiment can proceed when the method for preparing fuel has been demonstrated.

V. NEXT REPORT PERIOD ACTIVITIES

Experimental work on (U,Pu)C-W alloys will be continued until a method for preparing carbide fuel samples with a fine-particle dispersion has been worked out.

Program:	General Chemistry and Chemical Engineering		
AEC Task:	12-A, Sodium Chemistry		
Project Manager:	J. L. Ballif		
Reporting Period:	January-March 1970		
General Order:	7708	Subaccount:	36110
		AEC Category:	04-40-04-04.1

Principal Investigators: R. L. McKisson, R. L. Eichelberger, and J. Guon

I. PROJECT OBJECTIVES

The objectives of this project are to advance the understanding of the chemistry of liquid sodium as a solvent for metals and nonmetals through study of: (1) the solubility of selected metals and nonmetals in liquid sodium as a function of temperature by using the highest-purity materials available to minimize the perturbations caused by unwanted side reactions; (2) the nature of solute entities and their interactions in liquid sodium, from which thermodynamic properties of the solutions may be deduced; and (3) the analytical chemistry of solutes in liquid sodium at very low concentrations, in order that the data developed under (1) and (2) above may have the greatest possible reliability.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

A Memo report, "Solubility Experiments with Mo, Ti, Nb, Ni, and Cr in Liquid Sodium," was issued.

Experiments with oxidized nickel in sodium in the Sodium Chemistry Loop (SCL) have indicated that:

- 1) Na at 500°F, whether liquid or vapor, causes essentially complete reduction of oxidized Ni to metallic Ni.
- 2) The reduction of oxidized Ni in liquid Na proceeds by simple removal of O₂ rather than by a more complex reaction involving a soluble compound of nickel oxide and sodium or sodium oxide.

Experiments with acid-etched hydrogen-reduced 304 stainless steel in the SCL have indicated that:

- 1) A reversible reaction (adsorption) between this steel and O₂ occurs in liquid Na.

- 2) At 500°F a steel coupon equilibrated with sodium cold-trapped at 225°F (~0.6-wppm oxygen) adsorbed 3.8- $\mu\text{g oxygen/cm}^2$ when exposed to sodium containing 1-wppm (weight parts per million) oxygen; and steel equilibrated with 1-wppm oxygen in Na adsorbed an additional 1.4- and 1.9- $\mu\text{g oxygen/cm}^2$ when exposed to Na containing 13- and 19-wppm oxygen, respectively.

III. PROGRESS DURING REPORT PERIOD

A. SODIUM CHEMISTRY LOOP EXPERIMENTS

Tests were completed in which an acid-etched hydrogen-reduced coupon of 304 stainless steel was equilibrated with sodium at 500°F containing different known oxygen concentrations. The test sequence for these experiments is as follows:

- 1) Equilibrate stainless-steel coupon with Na-cold-trapped at 225°F.
- 2) Drain Na from the coupon station and isolate the station from the rest of the loop.
- 3) Inject oxygen into the Na and equilibrate at a predetermined cold-trap temperature.
- 4) Circulate the Na (now containing a known concentration of oxygen) over the steel coupon until equilibrium is attained.
- 5) Drain Na from the coupon station and isolate the station from the rest of the loop
- 6) Cold-trap the Na at 225°F
- 7) Circulate the cold-trapped Na over the steel coupon until equilibrium is attained.
- 8) Cold-trap the Na (still keeping the coupon in the system) at 225°F.
- 9) Begin next series.

Changes in the O_2 level are monitored with an electrochemical oxygen meter, which thereby serves to determine when equilibrium is reached in the system.

Two complete test series were completed. The O_2 concentrations in Na after equilibration for the two tests were 13 and 19 wppm, respectively. Subsequent equilibration with Na which had been cold-trapped at 225°F raised the O_2 level in the Na to about 1 wppm in each case. The results showed that the amount of O_2 on the steel was $1.4 \mu\text{g}/\text{cm}^2$ less at 1-wppm oxygen in the Na than it was at 13-wppm in the Na. The equivalent number for the 19-wppm test was $1.9 \mu\text{g oxygen}/\text{cm}^2$. The coupon apparently adsorbed $3.8 \mu\text{g oxygen}/\text{cm}^2$ more from sodium with 1-wppm oxygen than from sodium cold-trapped at 225°F (nominally 0.6-wppm).

An attempt made to perform a kinetic analysis of the data from the electrochemical oxygen probe was unsuccessful, probably because of the relatively long time constant of the probe.

B. METALS-IN-SODIUM SOLUBILITY STUDIES

Sixteen metal-solubility experiments were equilibrated in the range 600 to 900°C as follows: seven manganese (electrolytic deposit), five vanadium (zone-refined), and four cobalt (zone-refined). Of these only the first three Mn experiments have been analyzed, the other 13 results being delayed by a work backlog in the Analytical Chemistry Group.

The experiments completed were equilibrated at temperatures of 700, 800, and 900°C. When the capsules were opened it was found that in the 700 and 800°C tests the solute coupon had broken away from the mounting wire and was imbedded about 1/8-in. into the sodium. The appearance of the capsules suggested that the coupons had come loose during the machining operation to open the capsules, and had not fallen into the molten sodium when the capsule was inverted at temperature. Although the coupons were carefully removed from the sodium in which they were imbedded, it is probable that the results for these two experiments are too high. The results for the three runs at 700, 800, and 900°C are 200-, 54-, and 62-wppm manganese respectively.

The crucible-collector assemblies for all 16 experiments were fabricated from commercial Cb-1Zr alloy and were loaded with Na and welded closed in the ion-pumped vacuum facility used for loading capsules in previous solubility studies.

IV. EVALUATION OF EFFORT TO DATE

This program has been fruitful, and has resulted in substantial advances in understanding sodium and its interactions with metals, oxygen, and carbon. The work has been reported in topical reports issued when appropriate over the four years of the project, and two more are planned for the current fiscal year.

V. NEXT REPORT PERIOD ACTIVITIES

The chemical analyses needed to complete the solubility experiments which have been run will be made and the solubility results which have not yet been included in topical reports will be so reported. Several oxygen-meter tests will be run in the SCL, and this work will be terminated with the issuance of a topical report.

Program: Reactor Development		
AEC Task: 12-B, Fission-Product Contamination		
Project Manager: J. L. Ballif		
Reporting Period: January-March 1970		
General Order: 7708	Subaccount: 29410	AEC Category: 04-60-10-01.1

Principal Investigators: M. Silberberg, J. Guon, G. B. Zwetzig

I. PROJECT OBJECTIVES

The objectives of the project are to (a) determine the behavior of fission products and other radioactive contamination released to the sodium coolant in a fast reactor in order to provide information on the distribution of radioactivity between the sodium and the other system elements of the LMFBR, and (b) develop a digital-computer program for estimating the distribution of fission and corrosion products in the coolant, cover-gas, and primary-system components and on system surfaces of a LMFBR.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1970

A new integration method was developed for use in the Source Term Computer Program (STP-1), which substantially reduces the problem running time and increases the stability of the computation procedure. A variable-time step feature was incorporated in the code to provide further operating economies and increased accuracy.

Parametric engineering studies were begun with STP-1 for activated corrosion products. Preliminary results indicate that for the range of input variables studied the bypass cold-trap of an LMFBR primary loop operating at slightly increased utilization may be capable of reducing primary-loop contamination to levels consistent with contact maintenance for all areas except the cold-trap itself.

Experimental work on the cesium-sodium-stainless-steel system was completed. Although certain details of the system are still lacking, adequate quantitative information has been obtained to provide input for assessing the extent of radiological problems associated with cesium release to a LMFBR. Using

cesium data a preliminary estimate was made for FFTF. This estimate indicated that for 1% fuel failure and 10% cesium release, cesium adsorption will contribute insignificantly to the radiation level in the intermediate heat exchanger (IHX). The cesium level in the IHX will depend to a much greater extent on the ability to drain primary sodium from this component.

Experimental work on the barium-sodium-stainless-steel system was completed, though only data from the first test series (below saturation) were reduced. Reduced data indicate analogous behavior of cesium and barium. This is the first time this has been experimentally confirmed and represents a major advancement in the prediction of fission-product distribution in a LMFBR. A preliminary estimate was made for FFTF using available barium data. This calculation indicated that for 1% fuel failure and a sustained release of 0.1% per year of the barium, Ba radiation in the IHX would be 2 r/hr. A similar estimate was made for zirconium assuming a log-log dependence of K (the equilibrium-deposition constant) on solubility. This estimate indicated for 1% fuel failure and 0.1% per year sustained release rate, zirconium radiation in the IHX would be 0.7 r/hr.

III. PROGRESS DURING REPORT PERIOD

A. SOURCE TERM

1. Code Development

The following revisions and improvements were made to STP-1:

- a) The equations governing fission-product release from the fuel were revised to include the mechanisms of recoil, knockout, and direct release from near-molten regions in addition to the existing diffusion mechanism.
- b) The equations governing the rate of cladding failure were modified to permit the rates to be varied within each time block.
- c) More realistic fission-product adsorption and isotopic-exchange models were developed and incorporated in the code.
- d) Provisions for automatically initializing the adsorbed and cover-gas inventories were added to the code.

- e) Physically realistic limitations were imposed on the adsorption and vaporization-transport rates to suppress "overshoot" instability experienced in some test problems.
- f) A plotting (CRT) subroutine was developed, incorporated into STP-1, and checked out. Typical output for a simplified test problem is shown in Figures 1 and 2. Figure 1 shows the radiation levels as a function of time in the cold-half of the IHX and in the cold-leg of the primary loop. Figure 2 shows the surface deposit concentrations as a function of time for ^{54}Mn and ^{60}Co in the above two regions. Since the test problem was formulated for code development purposes only, the ordinate values shown in Figures 1 and 2 are illustrative only. More realistic results are given in Section A-2, below.

A draft of a User's Manual for STP-1 was prepared.

2. Engineering Studies

A preliminary series of parametric studies was started using STP-1 to investigate the effects of certain variables on the residual radiation levels produced by activated corrosion products. The basic system consisted of a 2500-Mwt LMFBR with an average neutron flux of 5×10^{15} nv. The average core-corrosion rate was 1 mil/year over a core surface area of $1.75 \times 10^7 \text{ cm}^2$, with chromium assumed to corrode at a rate equal to twice that of the other stainless-steel constituents. The volume of coolant in the core was $2.62 \times 10^6 \text{ cm}^3$ and the total coolant volume was $8.51 \times 10^8 \text{ cm}^3$. The several primary loops of the reactor were lumped together and divided into five regions: (1) hot-leg, (2) hot-half IHX, (3) cold-half IHX, (4) cold-leg, and (5) cold-trap. The average temperatures in these regions were as follows:

<u>Region No.</u>	<u>Component</u>	<u>Temperature (°F)</u>
1	Hot-leg	1060
2	Hot-half IHX	985
3	Cold-Half IHX	835
4	Cold-leg	760
5	Cold-trap	302

Figure 1. Sample Problem Radiation Levels,
Cathode-Ray Tube Output

Figure 2. STP-1 Sample Problem ^{60}Co and ^{54}Mn Surface Deposits, Cathode-Ray Tube Output

The total flow in the primary loop was 2.4×10^5 gpm and in the bypass cold-trap was 250 gpm.

The deposition model used in STP-1 for corrosion products assumes that partial precipitation of corrosion products will occur in those regions where the concentration of an element exceeds its solubility. The fraction of the concentration above saturation that precipitates per unit time in such a region is the precipitation efficiency for that region. The precipitation efficiencies used for the initial computations in Regions 1 to 4 were as follows: Region 1, 1.27×10^{-5} ; Region 2, 3.0×10^{-4} ; Region 3, 3.0×10^{-4} ; and Region 4, 7.82×10^{-5} . Investigation of the importance of this parameter is planned.

An additional factor considered in the calculations is that some of the deposited precipitate may re-enter the coolant either by erosion or by statistically sufficient energy to break its bonds to a precipitate clump. To represent this mechanism a re-entry factor of $0.02154 \exp(-4989/T(M) \text{ sec}^{-1})$ was used, where $T(M)$ is the coolant temperature ($^{\circ}\text{C}$) in Region M. This relation provides a fractional re-entry rate of 10^{-6} sec^{-1} at 500°C (932°F) and $10^{-10} \text{ sec}^{-1}$ at 260°C (500°F). Early investigation of the importance of this factor is planned.

Initial coolant concentrations of stable chromium, nickel, iron, manganese, and cobalt were set equal to their estimated solubilities at the cold-leg temperature, 760°F .

The following cases were considered:

Case 1: Precipitation efficiency in cold-trap = 0.006%

Case 2: Precipitation efficiency in cold-trap = 100%

Case 3: Precipitation efficiency in cold-trap = 10%.

(A similar result would be obtained for 100% cold-trap efficiency and 1/10 the reference flowrate).

Case 4: Precipitation efficiency in cold-trap = 10%, and corrosion-product solubilities are 1/10 their reference estimated values.

The radiation levels calculated by STP-1 for the various primary-loop regions for the above cases are shown in Figures 3 to 6. (The results shown in Figures 3 to 6 were plotted by hand since these studies were completed before the CRT subroutine for STP-1 had been completely checked out.)

Figure 3. Radiation Levels, Case 1

Figure 4. Radiation Levels, Case 2

Figure 5. Radiation Levels, Case 3

Figure 6. Radiation Levels, Case 4

Figure 3 shows that low cold-trap efficiency for removal of corrosion products from the coolant (or equivalently, limited cold-trap operation) would produce residual radiation levels in the primary loop of the order of 100 r/hr near the cold-half of the IHX and of the order of 1000 r/hr in the vicinity of the cold-leg piping. The inflection point in the cold-half IHX curve near 240 days is due to the fact that the concentration of Co in the coolant has reached the point where it has started to precipitate in that region.

Figure 4 shows the drastic reduction in primary-loop residual radiation levels which are achieved if the cold-trap is operated at about three times typical full-capacity flow (about 0.1% of total primary flow) and is 100% efficient in reducing coolant concentrations to saturation level at the cold-trap temperature. The only problem may be the 18,000 r/hr dose rate at the cold-trap.

Figure 5 shows that even if the cold-trap is only 10% efficient in corrosion-product removal, primary-loop radiation levels are suitable for limited contact maintenance except at the cold-trap.

Figure 6 shows that even if corrosion-product solubilities are 0.1, their estimated reference values,* and the cold-trap is only 10% efficient, serious residual-radiation levels occur only in the cold-trap.

The limitations of the above analysis should be noted:

- 1) The possible mechanisms of corrosion-product adsorption on the primary-loop walls and diffusion into the steel are not included in the present version of STP-1. This would not be expected to produce a significant change in the results, however, since these mechanisms can accommodate only a relatively small number of atoms. Further, since there are many more stable atoms of each corrosion-product element present in the coolant than radioactive atoms the vast majority of atoms fixed by these mechanisms would not be radioactive. Hence only very low residual radiation levels would be expected to result.
- 2) A more important limitation is the assumption that the corrosion products present are in elemental form. If, however, one or more is a highly insoluble compound, significantly higher residual-radiation

*R. L. Eichelberger, personal communication

levels may result. As noted earlier Figure 6 shows the effect of a solubility reduction by a factor of 10. An investigation of the effect of further reductions in solubility is planned for the next report period.

B. DEPOSITION STUDIES

A plot of $\log K(T)/K(700^\circ)$ vs $1/T$ for data from the first barium-test series is shown in Figure 7. For these tests nothing inconsistent with the K concept was found. The comparison of various parameters is sufficiently good to pretty well preclude the possibility of another deposition mechanism. It is interesting to note that the heat of activation for deposition of cesium (-3 kcal/gmole) is similar to the heat of activation for deposition of Ba (-3.5 kcal/gmole). This tends to bear out prior judgments that the values of K for these two elements is related to how strongly the sodium is rejecting the element, with the surface being a passive participant. If as more elements are studied a similar behavior is observed a generalized expression, perhaps based on solubility, can be developed.

The second Ba-test series (near saturation) was completed and data reduction is in progress. In the last week of the barium tests, four of the five heaters on test section No. 1 failed. These failures did not interfere with the completion of the test or taking of chemical samples. Each of these failures was internal (heating elements are Inconel-clad MgO-insulated nichrome) occurring in the nichrome adjacent to the transition to the nickel lead wire. Re-evaluation of the experimental program with respect to the next element to be studied is being completed before heater repair and resumption of loop operation. It is estimated that repairs will take less than one man-week.

Using available data preliminary radiation-level calculations were made for the IHX in the Fast Flux Test Facility. For Cs it was found that virtually all will remain in solution in the sodium. For Ba 2.5% will remain in solution with the remainder distributed on the surfaces. For zirconium, assuming a log-log relationship between K and solubility, 80% remains in solution with the remainder distributed on the surfaces. Two modes of release were assumed: a sustained release of 10%/yr for Cs and 0.1%/yr for Ba and Zr; and a burst release of 10% of the Cs and 0.1% of the Ba and Zr with sufficient circulation after

Figure 7. $K(t)/K(700^\circ\text{F})$ vs $1/T$ for Barium-Deposition Tests, $K(700) = 500 \text{ cm}$

release to homogenize the reactor system. Results of the calculation for a 1-yr operating time are reported in Table 1. It was assumed that all Na was drained from the IHX.

TABLE 1
FAST FLUX TEST FACILITY INTERMEDIATE HEAT
EXCHANGER RADIATION, 1-yr OPERATION

	IHX Radiation, (r/hr)	
	Burst Release Model	Sustained Release Model
Cs ($^{134}\text{Cs} + ^{137}\text{Cs}$)	0.004	0.004
$^{140}\text{Ba-La}$	0.54	2.1
$^{95}\text{Zr-Nb}$	0.35	0.7

IV. EVALUATION OF EFFORT TO DATE

The development of the Source Term Program is a significant achievement toward developing an economical engineering code for studying corrosion- and fission-product contamination of LMFBR's. The importance of STP-1 development is shown by the preliminary studies reported herein which indicate a potential means for drastically reducing LMFBR primary-loop residual-radiation levels with only moderate additions to existing plant equipment, on the basis of the range of parameters investigated to date. Further parameter explorations must be made and evaluated before firm conclusions can be drawn.

A major step has been made in both understanding and predicting deposition of cesium and barium in a LMFBR. Experimental studies have shown the barium can redistribute in both an isothermal and nonisothermal system. Previously it was believed (on "a priori" arguments because no experimental work had been done in this area) that once Ba found a deposition site, no further movement would occur. Because Ba is capable of movement through a system, in-line trapping procedures may be practical.

V. NEXT REPORT PERIOD ACTIVITIES

Parametric studies will be continued using STP-1 and extending studies to include fission products. The User's Manual for STP-1 will be issued.

Reduction of data from the second barium series will be completed. Following completion of the experimental program re-evaluation, the Fission Product Behavior Loop (FPBL) operation will be resumed with deposition studies on a new element. A topical report on FPBL studies with cesium and barium will be issued.