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QUARTERLY TECHNICAL PROGRESS REPORT
FUELS, MATERIALS, AND COOLANT
CHEMISTRY PROGRAMS
JULY-SEPTEMBER 1970

AEC Research and Development Report

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BY J H Kahn Lamb
DATE 12/4/70

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QUARTERLY TECHNICAL PROGRESS REPORT
FUELS, MATERIALS, AND COOLANT
CHEMISTRY PROGRAMS
JULY-SEPTEMBER 1970

The preceding Progress Report (Annual) was
AI-AEC-12971

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Program: Sodium Technology

AEC Task: Task 1, Phase I, Coolant Related Mechanical Properties

Project Manager: J. L. Ballif A. Mechanical Properties in Dynamic Sodium

Reporting Period: July-September 1970

General Order: 06507

Subaccount: 39140

AEC Category: 04-01-61-03.1

Principal Investigator: J. H. Shively

I. PROJECT OBJECTIVES

The objectives of Task A are: (1) to determine the effects of mass transport on the mechanical behavior of cladding and structural alloys under LMFBR system conditions; (2) to provide data relating the quality of the sodium as indicated by on-line instrumentation and conventional analytical methods to the degradation in biaxial creep and fatigue behavior of cladding and structural alloys; (3) to investigate the effects of the sodium system, its operation, and alloy type and composition being studied on the quality of the sodium; and (4) to provide a basis for extrapolating sodium impurity effects on mechanical behavior in a flowing sodium system under nonisothermal conditions to allowable materials property degradation in large engineering systems.

II. PROGRESS DURING REPORT PERIOD

A. SUMMARY

A second " ΔT Leg" experiment was initiated this month. The test involves the following: (1) biaxial stress-rupture specimens of the same lot of previous ΔT leg experiment but without wire wrap, (2) five heats of biaxial creep specimens in series, (3) two linear pressure specimens in support of FFTF gas plenum modeling, (4) a series of mass transfer rings for weight loss and carbon/nitrogen measurements, and (5) post-exposure stress-rupture specimens. The test conditions are displayed in Table 1.

Weight and counting measurements were completed on Type 304 and Type 316 stainless-steel rings exposed to flowing sodium under a high heat flux. Preliminary evaluation indicates the results duplicate those obtained in earlier

TABLE 1
TEST CONDITIONS

Test Temperature	1200° F $\pm$ 2.0 F°
Sodium Velocity	8.4 ft/sec $\pm$ 0.2 ft/sec
Leg Δ T	50° F $\pm$ 2.0 F°
Bulk Sodium Temperature	1150° F $\pm$ 2.0 F°
Δ T Axial-Test Section	12° F $\pm$ 2.0 F°

experiments on the two alloys for comparable conditions of heat flux, temperature, and time.

Carbon and nitrogen measurements were made on the cladding used in the first Δ T leg experiment. The results are presented in Table 2. Both carbon and nitrogen have migrated out of the cladding.

TABLE 2
NITROGEN AND CARBON ANALYSES Δ T LEG CLADDING

Specimen Number	Test	Exposure	Nitrogen, Two Samples (ppm)	Carbon (ppm)
6	Δ T-1	1200° F flowing sodium	679, 676	-
15	Δ T-1	1200° F flowing sodium	630, 645	-
25	Δ T-1	1200° F flowing sodium	618, 583	-
7	Δ T-1	1200° F flowing sodium	- -	164 (average)
Control	-	Unexposed	794, 769	570 (average)
363	Retort	1200° F static sodium	592, 580	540
360	Retort	1200° F static sodium	615, 581	620

Fabrication of a third Δ T leg experimental assembly was started. The assembly will be inserted in the loop following the completion of the present test. This test includes biaxial creep and stress-rupture specimens and corrosion rings.

B. EVALUATION

1. Carbon and Nitrogen Analysis

Carbon and nitrogen measurements were made on the cladding used in Task A. The carbon content was measured using the conventional combustion method. The nitrogen content was determined by the Kjeldahl method. The sample is dissolved in HCl, reacted with concentrated NaOH, and the resultant ammonia is distilled in a Kjeldahl flask. The distillate is reacted with H₂SO₄ plus Nessler's color reagent. The ammonium salts in solution are analyzed by absorption using a spectrophotometer.

The carbon analysis reported here in Table 2 and in the previous quarterly publication (AI-AEC-12971) show that carbon was removed during exposure to static sodium. The nitrogen analysis reported in Table 2 has an accuracy of 10 to 15%. On this basis, it is evident that the stainless steel has slightly lost nitrogen. The extent of denitriding was about the same in both static and flowing sodium. Therefore, the differences in properties cannot be explained by a loss of nitrogen in these two environments.

2. Second ΔT Leg Test - Description

The test conditions for the second " ΔT Leg" experiment in the 2-in. flowing sodium loop are presented in Table 1. The geometrical conditions are outlined in Table 3.

The test involves a combination of tests to determine the effect of flowing sodium on the mechanical properties of Types 304 and 316 stainless steels. This is accomplished by performing stress-rupture tests, biaxial creep tests, pressure ramp creep tests, weight loss measurements, and post-exposure stress-rupture tests. The specimen designs are shown in Figure 1. The individual test parameters are presented in Table 4. The locations in the array are shown in Figure 2. It should be noted that while the pressures are identical in the series tubes (see Figure 1), minor differences in wall thickness between lots cause slight differences in the hoop stress. To prevent early rupture, the stresses were set to give rupture in approximately 2500 hr on the weakest specimen in the chain, viz., Heat No. 03574.

10-14-70

6507-4740

Figure 1. Mechanical Properties Test Pins for Loop ΔT

10-14-70 UNCL

6507-4739

Figure 2. Test Configuration for ΔT Test No. 2
(View looking from outlet)

TABLE 3
TEST ASSEMBLY INFORMATION

Materials	10-15 CW Type 316 Stainless Steel Annealed Type 304 Stainless Steel Annealed Type 316 Stainless Steel
Cladding Dimensions (in.)	0.250 OD by 0.010 wall by 20 long 0.297 OD by 0.010 wall by 4 long
Strain Allowance in Bundle	7.7 $\Delta D/D$
Shroud Material	Annealed Type 316 Stainless Steel
Pressurization Gas	Helium
Pellet Dimensions (in.)	0.250 by 3.230 long
Pellet Material	Annealed Type 316 Stainless Steel
End Cap Weld Type	Electron Beam Lap
End Cap Material	Matches Cladding
End Cap Type	Solid Plug

Index lines were placed on the end caps of all tubes. Measurements made before exposure will be compared to those after exposure. The rings were weighed prior to exposure. A control sample has been kept back to determine the effects of handling and cleaning on the weight loss measurements after exposure.

It should be noted that Specimens 30 and 31 have failed. It is not known which heat of cladding failed in the chain. It is highly likely that the failures are in tubes for Heat No. 03574. The failures can be compared to the biaxial stress-rupture performance of annealed tubes of Type 316 stainless steel made from Heat No. 65808. This is displayed in Figure 3. At the present time, there does not appear to be any effect of flowing sodium on the properties of annealed tubing. However, without completion of the test, this conclusion is speculative.

III. EVALUATION OF EFFORT TO DATE

This task continues to provide data on the effects of flowing sodium on mechanical behavior of stainless steels. The current effort is aimed at studying

TABLE 4
SPECIMEN TEST PARAMETERS

Specimen Number	Pressure at Temperature (psi)	Time at Pressure (hr)	Stress for Several Heat Numbers and Materials (psi)				
			20013 304A	65808 316A	03574 316A	20303 316A	65808 316 CW
30	1440	1525	19,990	19,910	21,000	19,970	20,070
31	1440	1532	19,990	19,910	21,000	19,970	20,070
32	1440	1433	19,990	19,910	21,000	19,970	20,070
33	1440	1001	19,990	19,910	21,000	19,970	20,070
34	1440	600	19,990	19,910	21,000	19,970	20,070
36	1440	305	19,990	19,910	21,000	19,970	20,070
37	1440	114	19,990	19,910	21,000	19,970	20,070
38	1540	114	21,400	21,300	22,500	21,370	21,470
39	1540	194	21,400	21,300	22,500	21,370	21,470
45	1540	305	21,400	21,300	22,500	21,370	21,470
46	1540	500	21,400	21,300	22,500	21,370	21,470
47	1540	762	21,400	21,300	22,500	21,370	21,470
48	1540	1001	21,400	21,300	22,500	21,370	21,470
49	1540	1200	21,400	21,300	22,500	21,370	21,470
50	2117	Continuing					25,148
51	2117	Continuing					25,148
8	2117	Continuing					25,148
43 [†]	Ramp	Continuing					Ramp
44 [§]	Ramp	1250					Ramp

Specimens identical to ΔT Test No. 1 material.

[†]Two specimens of 316 CW, Ht. 65808, and two specimens 316A, Ht. 65808, filled with 1 atm argon at room temperature.

[§]Twenty-one mass transfer rings of cold-worked and annealed Type 316, Ht. 65808.

Figure 3. Comparison of Creep Rupture in Flowing Sodium With Reference Heat in Static Sodium

the effects of carbon losses on several heats of Type 316 stainless steel. Evidence of nitrogen removal from the stainless steel is a new finding of this program.

IV. NEXT REPORT PERIOD ACTIVITIES

The following major activities are anticipated for the next report period: (1) complete "heat to heat" flowing sodium test, and (2) initiate FFTF cladding test in flowing sodium.

TABLE 5
BIAXIAL STRESS-RUPTURE TESTS - 12 SPECIMENS PER RETORT

Retort Number	Test Temperature (°F)	Alloy	Cladding Condition ,†	Environment	Life Span (hr)	Status		Remarks	Start/Completed
						Test	Examination		
17	1200	304SS	Ann	Na	5000-10,000	3/4 Comp.	-	Extension of Data	2-22-68
22	1000	304SS	Ann	Na	5000-10,000	Comp.	-	Extension of Data	2-29-68/7-23-70
25	900	304SS	Ann	Na	1000-10,000	Comp.	-	Extension of Data	10-8-68/8-10-70
26	900	304SS	AR	Na	1000-10,000	Comp.	-	Extension of Data	3-27-69/8-21-70
110	1000	316SS	AR	Na	1000-10,000	3/4 Comp.	-	Extension of Data	4-23-68
113	900	316SS	AR	Na	100-10,000	In Prog.	-	Extension of Data	8-16-68
115	1000	316SS	AR	He	100-5000	3/4 Comp.	-	Effect of Sodium	10-18-68
116	1000	316SS	AR + Aged	Na	100-5000	Comp.	-	Effect of Thermal Aging	11-5-68/7-31-70
120	900	316SS	Ann	Na	1000-10,000	1/2 Comp.	-	Extension of Data	12-20-68
126A	1200	316SS	AR	Na	1000	On Test	-	Cleaning Study	8-20-70
127	1000	316SS	AR	Na	2000 Soak	Soak Comp.	-	Cleaning Study	1-18-70
130	1200	316SS	AR	Na	200-3000	Comp.	-	Lot to Lot Study	4-27-70/8-31-70
133	1200	316SS	Ann	Na	200-2000	In Prog.	-	Heat No. 03574	9-15-70
134	1200	316SS	Ann	Na	200-2000	In Prog.	-	Heat No. 20303	9-17-70

Ann - Annealed tubing

†AR As-received tubing containing 10 to 15% cold work

Program:	Sodium Technology		
AEC Task:	Task 1, 824 Contract		
	Phase I - Coolant Related Mechanical Properties		
Project Manager:	B. Mechanical Properties in Static Sodium		
	J. L. Ballif		
Reporting Period:	July-September 1970		
General Order:	06507	Subaccount:	39130
		AEC Category:	04-01-61-03.1

Principal Investigator: D. F. Atkins

I. PROJECT OBJECTIVES

The objective of this task is 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 Types 304 and 316 stainless-steel cladding alloys.

II. PROGRESS DURING REPORT PERIOD

Biaxial stress-rupture tests of Types 304 and 316 stainless-steel tubing were continued in multiple specimen static sodium retorts. The status of these retorts is given in Table 5. A number of these retorts have been under test for two years or longer and long-term stress-rupture data are becoming available. Table 6 lists the test conditions and rupture life of several long-term specimens which failed during this quarter.

During this period, five 12-specimen retorts completed biaxial stress-rupture test. These retorts contained a total of 36 specimens of Type 304 stainless-steel tubing and 24 specimens of Type 316 stainless-steel tubing. The retorts were drained of sodium and cleaned, and the specimens were submitted for profilometer measurements of the diameter. These measurements will be used to calculate the biaxial rupture strain of the specimens.

Twelve specimens of Type 316 stainless-steel tubing were biaxial stress-rupture tested in 1200°F sodium. The specimens were taken from the second re-draw lot of the reference heat, No. 65808. The data points are plotted in Figure 4 and are compared with the curve representing the initial lot of tubing. It is observed that there is little difference in the stress-rupture strength

Figure 4. Biaxial Stress-Rupture Comparison

TABLE 6
LONG-TERM BIAXIAL STRESS-RUPTURE VALUES ON TYPES 304
AND 316 STAINLESS-STEEL TUBING
(0.275 in. ID by 0.010 in. WALL)

Alloy Condition ^{*†}	Temperature (°F)	Environment	Hoop Stress (psi)	Rupture Life (hr)
Ann-304	1200	Na	13,000	21,896
Ann-304	1000	Na	37,000	20,942
Ann-304	900	Na	48,000	15,933
Ann-304	900	Na	48,000	14,605
Ann-304	900	Na	48,000	16,007
CW-304	900	Na	59,000	12,321
CW-304	900	Na	59,000	10,856
CW-316	900	Na	50,000	14,268
Ann-316	900	Na	48,000	14,883
Ann-316	900	Na	48,000	14,618
CW-316 [§]	1000	Na	40,000	12,181

*Ann = Annealed

†CW = Coldworked

§Thermally aged in 1000°F Na for 3000 hr prior to stress-rupture testing

of the two tubing lots. Both lots were processed by the same tube mill to the same specification. The data indicate that both lots have almost the same degree of cold work.

Twelve specimens of Type 316 stainless-steel tubing (10 to 15% coldwork) were assembled in a static sodium retort and placed on biaxial stress-rupture test at 1200°F. These specimens had previously been exposed to 1200°F sodium for 2000 hr and then subjected to various cleaning and storage procedures. The specimens are currently under 28,000-psi hoop stress and the stress-rupture data will be used to evaluate the cleaning and storage operations.

A second series of twelve Type 316 stainless-steel specimens were subjected to cleaning and storage procedures. These specimens had received a 2000-hr exposure to 1000°F sodium. After 1000-hr storage, they will be stress-rupture tested in 1000°F sodium.

Two 12-specimen retorts, containing annealed tubing from two new heats of Type 316 stainless steel, were assembled and placed on test in 1200°F sodium. The stress-rupture results on the as-received tubing (10 to 15% coldwork) from these two heats were reported previously. The results on the annealed material will be compared to separate coldwork effects from alloy chemistry variations. Three specimens in each retort are to be crept tested with the internal pressure being removed after 200, 500, and 1000 hr. The stress-rupture and creep data to be obtained from these retorts are required for base-line data in support of the current ΔT loop test on Task A.

A uniaxial stress-rupture test of Type 316 stainless-steel tubing (10 to 15% coldwork) ruptured after 787 hr in 1000°F sodium under a stress of 48,000 psi. The specimen strain at rupture was 2% and the steady-state creep rate was 1.5×10^{-3} %/hr.

A topical report, "Stress-Rupture Behavior of Types 304 and 316 Stainless-Steel Cladding in High Temperature Static Sodium," has been prepared and published.⁽¹⁾

III. EVALUATION OF EFFORT TO DATE

The static sodium testing program continues to provide data for comparison with flowing sodium tests. The test matrix for providing base-line stress-rupture data on the reference heats of Types 304 and 316 stainless steels has been completed. Testing in static sodium this fiscal year will concentrate on the effect of process variables with the main effort on providing support information for the flowing sodium tests.

IV. NEXT REPORT PERIOD ACTIVITIES

Biaxial stress-rupture testing will continue on the long-term tests involving Types 304 and 316 stainless-steel tubing.

Rupture strains and creep rates will be determined for the specimens in the five recently completed 12-specimen retorts. Metallographic examinations will be performed to evaluate structure in relation to stress level, rupture life, and fracture mode.

Retorts involving sodium cleaning and heat-to-heat studies will yield data for preliminary evaluation of these parameters.

REFERENCE FOR TASK 1-I-B

1. D. F. Atkins, "Stress-Rupture Behavior of Types 304 and 316 Stainless Steel Cladding in High Temperature Static Sodium," AI-AEC-12976 (September 30, 1970)

Figure 5. Rotating Beam Fatigue Tests for Type 316 Stainless Steel Tubing
(0.275 ID x 0.010-in. Wall) at 1200°F

Program: Sodium Technology

AEC Task: 1, 824 Contract, Phase I – Coolant Related Mechanical Properties
C. Interstitial Transport

Project Manager: J. L. Ballif

Reporting Period: July-September 1970

General Order: 06507

Subaccount: 39120

AEC Category: 04-01-61-03.1

Principal Investigator: D. F. Atkins

I. PROJECT OBJECTIVES

The objective of this task is to study the influence of interstitial compositional changes (initially carbon) on the engineering design properties of structural alloys to be used in high temperature sodium service. Conditions which produce both surface and bulk modifications to the alloy chemistry will be evaluated.

II. PROGRESS DURING REPORT PERIOD

Biaxial stress-rupture specimens of 316 stainless steel tubing (10 to 15% cold work) which contain a 2.5 wt % carbon case are under test in 1200°F helium. Stress-rupture data are given in Table 7. The as-received control specimen stressed to 33 ksi hoop failed in 278 hr. This value is in excellent agreement with the data previously generated on 316 stainless steel in 1200°F sodium. The case-carburized specimen stressed to 33 ksi hoop failed prematurely after 375 hr. The specimen has not been removed from the furnace but it is presumed that a weld failure occurred. The two other case-carburized specimens stressed to 35 and 40 ksi have not ruptured after approximately 2500 hr of test. This indicates that the carburization has greatly increased the rupture strength of the tubing.

Rotating beam fatigue tests at 1200°F in air were performed on both as-received and case-carburized tubing. Figure 5 shows a comparison of these data with an S-N curve for annealed material presented in the literature.⁽¹⁾ The displacement of the data points is due to the 10 to 15% cold work in the tubing material used in the current tests. The scatter in data is normal for fatigue tests. The limited data points for the case-carburized tubing were found to lie

9-3-70

6507-4018

Figure 6. Specimen Complex for Bulk Carburization

TABLE 7
BIAXIAL STRESS-RUPTURE DATA FOR AS-RECEIVED AND
CASE-CARBURIZED 316 SS TUBING
IN 1200° F HELIUM

Specimen Number	Condition	Hoop Stress (ksi)	Time to Rupture (hr)
62-14	As-received	33	278
C6-5	Case-carburized	33	375
C6-6	Case-carburized	35	2500 (No rupture)
C6-8	Case-carburized	40	2500 (No rupture)

within the scatter band for the noncarburized tubing at the lower strain levels. However, at 2% strain range, the case-carburized specimen seems to have greater fatigue strength. The lack of additional carburized specimens prevented further testing in this high strain region.

A rotating beam fatigue test was performed on annealed 316 stainless-steel tubing and it was found that strain hardening occurred under a 2% initial strain rate. A tensile test on the specimen after 21,435 cycles (no failure) revealed a significant amount of strain hardening. Strain hardening is not considered to be a problem with the 10 to 15% cold-worked tubing since the strains induced by the cold work exceed the level which could be induced during testing. However, an attempt will be made to modify the test equipment to provide a strain controlled test rather than the present load control.

Forty annealed 304 stainless-steel tubular specimens (Ht. 20013) were fabricated and diameter and wall thickness measurements were made. These specimens will be used in the bulk carburization studies.

A prototype retort for the bulk carburization studies was fabricated and placed on test. The retort utilizes the basic 12-specimen static retort design with the added feature of a pneumatically operated agitation device to mix the sodium. Figure 6 shows the agitator in relation to the test specimens. A special timing device (Figure 7) activates the agitator for eight cycles each hour. Twelve specimens of annealed Type 304 stainless steel were assembled for this initial test. The carbon source is 2-1/4 Cr - 1 Mo steel which is used in the agitator and the retort shell. The sodium temperature is 1100° F and the

9-28-70

Figure 7. Timer and Cycle Controller for Sodium Agitation

6507-4019

exposure time is planned to be 3000 hr. This should increase the carbon level in the specimens to 0.10 to 0.20 wt %. Three of the specimens in the retort will be stressed to obtain creep data during carburization and to evaluate the effect of stress on the rate of carburization. The remaining nine specimens will be unstressed and will be subsequently tensile and stress-rupture tested in 1100° F helium. Figure 8 shows the retort assembly mounted in the furnace just prior to the start of testing.

The fabrication of a second bulk carburization retort has been initiated. This retort will utilize a 1095 steel liner as the carbon source. The agitator will also be made of 1095 alloy.

III. EVALUATION OF EFFORT TO DATE

Initial tests on case-carburized Type 316 stainless-steel cladding indicate that the carburization enhances the biaxial stress-rupture strength of the alloy. This behavior was not entirely unexpected but one could have predicted premature ruptures if cracking developed in the brittle carbon case. Perhaps at a lower temperature, cracking might occur. High strain fatigue testing provides an ideal condition for the development of surface cracks in a brittle surface layer. However, on the limited fatigue tests conducted, no loss in strength was observed.

The design and construction of the retort for bulk carburization progressed smoothly with no major technical set-back. It appears that the retort will function in a consistent and reliable manner. The one area of concern is the potential fatigue of the bellows used to translate the drive to the sodium agitator. However, calculations of the stress on the bellows indicates that there is a considerable safety margin.

IV. NEXT REPORT PERIOD ACTIVITIES

Stress-rupture tests on case-carburized 316 stainless steel tubing will continue.

Rotating beam fatigue testing will be resumed on annealed and cold-worked Type 316 stainless steel with constant strain.

9-28-70

6507-4020

Figure 8. Bulk Carburization Retort Ready for Testing

The bulk carburization (Croloy-304 stainless steel system) test in 1100° F Na will be continued.

A new bulk carburization retort will be fabricated and assembled using 1095 steel as the carbon source.

REFERENCE FOR TASK 1-I-C

1. "A Study of Piping Materials for Sodium Piping in LMFBR Plants," CFB-4122-228, Technical Report 228, C. F. Braun & Company, April 1969

Program: Sodium Technology		
AEC Task: 1, Phase II: Sodium Quality Measurement		
Project Manager: J. L. Ballif		
Reporting Period: July-September 1970		
General Order: 06507	Subaccount: 39200	AEC Category: 04-01-61-03.1

Principal Investigator: E. W. Murbach

I. PROJECT OBJECTIVES

The objectives of this task are to: (1) apply on-line instrumentation to the experimental systems in support of the mechanical properties work, thereby providing reliable monitoring of sodium quality for correlation with the measured mechanical properties changes; (2) evaluate the currently available instrumentation for its applicability and reliability for continuous operation, and to evaluate advanced instruments as they become available; (3) develop and evaluate advanced purification methods for the removal of carbon, oxygen, and other deleterious impurities in large sodium systems; and (4) develop methods for controlling the carbon activity in systems which have both carbon sources and carbon sinks.

II. PROGRESS DURING REPORT PERIOD

A. LOOP OPERATION

The sodium supply loop was operated continuously during the quarter. Five retorts were loaded for the program and one vessel was loaded for the reactor safety program. The loop was cold trapped continuously at 250°F to insure low oxygen in the sodium.

The electrochemical oxygen meter continued to operate with emf readings of 1.161 ± 0.002 volts.

B. SAMPLING AND ANALYSIS

During the report period, eight retort samples were collected and the 2-in. loop was sampled three times. Three retort samples gave high oxygen results, 30 to 50 ppm. There was no obvious reason for the high results. One of the fittings on the vacuum manifold of the sampler was replaced as a precautionary measure and some clean-up of the analytical glovebox was accomplished.

The sodium samples from the 2-in. loop were all normal, i.e., approximately 5 ppm of oxygen and carbon less than 10 ppm.

C. VACUUM DISTILLATION

Five additional experiments were carried out with the in-line vacuum distillation sample.

In Run No. 2, distillation was continued for 6 hr with an indicated temperature of $\sim 650^{\circ}\text{F}$ at the crucible support pedestal. Upon opening the chamber at the end of the run, it was found that very little sodium had distilled.

The unit was modified by adding a thermocouple to dip into the sodium sample.

Run No. 3 was carried out at a distillation temperature of $\sim 685^{\circ}\text{F}$. A temperature differential of $\sim 100^{\circ}\text{F}$ was observed between the immersion thermocouple and the thermocouple in the heater pedestal. After 4 hr of heating, the temperature of the immersed thermocouple began to increase. After one additional hour, the run was terminated. When the unit was disassembled, a small amount of sodium was found in the sample cup. Further examination of the unit showed that the vapor line had become clogged with sodium which caused cessation of distillation.

The unit was modified by enlarging the vapor line and a second chamber was added at the bottom of the containment vessel to collect residual sodium not removed during the helium flushing step.

Run No. 4 was completed satisfactorily. The sodium was distilled at 670°F . After 4 hr, the temperature began to rise indicating that distillation was complete. After disassembly, the sample cup was rinsed with dilute HCl ($\sim 0.1\text{ N}$) and the resultant solution was analyzed by means of flame photometer. The final result, assuming all the sodium containing residue was Na_2O was 4 ppm of oxygen.

Run No. 5 was carried out at a distillation temperature of 650°F for 5 hr and then at 690°F for one additional hour. Approximately one-third of the sodium sample had distilled indicating that the distillation temperature was too low.

Run No. 6 was carried out at a distillation temperature of 675°F . After 6.5 hr, the temperature of the immersed thermocouple began to rise indicating that

distillation was complete. The cover of the chamber was maintained at $\sim 745^{\circ}\text{F}$ during the run. On disassembly no condensate was found on the cover as in previous runs. The crucible was rinsed with dilute HCl ($\sim 0.1\text{ N}$) and the sodium determined on a flame photometer. Again, assuming the sodium was Na_2O , the final result was 2 ppm of oxygen.

D. INSTRUMENTATION

Plans for modification of the 1-in. loop for carbon meter testing are continuing. The piping layout has been completed and detailed drawings prepared. The pipe and welding fitting have been received as have the internal heaters for the carbon injector and the sodium heater.

Two electrochemical carbon meters have been received.

The new hot trap has been received and was helium leak-checked. No leakage was found. It is planned to install the hot trap at the same time the loop is down for installation of the instrumentation package.

E. OXYGEN DETERMINATION BY WIRES

Two 10-mil wires, one vanadium and one niobium furnished by ANL, were mounted on a holder and inserted in the sample station on the 2-in. loop. Sodium was circulated through the sample station for 24 hr at 1070 to 1120°F . The sample was chilled rapidly and transferred to a glovebox. The sodium in the sample tube was melted to remove the wires. The residual sodium on the wires was removed by reaction with isopropyl alcohol. The wires have been returned to ANL for oxygen determination.

III. EVALUATION OF EFFORT TO DATE

The latest results on the in-line vacuum distillation samples have shown that the method is operable. By flushing the samples with sodium, surface oxides should be effectively removed so that the only blank involved in the analysis is the sodium blank in the solution used to rinse the sample cup following distillation.

One problem with the current design is the poor heat transfer between the heater pedestal and the sample cup. For the last run, an overall heat transfer

coefficient of ~ 35 was calculated. It is planned to modify the sample cup by welding it to a base which will be a slip fit over the heater pedestal.

The modifications to the 1-in. loop have been delayed by late delivery of the electrochemical carbon meters and by a shortage of funds.

The routine operations of dispensing, sampling, and analysis of sodium continue to be carried out without difficulty.

IV. NEXT REPORT PERIOD ACTIVITIES

Activities for the next quarter in loop modifications and instrumentation will depend on scheduling and funding. Detailed plans for the 2-in-loop modifications are required.

Routine operations of retort loading, sampling, and analysis will continue on an as-required basis.

Program:	Sodium Technology		
AEC Task:	1, Phase III: Special Compatibility (Sodium Frost)		
Project Manager:	J. L. Ballif		
Reporting Period:	July-September 1970		
General Order:	6507	Subaccount:	39300
		AEC Category:	04-01-61-03.1

Principal Investigator: L. G. Kellogg, E. Matlin

I. PROJECT OBJECTIVES

The objectives of this work are to evaluate the transport of sodium vapor and its compounds (frost) and its subsequent deposition within the annuli of the IVHM rigid arm and rotating plug. Also, it is an objective to verify the argon purge rate of the reference concept for its effectiveness in suppressing xenon diffusion and sodium-vapor transport into the annuli.

II. PROGRESS DURING REPORT PERIOD

None. This project has been temporarily stopped pending a decision on work direction toward a basic information program.

III. EVALUATION OF EFFORT TO DATE

None.

IV. NEXT REPORT PERIOD ACTIVITIES

Redirection of this project is under consideration by the Coolant Chemistry Branch, Division of Reactor Development and Technology, toward more specific FFTF needs.

Program: Sodium Technology		
AEC Task: 1, Phase IV: Radioactivity Transport		
Project Manager: J. L. Ballif		
Reporting Period: July-September 1970		
General Order: 06507	Subaccount: 39410	AEC Category: 04-60-10-01.1

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

I. PROJECT OBJECTIVES

The objective of this work is to develop sufficient understanding of the problems of radioactive material transport to permit realistic evaluation of radiation problems that may affect maintenance or repair of a LMFBR primary system. The work includes studies of the transport of both corrosion and fission products.

II. MAJOR ACCOMPLISHMENTS DURING FY 1971

The limited post-shutdown examination of the SNAP 8 Developmental Reactor (S8DR) to assess radioactivity-transport problems related to LMFBR (FFTF) was completed. The results were summarized in a report "Some Observations on Mass and Radioactivity Transfer in the S8DR," TI-707-33-002 (July 24, 1970) and issued to RDT.

An abbreviated test series on manganese deposition was completed in the Fission Product Behavior Loop.

III. PROGRESS DURING REPORT PERIOD

A. FISSION PRODUCT BEHAVIOR LOOP

The systematic study of the transport of manganese in a sodium system was started. After sufficient manganese was injected into the loop (started in Fiscal Year 1970), the initial portion was completed of a parametric test series similar to that used to study barium deposition. The test series involved varying the temperature of one test section while maintaining constant the temperature of the rest of the loop. The experimental portion of the program was terminated prematurely because of a change in current Task 1 priorities. Sufficient data were obtained to permit some evaluation of the manganese/sodium-stainless-steel system. While data reduction is not complete, preliminary results indicate that

manganese is not as strongly adsorbed onto stainless steel as would be expected from a priori consideration and the adsorption constant has a positive temperature coefficient. These results are consistent with the results from the S8DR examination. (See Section III-C.)

The final set of chemical samples have been taken from the FBPL and submitted for analysis.

B. S8DR LOOP SURVEY

Evaluation relative to radioactive specie transport from the analysis of available S8DR data and post-shutdown examination was completed. Results are reported in detail in the referenced publication. (See Section II.) In brief, the results of the study can be summarized as follows:

- 1) The deposition of the corrosion products in the cold leg piping was in the form of a dark powder (~ 0.5 mil thickness), which could be removed by vigorous wire brushing. The major constituents in the deposit were identified as Fe (austenite) and NaCrO_2 . By contrast, the hot leg piping was free of a visible deposit, and actually showed evidence of corrosion.
- 2) Comparison of the corrosion product deposition rate observed on the cold leg piping with predictions from empirical equations (G. E. equations) derived from LMFBR mass transfer loop experiments shows drastic differences. Specifically, the observed deposition rate is more than an order of magnitude higher than the predicted rates. It is recognized that differences exist between the S8DR system and the loop systems, when making such a comparison. The differences between the observations made in this study are of major concern.
- 3) Shutdown radiation levels from pipe elbows (both hot and cold leg), and from the pump outlet, were appreciable fractions of, or greater than, the radiation from the cold end of the IHX.
- 4) The deposition pattern in the primary loop is consistent with simple mass transfer consideration — higher deposition occurs in regions of higher Reynolds number (e.g., pipe elbows vs straight pipe).

- 5) Radiation levels in the hot leg were not insignificant when compared to those observed in the cold leg and IHX.
- 6) The activated corrosion products, Cr^{51} , Co^{58} , Co^{60} , and Mn^{54} , were detected on both the hot leg and cold leg piping. Deposition of Co and Mn were enhanced in the hot leg. Preferential deposition of Cr was not observed.
- 7) Fission product deposition was observed only in the cold leg; and, if present in the hot leg, was below the limit of detection. The fission products detected in the cold leg were Ba-La¹⁴⁰, Ce¹⁴⁴, Nb⁹⁵, Sr⁸⁹, and Sr-Y⁹⁰.
- 8) The results from the S8DR examination support the data obtained thus far in the Fission Product Behavior Loop — that cesium is weakly adsorbed, and barium is strongly adsorbed, on stainless steel.
- 9) New information has been obtained on the behavior of three important radioisotopes, Co^{60} (and Co^{58}), Mn^{54} , and Nb⁹⁵. Cobalt and manganese are strongly adsorbed onto stainless steel; niobium is poorly adsorbed, and should be reasonably mobile in an LMFBR.
- 10) The data from the one NaK flush (system drained, refilled, and circulated for 24 hr) indicate that decontamination by liquid metal flushing is effective for Cs and Nb (weakly adsorbed), but not for Ba-La¹⁴⁰ (strongly adsorbed).
- 11) Post-shutdown radiation levels predicted from STP-1 are in general agreement with gross radiation levels observed following the draining of the primary loop. An improved evaluation in this area will be possible, when final radiation levels are obtained from the loop gamma scan.

The following recommendations are made:

- 1) Detailed evaluation of discrepancies and normalization of the STP-1 Code should be performed.
- 2) Radioactivity transfer problems in FFTF and the LMFBR Demonstration Plant should be re-evaluated using updated technology derived from this study.

- 3) Sufficient information is not available, at the present time, to extrapolate the results from a given reactor to confidently predict mass and radioactivity transfer in a different reactor, without conducting additional surveys and complementary laboratory tests required for proper correlation and scaling between reactor systems.
- 4) A recommended method should be prepared for planning and performing mass and radioactivity transfer surveillance studies on existing and future LMFBR's.
- 5) Immediate planning should be carried out to apply the techniques and requirements in Item 4 to operating reactors, such as EBR-II, Fermi, and, if possible, Rapsodie and DFR. Following proper planning, system surveillance programs should be initiated for these reactors.
- 6) Detailed information should be obtained on the results of any radiation survey and/or radiochemical analyses performed during the current Rapsodie system modification shutdown.
- 7) When the S8DR IHX becomes available in the future, it should be sectioned, and examined for corrosion product deposits, in connection with possible degradation of heat transfer characteristics in an LMFBR IHX.
- 8) The computations required to derive radiation dose levels from the primary loop gamma scan should be performed to assist with STP-1 normalization.

IV. EVALUATION OF EFFORT TO DATE

The premature termination of the manganese test series will restrict to some degree a complete quantitative interpretation of test results. Assuming the applicability of the equilibrium constant approach to the manganese/sodium/stainless-steel system, suitable parameters can be developed that will explain FPBL data and hopefully can be applied to reactor calculations. The post-shutdown examination of the S8DR primary loop has provided significant supplementary information for the systematic study of radioactivity transport and provides an excellent base for developing detailed LMFBR primary system surveillance procedures.

V. NEXT REPORT PERIOD ACTIVITIES

Reduction and analysis of data from the Mn deposition test series will be completed and summarized.

Program:	Radiation Effects		
AEC Task:	2-A, Irradiation Damage in Cladding and Core Structural Material		
Project Manager:	H. Pearlman		
Reporting Period:	July-September 1970		
General Order:	06506	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 fluence, temperature, helium concentration, and microstructure on void formation and helium embrittlement; and (3) perform preliminary evaluations of modified and new alloys developed by other laboratories.

II. PROGRESS DURING REPORT PERIOD

A. VOID FORMATION IN TYPE 316 STAINLESS STEEL

Four samples of Type 316 stainless steel were proton irradiated and the irradiation conditions of these samples appear in Table 8. Sample 56 had one half irradiated to a higher fluence by shielding the other half after a fluence of 1.9×10^{19} p/cm² had been achieved. The two halves are designated as 56A and 56B. Void formation at the end-of-range region of the low-fluence part of the sample will be compared with void formation at the front portion of the high fluence part.

TABLE 8
PROTON-IRRADIATED SAMPLES OF TYPE 316 STAINLESS STEEL

Irradiation Number	Sample Number	Treatment	Atom Fraction He (x 10 ⁻⁶)	Temperature (°C)	Flux (p/cm ² /sec)	Fluence (p/cm ²)
4-1	56A	6*	5.9	500	1.6×10^{15}	1.9×10^{19}
4-2	56B	6	5.9	500	1.6×10^{15}	6.7×10^{19}
4-3	47	6	5.8	400	9.0×10^{14}	2.3×10^{19}
4-4	38	6	5.8	600	9.8×10^{14}	2.3×10^{19}
4-5	6 - 25	6 + 25% CR	5.9	500	7.9×10^{14}	7.0×10^{18}

*Treatment 6: 1 hr 982 °C + 8 hr 760 °C

Examination of Samples 28, 56A, and 56B has yielded data that are shown on Table 9, along with previous results. A plot of swelling as a function of vacancy production appears in Figure 9. The irradiation damage is now expressed as vacancy production on an atomic basis, displacements per atom, instead of displacements per unit volume. This will allow a meaningful comparison among alloys having significantly different numbers of atoms per unit volume.

TABLE 9
VOID FORMATION IN TYPE 316 STAINLESS STEEL,
PROTON-IRRADIATED AT 500°C

Sample*	Damage (displacements/ atom)	Void Size (Å)	Void Density (/cm ³)	Volume Increase (%)
28	0.5	47	5×10^{15}	~0.04
6	0.8	110	3×10^{15}	0.2
6	1.2	70	1×10^{16}	0.2
36	2.4	190	9×10^{14}	0.4
36	3.2	150	5×10^{14}	0.1
6	4.1	150	5×10^{15}	1.0
36	9.8	230	3×10^{15}	2.1
56A	16	260	7×10^{15}	6.0
56B	55	-	-	≥10

*Samples contained between 2 and 9 appm helium

The volume change increases linearly with vacancy production up to about 6%. The next data point (~10%) lies significantly below the straight line if extrapolated. That point comes from Sample 56B where the dislocation density is so high that it is difficult to observe all the voids present. Further electron microscopy is being done on this sample. Figure 10 shows the void formation in that sample and in 56A.

Sample 63, containing no helium and irradiated at 500°C, had only a few voids where the vacancy production was 4.7 displacements/atom. This result demonstrates the effect of helium since a volume change of about 1% would be expected had helium been present.

70S9-86-9

Figure 9. Volume Increase as a Function of Vacancy Production in Proton-Irradiated Type 316 Stainless Steel at 500°C

56A

16 DISP./ATOM

56B

55 DISP./ATOM

1000 Å → | ←

1000 Å → | ←

Figure 10. Transmission-Electron Micrograph of Voids in Samples 56A and 56B

B. CREEP OF TYPE 316 STAINLESS STEEL

Experimental difficulties have prevented the initiation of creep of helium containing samples at 700°C. This work should commence shortly.

C. EBR-II IRRADIATIONS

Two AI capsules in ORNL X034A and one in ORNL X035 were removed from the EBR-II. The estimated peak total fluence of the former capsules is 8×10^{22} n/cm² and that of the latter capsule is 5×10^{22} n/cm². Other AI samples in ANL X018B were also removed from the EBR-II after an estimated peak total fluence of 3×10^{22} n/cm².

D. EVALUATION OF STAINLESS STEELS FROM OTHER LABORATORIES

Type 316 stainless steel from PNL and ORNL is being rolled to thicknesses of 0.0005 in. and 0.008 in. for proton irradiation and helium injection. The shock-hardened Type 316 stainless steel from BMI will be ground down to 0.0005 in. for proton irradiation but no tensile samples will be made owing to the poor surface of the 0.008-in. strip.

III. EVALUATION OF EFFORT TO DATE

Results on the effect of irradiation damage on swelling have been quite valuable. The data that have been produced are unique and it is unlikely that such high swelling values ($\sim 10\%$) will be achieved by reactor irradiations in the near future. We will soon be able to study other variables of the swelling process at useful amounts of radiation damage.

Progress has not been satisfactory on creep testing of Type 316 stainless steel with helium. However, no major problems exist and this effort should begin to give results in the next quarter.

IV. NEXT REPORT PERIOD ACTIVITIES

(1) Type 316 stainless steel will be proton-irradiated to a very high fluence, (2) creep tests of Type 316 stainless steel with helium will begin, (3) examination of proton-irradiated Type 316 stainless steel will continue, (4) samples for proton irradiation will be injected with helium, and (5) fabrication of samples of Type 316 stainless steel from other laboratories will be completed.

Program: Fuel Cladding and Structural Materials		
AEC Task: 2-B, Integral (n, α) Cross Section Measurements		
Project Manager: H. Pearlman		
Reporting Period: July-September 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. PROGRESS DURING REPORT PERIOD

Approximately 150 specimens of the elements and compounds Be, Si, V, Mn, Co, MnF₂, FeF₂, NiF₂, and PbS have been prepared for mass spectrometric helium analysis. These samples were irradiated at five different locations in and out of the core of EBR-II during the dosimetry test Run 31F. Individual etching of the irradiated pieces was followed by cutting them into two or more specimens for duplicate analysis, microscopically examining them for surface impurities and finally weighing them. The fluorides are to be analyzed because the integrated (n, α) cross section of fluorine is needed for the subtraction of the fluorine contribution to the helium production in LiF, NaF, and CaF₂.

A new multiple-sample oven similar in most respects to existing ovens has been designed and built with a much smaller volume. This should increase the sensitivity of the gas mass spectrometer system by lowering the helium leak-rate into the oven through the glass walls and by increasing the fraction of the gas sample that is taken for mass spectrometric analysis.

Two methods are being investigated to extract helium from high melting-point-temperature materials. One is the use of a platinum flux to dissolve the specimen, using an HF induction furnace that is part of the recently-designed system to measure extremely small quantities of nitrogen and hydrogen in metals. The other depends on laser energy for melting, and is being evaluated using an available laser to conduct preliminary tests. Irradiated compounds such as TiN, HfN, TaN, SiC, and TiC whose melting points range from 2700 to 3360°C should give information on the (n,α) cross sections of N and C.

Helium analyses of 32 additional specimens of neutron-irradiated Si, Fe, Ni, PbS, and Type 304 stainless steel have been completed. The results have been added to a table now containing a matrix of 119 measurements of helium concentrations in Fe, Ni, Al, Cr, Si, and two heats of Type 304 stainless steel resulting from neutron exposures at the five locations in EBR-II. In nearly all cases, each original sample was cut into two or more pieces for separate helium analysis. The resulting data are in excellent agreement, except in a few cases where the duplicate specimens showed small but significant differences. Twenty tungsten wire coils that had been used to melt and vaporize those duplicates with the lower helium concentrations were then reheated by placing them within new tungsten coils. Most of the reheated coils released additional amounts of helium (ranging from 0.1 to 10 parts per billion) which were just enough to remove the discrepancies. Final agreement in the data for duplicate specimens strongly indicates a complete recovery of helium. Remaining differences will be reduced partly by adjustment for small spatial separations. Correlation of helium concentrations with fluence data obtained from the same EBR-II dosimetry test, Run 31F, is progressing in cooperation with personnel at WADCO.

Progress in the task was reviewed with the USAEC in Washington on September 22-23, 1970. This was followed by a visit to ORNL where cooperation in future experiments and irradiations was discussed.

III. EVALUATION OF EFFORT TO DATE

The helium measurements in irradiated Al, Si, Cr, Fe, Ni, and Type 304 stainless steel for the five widely differing EBR-II locations have given rather precise values of the integrated (n,α) cross sections of these elements. Comparison with the steel results, moreover, have shown that up to 70% of the helium

generated in the stainless steel cannot be attributed to the major steel components, and must therefore have come from (n,α) reactions with trace constituents.

The mass spectrometer system has been working very well with a helium sensitivity of less than 0.05 parts per billion (atomic) for samples weighing a few milligrams. The next most important elements and compounds to be studied — B, Al_2O_3 , TiN, HfN, TaN, SiC, TiC, and ZrC — all have melting points well above 2000°C and therefore require a higher temperature method of helium extraction. If the new method that must be devised proves more convenient and displays a 100% helium recovery on all specimens without any important disadvantages, it might replace the existing system. While this is being devised, however, other important elements and compounds with lower melting points, such as Be, V, Mn, Co, MnF_2 , FeF_2 , and NiF_2 , will be analyzed using the existing method of helium extraction.

Although very high purity elements and compounds were used in this experiment, it is nevertheless necessary to determine what contributions of helium if any, come from (n,α) reactions with impurities. At present, the principal impurity sources of helium are thought to be N and B and to a lesser extent Si, S, P, C, and O. Thus, before final (n,α) cross sections can be reported, it is necessary to perform further analyses for impurity content on each substance. Fortunately all samples were taken from the same sources so that one analysis of each substance by spark source mass spectrometer at Atomics International will accomplish most of this effort, giving a complete elemental impurity analysis, with the exception of some of the gases that appear in the atmosphere. Analyses of nitrogen concentrations will be performed with a new technique developed at Atomics International but these will have to be done both before and after irradiations because N evidently is very mobile under some circumstances.

IV. NEXT REPORT PERIOD ACTIVITIES

Correlation of helium concentrations at the five EBR-II locations with WADCO fluence data will continue.

Preliminary experiments will continue to establish whether laser or induction furnace methods of extracting helium can be used effectively with the high sensitivity gas mass spectrometer system.

Plans will be made in cooperation with WADCO for submitting capsules containing pure elements and compounds for the next EBR-II dosimetry test at the higher power level.

Program:	LMFBR Technology	
AEC Task:	3, Fast Reactor Fuels	
Project Manager:	C. W. Wheelock	
Reporting Period:	July-September 1970	
General Order:	06515	Subaccount: 28210 AEC Category: 04-40-02-01.

Principal Investigator: G. Ervin, Jr.

I. PROJECT OBJECTIVES

The objectives of this task are to: (1) determine selected microstructure attributes and mechanical and physical properties of mixed carbide fuels, both to establish the potential for these materials as nuclear fuels and to provide a basis for interpreting and understanding irradiation behavior in terms of atomistic transport phenomena; and (2) develop, where necessary, preparative techniques for polycrystalline mixed carbide materials of controlled structure and stoichiometry.

II. PROGRESS DURING REPORT PERIOD

A. PREPARATION OF MODIFIED (U,Pu)C

Carbothermic reduction of a mixture of the oxides UO_2 , PuO_2 , and WO_3 followed by sintering at 2050°C , has produced single phase monocarbide, $\text{U}_{0.8}\text{Pu}_{0.2}\text{C}$, with tungsten in solid solution. The evidence for this is optical metallography which shows single phase monocarbide with no indication of higher carbide or free metal and lattice parameters of the monocarbide which are sharply lower than for pure $\text{U}_{0.8}\text{Pu}_{0.2}\text{C}$. Experiments are in progress to precipitate the dissolved tungsten by annealing at intermediate temperature. Lattice parameter data for preparations containing 1.6% W are as follows:

	<u>Major Phase</u>	<u>Minor Phase</u>
Initial material, 1.6% W, sintered at 2050°C	4.945 Å	4.957 Å
Subsequent anneal at 1600°C	4.947 Å	4.948 Å
Subsequent anneal at 1450°C	4.947 Å	4.961 Å
Subsequent anneal at 1350°C	4.956 Å	none
Tungsten-free $\text{U}_{0.8}\text{Pu}_{0.2}\text{C}$	4.969 Å	-

The last item, tungsten-free carbide, is the figure obtained for a sample of mixed carbide which we purchased from United Nuclear Corporation. The figure for the United Nuclear material is in good agreement with published lattice parameter data for mixed carbide.

The much lower figure for the 1.6% W material sintered at 2050°C, 4.945 Å, compared with 4.969 Å for unmodified carbide indicates that the tungsten is in solid solution in the monocarbide lattice. Minor amounts of a second phase with higher lattice parameter is an indication of inhomogeneous distribution of tungsten. Annealing at 1600°C did not precipitate tungsten to any extent but apparently caused some improvement in homogeneity. Annealing at 1450°C precipitated little if any tungsten, but at 1350°C a significant proportion of the tungsten was forced out of solution as shown by the increase in lattice parameter from 4.945 to 4.956 Å.

Surface replicas of two of these samples have been prepared, the one in the as-sintered (2050°C) condition and the other in the 1350°C annealed condition. On the basis of the lattice parameter data, it is to be expected that the as-sintered sample will not show tungsten particles but that the 1350°C sample will. Only the replica of the 1350°C sample has been examined. A few particles were found but they were about 1 micron in diameter or about ten times as large as desired. Also the optical microstructure is puzzling in that it indicates considerable grain growth during this relatively low temperature anneal. The best conclusion at this time is that more experimental work is needed, perhaps annealing at still lower temperature.

B. HELIUM INJECTION STUDIES

Surface replicas of thermal-gradient-annealed samples of helium-injected UC-W have been prepared and both helium bubbles and tungsten particles have been observed. The specimens as irradiated in the cyclotron contained a band of helium about 7 mils below the surface. The annealing was performed at a top temperature of ~1550°C with a thermal gradient of 800°C/cm in the direction of the irradiated face, by placing this face against an induction-heated tungsten cylinder, with water-cooled copper pressed against the other end. The replicas show that helium bubbles moved up the thermal gradient a substantial distance, i.e., 3 or 4 mils, toward the heated face. More detailed examination

and counting of bubbles will be needed to evaluate the effect of the particles on the bubble motion; however, the observed bubble motion is itself of great significance to understanding fuel swelling. This much bubble motion is orders of magnitude greater than is predicted by current bubble migration theory but agrees with an earlier report* on helium bubble migration in UC. The observation of extensive bubble motion out-of-pile also tends to discredit the popular argument that fission gas is transported in fuel primarily by bubble resolution and atomic diffusion, rather than bubble motion.

Some effort has been made toward preparing thin foils for transmission electron microscopy of helium-injected-and-annealed specimens, but routine difficulties have prevented success thus far. The transmission technique is a superior way to observe bubbles and bubble-particle associations, and our earlier success in photographing thin foils of tungsten-containing UC is evidence that the technique is feasible.

III. EVALUATION OF EFFORT TO DATE

A method of preparation of tungsten-modified mixed carbide (i.e., carbo-thermic reduction of a mixture of UO_2 , PuO_2 , and WO_3) was developed to the point that suitable fuel for a fission gas swelling test could be fabricated. Further intermediate temperature annealing studies are desirable to find conditions for converting the solid solutions to structures containing a high density of small particles.

The helium injection work has made some progress in demonstrating qualitatively that helium bubbles migrate appreciable distances (mils) in a thermal gradient in the absence of an irradiation environment. The work has been slowed by problems of developing dependable electron microscope procedures for observing bubbles.

IV. NEXT REPORT PERIOD ACTIVITIES

Topical reports will be written on the two studies as final reports of progress to date for this project, which is now terminated. Equipment assigned to the project will be disconnected to make it available for transfer or other disposition.

*M. A. DeCrescente and E. G. Selleck, "Fission Gas Migration Studies in Uranium Carbide," PWAC-476, October 1965