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Nuclear Materials & Propulsion Operation

**HIGH-TEMPERATURE MATERIALS PROGRAM
PROGRESS REPORT No. 54, Part A**

December 20, 1965

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GENERAL ELECTRIC

ATOMIC PRODUCTS DIVISION

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TID-4500 (47th Ed.)

**HIGH-TEMPERATURE MATERIALS PROGRAM
PROGRESS REPORT No. 54, Part A**

December 20, 1965

United States Atomic Energy Commission

Contract No. AT(40-1)-2847

**NUCLEAR MATERIALS and PROPULSION OPERATION
ATOMIC PRODUCTS DIVISION**

GENERAL ELECTRIC

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CONVERSION TABLE

To Convert From	To	Multiply By
Atmospheres	Pounds/inch ²	14.7
Calories (mean).....	Btu (mean)	0.00397
Calories/gram-°C	Btu/pound-°F	1.0
Calories/sec-cm-°C	Btu/hr-ft-°F	241.8
Calories/sec-cm ²	Btu/hr-ft ²	1.32 x 10 ⁴
Calories/sec-cm ² -°C	Btu/hr-ft ² -°F	7370
Centimeters	Feet	0.03281
	Inches	0.3937
Cubic centimeters	Cubic feet	3.531 x 10 ⁻⁵
	Cubic inches	0.06103
Grams	Pounds	0.002205
Grams/cm ³	Pounds/ft ³	62.43
Grams/cm ²	psi	0.01422
Kilograms	Pounds	2.205
Kilograms/cm ²	Atmospheres	0.9678
	Pounds/ft ²	2048
	Pounds/inch ²	14.22
Kilograms/mm ²	Pounds/inch ²	1422.32
Kilowatts	Btu/sec	0.948
Liters	Cubic feet	0.0353
Meters	Inches	39.37
Millimeters of mercury	Atmospheres	0.001316
Square centimeters	Square feet	0.001076
	Square inches	0.155
Torr	mm of Hg	1.0
	Atmospheres	0.001316
Watts/cm-°C	Btu/hr-ft-°F	57.8
Watt-seconds	Btu	0.000948
Watts/cm ²	Btu/hr-ft ²	3170
Watts/cm ² -°C	Btu/hr-ft ² -°F	1760
Centimeters/sec	Feet/sec	0.03281
Meters/sec	Feet/sec	3.281

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1. INTRODUCTION AND SUMMARY

Introduction

This report is the unclassified portion of the fifty-fourth in a series of monthly reports of the work in process on materials research and development for the Atomic Energy Commission under Contract AT(40-1)-2847.

Included is a summary of the work from September 15, 1965 to November 15, 1965, on seven of the specific research and development efforts in process. Three of the remaining programs are reported in the classified portion of this report, GEMP-54B.

Seven other research and development efforts covered by the contract are reported in alternate months.

Summary

EFFECT OF RADIATION ON REFRACTORY METALS AND ALLOYS (57004)

Creep-rupture testing of Mo-TZM (Mo - 0.5Ti - 0.08Zr) alloy specimens was conducted at 1310°C. Results indicate only minor changes in these properties at this temperature for specimens irradiated to a fast neutron fluence of 6.8×10^{19} nvt, ($E_n \geq 1$ Mev).

Hot-hardness measurements of tungsten single crystals irradiated to three fast neutron fluence levels (4.6×10^{18} , 2.4×10^{19} , and 7.3×10^{19} nvt, $E_n \geq 1$ Mev) have been completed. Room-temperature hardness values following elevated temperature anneals of the specimen irradiated to 7.3×10^{19} nvt ($E_n \geq 1$ Mev) reached a maximum at a temperature of 800°C, an increase of approximately 31 percent compared to the as-irradiated specimen.

A comparison of the microhardness increment fraction versus homologous test temperature curves for irradiated molybdenum and tungsten show a much different defect recovery characteristic for tungsten in the region of 0.12 to 0.35 T_m . This is partially attributed to the formation of irradiation-induced defect - foreign atom complexes with rhenium and osmium atoms.

Resistivity measurements on isothermally annealed irradiated molybdenum specimens were conducted in the temperature region of 0.31 T_m . Preliminary analysis of the data shows that probably more than one recovery process is occurring in this temperature range with activation energies in the range of 2.0 to 2.5 ev. Further analysis and possible additional tests are required to determine the nature of the recovery kinetics involved and the defect migration activation energies.

Based on computer simulations, the collided atom volume produced by a cascade is multiply-connected but the region of anelastic strain produced by the associated displace-

ment spike can be taken to be simply connected and its volume is about 3.5 times the collided atom volume at 0°K in α -iron. The irradiation-produced increase in electrical resistivity is proportional to the cumulative collided atom volume. The electrical resistivity, therefore, saturates when the cumulative collided atom volume fills the specimens. Irradiation hardening, however, is greatest for a critical separation distance between dislocation barriers. If displacement spike anelastic strain regions are assumed to act as barriers to dislocation movement then irradiation hardening of a pure metal will be greatest when the displacement spike density is such that this critical separation distance is realized. This circumstance, the fact that the anelastic strain region volume exceeds the collided atom column, and the at-least three-fold-cascade-overlap condition, for electrical resistivity saturation, combine to give a smaller exposure for maximum hardening than for saturation of electrical resistivity in pure α -iron at 0°K. The exposure for electrical resistivity saturation is predicted to be at least 10^2 times larger than that for maximum neutron-irradiation hardening in pure α -iron at 0°K.

EVALUATION OF THE PLASTIC FATIGUE PROPERTIES OF HEAT RESISTANT ALLOYS (57016)

Testing was conducted to demonstrate the capabilities of the new fatigue testing machine. The tests verified the ability to control the machine with the aid of an extensometer that monitors the diameter variation in a push-pull type specimen.

EFFECT OF RADIATION ON HEAT RESISTANT METALS AND ALLOYS (57018)

The reduction in the creep-rupture life of Hastelloy N specimens tested at 650°C appears to be independent of the irradiation temperature; i. e., reactor ambient temperature or at 650°C.

Specimens of A-286 containing the least amount of boron have the greatest reduction in creep-rupture properties for both reactor ambient temperature irradiations and an elevated-temperature (650°C) irradiation.

Specimens of Fe - 15Cr - 4Al - 1Y (1541 alloy) irradiated at reactor ambient temperature showed only minor changes in post-irradiation tensile properties at 650°, 750°, and 850°C when compared to control specimens tested at the same temperature. In particular, the ductility was not affected when tested at elevated temperatures for specimens irradiated to a fast neutron dose of 7×10^{19} nvt (fission).

RADIATION EFFECTS IN BeO (57063)

The relative resistance to microcracking was determined for several BeO compositions in measurements of the dimension and strength changes following irradiation at 100°C. A fine-grained material (8 microns) containing 1 weight percent bentonite in UOX-grade BeO appeared somewhat more stable than materials of similar grain size examined previously while a composition containing 1 weight percent Al_2O_3 in UOX-grade BeO appeared particularly susceptible to microcracking. Other compositions examined, including specimens from five production blocks of the EBOR material, exhibited microcracks at dosages of 1 to 2×10^{20} nvt ($E_n \geq 1$ Mev), essentially in accord with previous observations.

The thermal diffusivity of an extensively microcracked specimen irradiated at 100°C was only about 10 percent of that of the unirradiated material at 300°C and varied only slowly with temperature. On annealing at 1000°C, the diffusivity recovered to about 90 percent of the unirradiated value; the lack of full recovery is attributed to uncured

microcracks remaining after the annealing treatment. In contrast, the diffusivity of a specimen irradiated at 1000°C to 2.5×10^{21} nvt ($E_n \geq 1$ Mev), decreased by amounts ranging from 20 percent at 300°C to about 4 percent at 1000°C.

FISSION PRODUCT TRANSPORT PROCESSES IN REFRACTORY-METAL FUEL SYSTEMS (57070)

Studies of krypton transport in tantalum at elevated temperatures were continued. Optimum discharge conditions for simultaneously loading and diffusing measurable levels of Kr^{85} into fine-grained arc-cast tantalum at 1830°C were established. Submicron sectioning work was continued on the W-Mo-Re alloys.

Anodization and mechanical stripping of the resulting oxide layer were established as the sampling procedure for obtaining fission product distribution data at successive depths in a tantalum can used to contain uranium fuel during irradiation. Surfaces may be prepared for anodization by grinding and polishing at approximately 100-micron-depth intervals.

Final design of the high-temperature furnace and creep stand for determining the effect of fission products on mechanical properties of refractory metals was completed and the tungsten samples for the first irradiation were fabricated.

PHYSICO-CHEMICAL STUDIES OF CLAD UO_2 IN POTENTIAL MELTDOWN ENVIRONMENTS (57075)

Corrosion tests of Zircaloy in steam at 1200°C to 1600°C show that tin or a tin-rich alloy is precipitated in the ZrO_{2-x} layer of the oxide film. In samples tested under isothermal conditions, this layer of tin is a well defined line near the metal-oxide film interface. In samples tested under transient thermal conditions the tin-rich alloy appeared as a network structure in the oxide film.

Type 304 stainless steel shows considerable "bloating" as the metal melts in steam after heating at a 0.3°C temperature rise per second. This effect is not seen where the heating rate is 3°C per second.

PHYSICO-CHEMICAL STUDIES OF Fe-Cr-Al CLAD FUEL SYSTEMS (57076)

Uranium encapsulated in Fe - 15Cr - 4Al was tested for various times at temperatures in the range 600°C to 1200°C. Interaction of the two materials varied from a trace of reaction at the interface after 750 hours at 600°C to extensive compound formation and penetration of a 0.48-cm cladding in 80 hours at 1200°C.

Additional alloys were prepared for a study of the diffusion behavior of Al in Fe-Cr-Al alloys.

Assembly of an apparatus for measuring the activity of alloy components by determining the emf of cells with molten salt electrolytes was begun.

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2. EFFECT OF RADIATION ON REFRACTORY METALS AND ALLOYS

(57004)

The objective of this program is to determine the effect of radiation on the time-, temperature-, and stress-dependent properties of selected refractory metals and alloys, to identify the causes of any observed changes in these properties, and to develop remedial measures.

EXPERIMENTAL PROGRAM

The experimental program to study the effects of neutron irradiation on the creep-rupture, tensile, resistivity, and hardness properties of refractory metals and alloys is continuing. In addition to studies on the above properties, theoretical calculations on the nature of the neutron-induced defects are also being performed as well as direct observations of defects by the use of the transmission electron microscope.

Studies of the creep-rupture properties of irradiated Mo-TZM (Mo - 0.5Ti - 0.08Zr) specimens at 1310°C, hot hardness of tungsten single crystals, resistivity of molybdenum specimens, and theoretical studies on neutron-induced defects in bcc metals are discussed. A summary of the experimental program, test conditions, and preliminary results are given in Table 2.1.

Status of Irradiations

The last capsule (ORM-37) of the ORR dose-dependency series of capsules is completing its 450-hour scheduled irradiation. Capsule ORM-38 will be removed from the reactor during the next reporting period. Capsule ORM-35 has been returned to GE-NMPO and disassembled.

An ambient temperature capsule (ORM-43) is being fabricated for insertion in the ORR-A2 facility following the ORM-38 irradiation. This capsule will contain sheet creep-rupture specimens fabricated from a W-Re-Mo alloy.

The ETR ambient irradiation series of capsules (GEFP2-232 to -241 inclusive)¹ are progressing on schedule. Capsules GEFP2-232 and -233 were returned to GE-NMPO following irradiations of 3920 and 2371 MwD, respectively. Capsules GEFP2-236 and -237 were irradiated in the ETR-E5 facility for 498 hours (3566 MwD) and are awaiting return to GE-NMPO. The GEFP2-234 capsule is presently in the second of its scheduled four ETR reactor cycles in the F-13 SW facility.

Capsules GEFP2-238 and -239 were inserted in the ETR-E5 facility late in the present reporting period. The design of the GEFP2-235 and -240 cartridges is complete and fabrication is in progress.

Two elevated temperature ETR irradiation cartridges were designed for tungsten creep-rupture specimens. A cartridge designated GEFP2-133 will contain nine speci-

¹"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, Table 2.2, p. 14.

TABLE 2.1
SUMMARY OF MAJOR EXPERIMENTS IN THE REFRACTORY METALS AND ALLOYS RADIATION EFFECTS PROGRAM

Exp. No.	Objective	Property	Test Specimen			Irradiation Conditions ^c			Post-Irradiation Heat Treatment		Test Conditions	Percent Change In Property ^d			Comments					
			Material ^a	Type	Lot Number ^b	Capsule Number	Temp, °C	nvt, neutrons/cm ² Thermal (E _n ≥ 1 Mev)	Temp, °C	Time, hr		Creep rate	Rupture life	Elongation						
1	To investigate the effects of neutron irradiation on the creep-rupture and tensile properties. To perform supporting studies on the effects of neutron irradiation on the hardness and resistivity properties.	Creep rupture	W	Rod	MKI37(PM)-X	ORM-4	70	2.2 x 10 ¹⁸	5.8 x 10 ¹⁷			1100	19.34	- 10.0	15	- 1	Warm-worked. To be tested. To be tested. To be tested.			
					MKI37(PM)-X	ORM-8	70	9.5 x 10 ¹⁸	2.5 x 10 ¹⁹			1100	19.34	- 40.0	74	- 1				
					MKI37(PM)-Y	ORM-6	70	2.0 x 10 ¹⁸	3.7 x 10 ¹⁸			1100	20.35	-	17	-				
					MKI37(PM)-Y	ORM-8	70	9.5 x 10 ¹⁸	2.5 x 10 ¹⁹			1100	20.35	- 60	160	- 7				
					MKI37(PM)-E	ORM-17	70	7.5 x 10 ¹⁸	1.6 x 10 ¹⁹			1100	18.28	- 16	28	-10				
					MKI37(PM)-E	ORM-19	70	3.9 x 10 ²⁰	8.2 x 10 ¹⁹			1100	18.28	- 76	490	- 6				
					MKI37(PM)-Y	ORM-6	70	2.0 x 10 ¹⁸	3.7 x 10 ¹⁸			1400	13.01	-	1	10				
					MKI37(PM)-Y	ORM-8	70	9.5 x 10 ¹⁸	2.5 x 10 ¹⁹			1400	13.01	-	9	14				
					MKI37(PM)-E	ORM-19	70	3.9 x 10 ²⁰	8.2 x 10 ¹⁹			1400	10.55	- 6	38	19				
					MKI37(PM)-J	ORM-19	70	3.9 x 10 ²⁰	8.2 x 10 ¹⁹			1500	8.44	- 8	10	-29				
					MKI37(PM)-J	ORM-19	70	3.9 x 10 ²⁰	8.2 x 10 ¹⁹			1700	5.62	- 13	9	-28				
					MKI37(PM)-J	33MT-137	1300	2.2 x 10 ²⁰	1.8 x 10 ²⁰			900								
					MKI37(PM)-E	ORM-17	70	7.5 x 10 ¹⁸	1.6 x 10 ¹⁹			900	22.64	- 76	280	-26				
					MKI37(PM)-J	ORM-12	70	9.1 x 10 ¹⁸	3.2 x 10 ¹⁸			1100	37.28	- 30	60	8				
					W - 25Re	Sheet	RWS9(PM)-4	ORM-20	70	3.5 x 10 ²⁰	8.0 x 10 ¹⁹			1100	43.9	- 52		55	-56	
			Mo	Sheet			KDM-515A(AC)-8	ORM-23	70	5.2 x 10 ¹⁹	1.4 x 10 ¹⁹			580	21.09	29		- 96	- 4	
							KDM-515A(AC)-2	ORM-13	70	5.5 x 10 ¹⁹	6.9 x 10 ¹⁸			980	10.55	- 2		10	14	
							KDM-515A(AC)-1	ORM-16	70	1.4 x 10 ²⁰	3.7 x 10 ¹⁹			580	21.09	210		- 65	-	
			50Mo-50Re	Sheet			KDM-515A(AC)-1	ORM-16	70	1.4 x 10 ²⁰	3.7 x 10 ¹⁹			600	21.09	57		- 32	-	
								ORM-20	70	3.5 x 10 ²⁰	8.0 x 10 ¹⁹									
							Ta	Sheet	ORM-23	70	5.2 x 10 ¹⁹	1.4 x 10 ¹⁹								
			Ta - 10W	Sheet				ORM-23	70	5.2 x 10 ¹⁹	1.4 x 10 ¹⁹									
			Tensile	W	Rod	MKI42(PM)-G	33MT-138	70	3.6 x 10 ²⁰	1.3 x 10 ²⁰			371		Yield Strength (Brittle fracture)	Tensile Strength		Elongation	Testing continuing at higher temperatures. To be tested. To be tested.	
		MKI42(PM)-B				ORM-9	70	2.8 x 10 ¹⁹	5.1 x 10 ¹⁸											
		MKI42(PM)-C				ORM-10	70	9.3 x 10 ¹⁹	1.7 x 10 ¹⁹											
		Resistivity	W	Rod	MKI42(PM)-E	ORM-7	70	2.5 x 10 ¹⁹	5.3 x 10 ¹⁸			-198		Resistivity			Warm-worked.			
					MKI42(PM)-D	ORM-11	70	1.2 x 10 ²⁰	3.3 x 10 ¹⁹			-198		105						
					MKI42(PM)-F	33MT-113	1300	2.2 x 10 ²⁰	8.0 x 10 ¹⁹			-198		315						
					W - 25Re	Rod	RWWS(PM)-A	ORM-14	70	6.2 x 10 ¹⁹	1.2 x 10 ¹⁹			-198		39				
							RWWS(PM)-C	33MT-113	1300	2.2 x 10 ²⁰	8.0 x 10 ¹⁹			-198		10				
			Mo	Rod	GEH(PM)-B	ORM-14	70	6.2 x 10 ¹⁹	1.2 x 10 ¹⁹			-198		15						
					GEH(PM)-C	ORM-14	70	6.2 x 10 ¹⁹	1.2 x 10 ¹⁹			-198		80						
					GEH(PM)-L	33MT-113	700	2.2 x 10 ²⁰	8.0 x 10 ¹⁹			-198		118						
			Mo-TZM	Rod	ZM8003(AC)-5	ORM-18	70	1.1 x 10 ²⁰	3.7 x 10 ¹⁹			-198		2						
					ZM8003(AC)-5	33MT-113	700	2.2 x 10 ²⁰	8.0 x 10 ¹⁹			-198		74						
		Hardness	W	Sheet	411-1	ORM-15	70	1.3 x 10 ²⁰	3.5 x 10 ¹⁹			15	500 grams	DPH Hardness			Single crystal. From isochronal annealing studies. Polycrystal. From isochronal annealing studies.			
											650		8							
											1300		60							
			Mo	Sheet	KDM-515A(AC)-M(10)	ORM-30	70	9.2 x 10 ¹⁹	2.6 x 10 ¹⁹			15	500 grams	5						
											300		18							
											1200		55							
													0							

SUMMARY OF MAJOR EXPERIMENTS IN THE REFRACTORY METALS AND ALLOYS RADIATION EFFECTS PROGRAM

[illegible]

^a All materials are in the recrystallized condition unless otherwise noted.

^b Material lot numbers are followed by parenthetical expressions denoting powder metallurgy (PM) or arc-cast (AC) processing. The last letter or number identifies the sheet or rod stock fabricated from the specified lot. Seven or more test specimens are generally machined from each sheet or rod stock.

^cThe listed irradiation conditions represent approximate values for specimens which were irradiated and represent target values for specimens to be irradiated. A summary of capsule irradiations, designations, and the number and type of specimens is given in Table 2.2.

^dProperty changes represent increases due to the indicated irradiations and test conditions when compared to a corresponding unirradiated test specimen. Negative values represent a decrease in the specified property.

mens for irradiation at 1300°C in a high-purity inert gas atmosphere. A similar cartridge (GEFP2-153) will be concurrently irradiated at a temperature of 1000°C. Fabrication was begun on both cartridges.

Hot-Hardness Tests

The hot-hardness testing of a series of tungsten single crystals is nearing completion. Annealing measurements have been made on three irradiated specimens as well as the control specimen. Data on the three irradiated specimens were reported previously,² and the data on the control are given in Figure 2.1. The relative number of rhenium and osmium atoms which are formed as a result of thermal neutron (n, γ) reactions with tungsten³ will be different in the three irradiated specimens due to (1) the different thermal neutron fluence⁴ levels and (2) the fact that the specimens were not tested at equivalent decay times.⁵ Therefore, any neutron-induced point defect – foreign atom complexes which may affect dislocation motion could completely mask out the influence of the foreign

Fig. 2.1 – Hardness of unirradiated tungsten single crystal {411} as a function of temperature

²Ibid., p. 19.

³"High-Temperature Materials Program Progress Report No. 50, Part A," GE-NMPO, GEMP-50A, August 20, 1965, pp. 17–21.

⁴Time integral of the neutron flux density generally known as the dose.

⁵ W^{185} , $\tau_{1/2} = 74$ days.

W^{187} , $\tau_{1/2} = 65$ days

atoms (Re and Os) which may also tend to strengthen the material at elevated temperatures. Therefore, the three irradiated and the one control specimen were given a further anneal (1 hour at 1700°C in a hydrogen atmosphere) where it is believed that these defect – foreign atom complexes dissociate; subsequent hardness measurements will be made at room temperature, 900°C, and 1200°C to determine the effect on hardness of only the foreign atoms (Re and Os).

The microhardness increment fraction, $\Delta H_f = \frac{H_I - \bar{H}_c}{\bar{H}_c}$, is given as a function of temperature for the three irradiated specimens in Figure 2.2. In this particular representation, H_I is the hardness value of the irradiated specimen at a given temperature and $\bar{H}_c$ is the hardness of the control at the same temperature. The microhardness increment fraction at approximately 320°C (0.16 T_m) is plotted as a function of nvt ($E_n \geq 1$ Mev) in Figure 2.3, and the slope of this line indicates that ΔH_f is proportional to the square root of the nvt ($E_n \geq 1$ Mev) for the range indicated, viz., approximately 4.6×10^{18} to 7.3×10^{19} nvt. In Figure 2.4, the microhardness increment fraction curves as a function of homologous temperature, T/T_m , are compared for molybdenum and tungsten. The higher value of ΔH_f in the case of tungsten in the region between 0.12 and 0.35 is probably at least partially attributable to the formation of the neutron-induced point defect – foreign atom complexes discussed above. In the case of molybdenum, transmutations to other elements is not very significant (low capture cross section) and those atoms that do absorb neutrons are mostly transformed to another stable isotope of molybdenum. Therefore, the foreign atom concentration will be much lower in molybdenum than in tungsten for equivalent thermal neutron fluence levels.

Fig. 2.2 – Microhardness increment fraction, ΔH_f , of irradiated tungsten single crystals {411} as a function of temperature

Fig. 2.3 – Microhardness increment fraction, ΔH_f , at approximately 320°C ($\sim 0.16 T_m$) as a function of fast neutron dosage

Fig. 2.4 – Microhardness increment fraction, ΔH_f , of molybdenum and tungsten as a function of homologous temperature

Annealing studies on tungsten single crystal specimens were performed in the same manner as previously reported for molybdenum.⁶ Results of room temperature annealing studies on tungsten are given in Figure 2.5. The specimen irradiated to $7.3 \times 10^{19} nvt$ ($E_n \geq 1$ Mev) showed a very large increase (31%) in hardness after elevated temperature anneals up to about 800°C; at higher temperature annealing caused the hardness values to decrease rapidly to the corresponding value of the control specimen. An annealing study was also performed on the tungsten single crystal irradiated to $7.3 \times 10^{19} nvt$ ($E_n \geq 1$ Mev); the specimen was annealed at various temperatures above 900°C, with hot-hardness being obtained at 900°C. The results are given in Figure 2.6. This particular study was performed to compare the hardness properties with changes in resistivity

⁶"High-Temperature Materials Program Progress Report No. 47, Part A," GE-NMPO, GEMP-47A, May 28, 1965, pp. 94-95.

Fig. 2.5—Room-temperature hardness of irradiated and unirradiated single crystal tungsten {411} as a function of annealing temperature

and creep-rupture properties.⁷ The slight increase in the 900°C hardness value following the 1100°C anneal is not yet understood but will be considered again following the completion of the 1700°C anneal studies.

Creep-Rupture Tests

Creep-rupture tests conducted on a series of arc-melted Mo-TZM (Mo – 0.5Ti – 0.08 Zr) sheet specimens at 1310°C are summarized in Table 2.2. A stress versus creep-rate plot for these data is shown in Figure 2.7. The results, as indicated by control and irradiated lines, show a decrease of approximately 21 percent in creep-rate due to irradiation. This decrease is considered insignificant based on results on other refractory

⁷"Fourth Annual Report – High-Temperature Materials and Reactor Components Development Programs, Volume I – Materials," GE-NMPO, GEMP-334A, February 26, 1965, p. 65.

Fig. 2.6 - Hot hardness (900°C) of tungsten single crystals that had been irradiated to 7.3×10^{19} nvt ($E_n \geq 1$ Mev) versus annealing temperature

Fig. 2.7 - Linear creep rate of Mo-TZM tested at 1310°C in hydrogen

TABLE 2. 2
CREEP-RUPTURE TESTS ON Mo-TZM SPECIMENS^a AT 1310°C

Specimen No.	Sheet No.	Condition	Stress, kg/mm ²	Linear Creep Rate, sec ⁻¹	Rupture Life, hr	Elongation, % in 2.54 cm Gage Length
667	4	ORM-21 ^b	11.25	4.57×10^{-6}	18.61	49.8
677	4	Control	10.55	3.89×10^{-6}	16.64	41.4
668	4	ORM-21 ^b	10.55	3.02×10^{-6}	17.95	45.9
675	4	Control	9.93	2.96×10^{-6}	13.6 < t_r < 17.5	48.7
669	4	ORM-21 ^b	9.93	2.36×10^{-6}		39.2
678	7	Control ^c	9.93	2.28×10^{-6}	17.97	39.5
684	7	Control	9.14	1.69×10^{-6}	22.95	47.8

^aSpecimens from 0.5-cm-thick sheet, heat KDTZM 974A; annealed after fabrication of specimens at 1200°C for 1 hour in H₂. Tested in hydrogen atmosphere.

^bORM-21 – irradiated to 6.8×10^{19} nvt ($E_n \geq 1$ Mev) at reactor ambient temperature.

^cSpecimen in the vendor-annealed condition only.

materials irradiated to comparable doses. The increase in the rupture life due to irradiation for the two comparable sets of data are also considered minor. It should also be noted that there was no apparent degradation in the ductility. Hence, it is believed that any irradiation-induced hardening generated at this fluence level (6.8×10^{19} nvt, $E_n \geq 1$ Mev) was essentially removed at the test temperature (1310°C). Subsequent testing of this alloy will be performed at a lower temperature.

Resistivity

The first phase for determining the isothermal recovery of defects present in irradiated molybdenum near the 0.31 T_m temperature region (Stage IV) was completed. Four samples of recrystallized molybdenum (1 hour at 1200°C in a hydrogen atmosphere) irradiated at reactor ambient temperature to a dose of approximately 1.3×10^{19} ($E_n \geq 1$ Mev), were annealed for 2 hours at 456°C. The purpose of this anneal was to normalize all of the specimens to a given nearly uniform defect concentration and to remove the defects responsible for the recovery peaks observed to occur below 0.31 T_m . With this procedure, only the defects mobile in the temperature region of 0.31 T_m would be observed and data evaluation would be simpler and more reliable.

The technique for performing the isothermal anneals was essentially the same as that reported for tungsten;⁸ i. e., a new sample was introduced at each new temperature being studied. The preliminary data for the four temperatures being studied are shown in Figure 2.8. Note that, except for the 556°C temperature condition, the annealing times were carried out until the resistivity values for the irradiated specimen approached asymptotically with the unirradiated control. The convergence of the irradiated and control resistivity data is quite different from that observed for tungsten⁸ (irradiated above 10^{19} nvt, $E_n \geq 1$ Mev) in that annealing of tungsten did not restore the resistivity values to those of the pre-irradiation conditions. This can be explained by the W-Re-Os transmutation as discussed previously in the section on hot-hardness.

Preliminary analysis of the data shows that apparently more than one process is occurring in this temperature range. The first portion of each isotherm appears to follow second-order kinetics,⁹ whereas, the second portion of the isotherm appears to follow first-order kinetics. Assuming the processes to be totally independent, analysis indicates that they have similar activation energies. The preliminary results on the uncorrected

⁸"High-Temperature Materials Program Progress Report No. 47, Part A," GE-NMPO, GEMP-47A, May 28, 1965, pp. 90–92.

⁹J. Motteff and J. P. Smith, "Recovery of Defects in Neutron-Irradiated Tungsten," *Flow and Fracture of Metals and Alloys in Nuclear Environments*, ASTM No. 380, published by American Society for Testing and Materials, 1965.

Fig. 2.8—Isothermal resistivity recovery of recrystallized molybdenum irradiated at reactor ambient temperature to 1.3×10^{19} nvt ($E_n \geq 1$ Mev)

data (without compensation for transient specimen temperature effects during the short-time annealing) indicate an activation energy, ΔH , in the range 2 to 2.5 eV for both processes. Further analyses and more measurements are being performed (1) to determine if the two processes are independent or if the second is dependent on the first and (2) to further assist in interpreting the mechanism of recovery in this important temperature region.

FUNDAMENTAL STUDIES

A study of neutron irradiation effects saturation in α -iron was made by simulating the production of point defects by several overlapping collision cascades on a digital computer. A basis for saturation behavior was introduced by using the Erginsoy-Vineyard re-

combination region for α -iron to represent vacancy-interstitial pair interactions.¹⁰ The simulations pertain to irradiation and observation at 0°K. They are, therefore, directly pertinent to low-temperature irradiation damage research work on bcc metals currently in progress. In addition, as previously reported,¹¹ it is possible to generalize the results to any irradiation temperature below 250°C, in the case of α -iron. The goal of the present study was to explore the use of the recombination volume approximation to vacancy-interstitial interactions, in describing the approach to saturation of the electrical resistivity and the approach to a maximum value for the lower yield point.

By definition, the effect of irradiation on a particular physical property of a specimen exposed to a constant irradiation flux is saturated when changes in this property are no longer observed. There is a widespread inclination to regard saturation of the displacement density as defining a universal saturated damage state. This inclination has its roots in the fact that electrical resistivity change measurement has been an overwhelmingly popular technique for monitoring irradiation damage. However, as shown by Quere et al.,¹² the neutron irradiation exposures required to saturate the effect of irradiation on electrical and mechanical properties are not generally the same. This means that it is necessary to consider more than the conditions for a vanishing time derivative of the irradiation-produced defect density in characterizing the saturated damage state for a particular physical property. A damage characteristic which suggests itself in this regard is the deployment of defects in a displacement spike. Since the computer simulation method (computer experiment method) enables one to describe the internal "defect" structure of a displacement spike, this method was used in an attempt to describe the possible influence of displacement spike structure on the saturation of irradiation effects, especially those on mechanical properties.

Two important levels of interaction between a distribution of existing defects and the formation of new defects naturally distinguish themselves in α -iron:

1. A weak interaction occurs between the defects produced by a current branch of an isolated cascade and the distribution of defects produced by the preceding branches. In this instance, less than 10 percent overlap usually occurs. This degree of interaction appears to be independent of the initiating primary knock-on atom energy for elastic collision cascades and gives rise to a characteristic defect cluster size distribution.
2. A strong interaction occurs between the defect distribution produced by an entire cascade and the formation of new defects by a second cascade of comparable energy when the second cascade is initiated within the displacement spike produced by the first cascade. In this instance, over 30 percent overlap usually occurs and the original defect deployment is completely altered. The defect cluster size distribution associated with this strong interaction appears to be different from that associated with the weak interaction.

Computational Model and Procedure

As many as three collision cascades were simulated in succession so that the course of each successive cascade was influenced by the cumulative defect distribution produced by its predecessors. Cascades initiated by primary knock-on atoms (PKA) with energies of 1 to 5 kev were considered. It was assumed that a collision cascade could be represented as a sequence of binary atomic collision events. A detailed account of the basic

¹⁰C. Erginsoy, G. H. Vineyard, and A. Englert, "Dynamics of Radiation in a Body-Centered Cubic Lattice," *The Physical Review*, Vol. 133, 1964, p. A595.

¹¹"High-Temperature Materials Program Progress Report No. 50, Part A," GE-NMPO, GEMP-50A, August 20, 1965, pp. 13-32.

¹²Y. Quere, private communication.

computational scheme has been reported previously.^{13,14} The model used in the present study, however, differed from that described earlier in two particulars:

1. In the present study, each binary collision event was determined exactly in accordance with the Erginsoy-Vineyard¹⁰ potential using the integration scheme of Robinson and Oen.¹⁵ In the earlier work, the collision angle was determined from the hard-sphere model approximation on the basis of an energy dependent collision radius given by the Bohr potential.
2. Only vacancies and interstitials which satisfied the Erginsoy-Vineyard stability conditions for vacancy-interstitial pairs were allowed in the present study. A recombination volume one-half as large as the Erginsoy-Vineyard recombination volume was used in the earlier work.

Erginsoy, et al., found that a split interstitial placed at any one of the non-central atom positions in Figure 2.9 would immediately recombine with a vacancy at the central atom position at 0°K; i. e., without requiring thermal excitation. This recombination region has a volume $V_R = 31 \Omega$ if one includes the central site (Ω is the atomic volume). According to Schilling,¹⁶ Lehmann has shown that the saturated displacement density is $d_{\max} = V_R^{-1}$, expressed as an atom fraction. Schilling has demonstrated that in addition to this

Fig. 2.9 – Erginsoy-Vineyard recombination volume for a vacancy and an interstitial in α -iron at 0°K. A split interstitial centered at any of the non-central positions will recombine with a vacancy at the center position. The region contains 31 atom sites.

¹³J. R. Beeler, Jr., and D. G. Besco, "Range and Damage Effects of Tunnel Trajectories in a Wurtzite Structure," *Journal of Applied Physics*, Vol. 34, 1963, p. 2873; "Effects of Channeling on Damage Production in Iron," *The Physical Review*, Vol. 134, 1964, p. A530.

¹⁴J. R. Beeler, Jr., "Primary Damage State in Neutron-Irradiated Iron," *Journal of Applied Physics*, Vol. 35, 1964, p. 2226.

¹⁵M. T. Robinson and O. S. Oen, "Computer Studies of the Slowing Down of Energetic Atoms in Crystals," *The Physical Review*, Vol. 132, 1963, p. 2385.

¹⁶W. F. Schilling, private communication.

direct recombination mechanism there exists an indirect mechanism whereby interstitials initially outside V_R are moved within recombination range with a vacancy as a result of radiation-induced lattice excitations. This indirect process was not considered in the present study. It serves to establish an effective recombination region volume which is larger than the Erginsoy-Vineyard recombination region volume.

Cascade overlap was simulated by first running an isolated cascade and then starting other cascades in the region traversed by the first cascade. This procedure is illustrated by Figure 2.10 wherein a set of primary and secondary knock-on atom trajectories is shown for each of two different cascades. The trajectories of third and higher order knock-ons were omitted to avoid confusion. The lower set in the figure belongs to the first cascade. The upper set belongs to the cascade which resulted when a second PKA, with the same initial energy and direction as the first PKA, was shot into the region previously damaged by the first cascade. The second PKA was started from a lattice site positioned three lattice constants along the $[010]$ direction from the starting position of the first PKA. In addition to the overlap of cascades initiated by PKA with a common initial direction and energy, arbitrary combinations of initial energies and directions were also used. The degree of overlap among the branches of single isolated cascades was also studied.

Fig. 2.10 – Interaction between two cascades. Projections of the primary (heavy solid line) and secondary (dotted and dashed lines) knock-on atom trajectories in two interacting cascades are shown.

Approach to a Saturated Displacement Density

There are two extreme models for the approach to a saturated displacement density, as indicated schematically by Figure 2.11. In each instance, it is assumed that a maximum displacement density exists, as defined above. For simplicity, a single symbol is used to represent a displacement event, rather than representing vacancies and interstitials separately.

Fig. 2.11 – Two extreme models for an increasing average macroscopic displacement density. (a) Random production of individual displacements. (b) Random production of groups of displacements in each of which the displacement density is saturated. New defects are denoted by open circles, old defects by filled circles. The number at the top of each frame is the number of stable displacements it contains.

Random Production of Individual Displacement Events - This case is most closely realized in electron bombardment of thin metallic specimens. It is assumed that displacement events are added at random at a constant rate. Initially, the displacement distribution is uniform and the total number of displacements builds up at a constant rate. In Figure 2.11a, displacements are added at the rate of 10 displacements per unit time (picture frame). Eventually, newly created displacements overlap old ones in a manner which causes mutual annihilation; the buildup rate for the number of displacements then starts to decrease monotonically, as indicated in the last two frames of Figure 2.11a. Sizmann¹⁷ has analyzed the approach to displacement density saturation for this case.

Random Production of Groups of Displacements - In neutron irradiation, PKA are dislodged by neutrons and each of these PKA initiates an atomic collision cascade. The resulting assembly of vacancies and interstitials is called a displacement spike. Except for the initiating PKA itself, all displacements in a spike are produced by the collision cascade. Previous calculations predict that the displacement density in a spike¹⁸ is saturated; i. e., it has the value $d_{\max} = V_R^{-1}$ for all PKA energies. This suggests the second extreme model in which groups of displacements are added at a constant rate, with the density in each group being $d_{\max}$. It is assumed that each group contains the same number of displacements but the shape of the group may vary. In Figure 2.11b, each group (displacement spike) contains 10 displacements and one spike is added per unit time. Eventually, a new spike overlaps an old one and defects are annihilated, as in the third frame of Figure 2.11b.

In general, the defect deployment produced by neutron irradiation is a mixture of the two extreme deployments described above. First of all, the displacement spikes do not

¹⁷G. Luck and R. Sizmann, "The Radiation Annealing of Frenkel Defects," *Phys. Stat. Sol.*, Vol. 5, 1964, p. 683.

¹⁸J. R. Beeler, Jr., "Saturated Damage State in Alpha-Iron," *Bull. Am. Phys. Soc.*, Vol. 10, 1965, p. 711.

have a common size.¹⁴ More important, however, the internal structure of a displacement spike is determined by the recombination region volume and the cohesive energy of the irradiated specimen material. It tends to degenerate from that of a compact group of displacements to that of a random assemblage as V_R and/or the cohesive energy increase. Increasing the irradiation temperature has the same degenerative effect.

Isolated Collision Cascade and Displacement Spike Characteristics

Two types of disturbed crystal regions, pertinent to an approach to saturation, are associated with a collision cascade. One of these, the collided atom volume, includes the initial positions of all atoms participating in collisions during a collision cascade. This volume of disturbed crystal was found to be that pertinent to defining the average macroscopic displacement density and hence to the saturation of electrical resistivity. The other region of interest is the region of anelastic strain set up by the vacancies and interstitials in a displacement spike. This type of crystal disturbance impedes the motion of dislocations and is, therefore, pertinent to the saturation of the lower yield point. The anelastic strain volume is larger than the collided atom volume for an irradiation at 0°K, but should be smaller than the collided atom volume for irradiation at a temperature above 120°K, the temperature at which interstitials begin to migrate in α -iron.¹⁹ A brief discussion of these two volumes for an isolated collision cascade will be given to form a basis for describing cascade overlap effects.

The shape and structure of the collided atom volume associated with a 5-keV cascade appears in Figure 2.12. It is within such a volume that the displacement density is saturated. Hence, it is important to point out that the collided atom volume is multiply-connected (Swiss cheese structure) and very frequently tends to exhibit a $\langle 110 \rangle$ orientation in α -iron.

Fig. 2.12—Collided atom volume for a 5-keV cascade in α -iron. This volume is multiply-connected. Note $\langle 100 \rangle$ orientation.

¹⁹P. G. Lucasson and R. M. Walker, "Production and Recovery of Electron-Induced Radiation Damage in a Number of Metals," *The Physical Review*, Vol. 127, 1962, p. 485.

An approximation to the region of inelastic strain produced by the vacancies and interstitials in a displacement spike is shown in Figure 2.13. The anelastic strain region shown in Figure 2.13 is that of the displacement spike associated with the collision-cascade collided atom volume in Figure 2.12. Because either a vacancy or an interstitial causes anelastic strains at least up to fourth neighbor distances, the anelastic strain region in α -iron at 0°K can be taken to be simply connected.

Fig. 2.13—An elastic strain region produced by the interstitial and vacancies created by the collision cascade whose collided atom volume appears in Figure 2.12

Cascade Overlap

Let $\vartheta(E)$ represent the number of displacements produced by a cascade initiated by a PKA with energy E . The average number of displacements per unit PKA energy is $K(E) = \vartheta(E)/E$ and is called the displacement efficiency of a cascade. The general effect of running a collision cascade through a region containing defects produced by previous cascades was to decrease the displacement efficiency of the cascade relative to that it would exhibit in an initially perfect crystal. In addition, the size distribution of vacancy clusters appeared to be changed relative to that for an isolated cascade. The interstitial cluster distribution, however, did not appear to be changed.

Table 2.3 lists the displacement production results for overlapping 2.5-keV cascades which were initiated by three PKA started from different starting positions a distance $\Delta z = a$ apart along the $[001]$ direction, where a is the lattice constant. The same initial direction was used for all three PKA. In this particular instance, the displacement efficiency fell off by about a factor of six in the course of three passes. On the average, the displacement efficiency of the second cascade was reduced by approximately 30 percent and that of the third cascade by approximately 70 percent in the case of 2.5-keV cascades. A maximum reduction of 90 percent was observed for the third cascade, hence at least a three-fold overlap of 2.5-keV collision cascades would be required to saturate the average macroscopic displacement density. Although the average displacement efficiency reductions for 5-keV cascade overlap were larger than those for 2.5-keV cascades, at least three-fold overlap also would be required, at 5 keV, to saturate the average macroscopic displacement density.

Maps of the vacancy deployment in a (001) slab section, two lattice constants thick, after the first, second, and third passes of the 2.5-keV cascades of Table 2.3 appear in Figure 2.14a, b, and c, respectively. Interstitials were omitted since the cluster size distribution of these defects was not changed significantly by cascade overlap. This se-

Fig. 2.14 – Vacancy map for three successive 2.5-keV cascade passes. Note the growth of a 20-vacancy cluster and the reduction of the tetravacancy in (a) to a divacancy in (c), for example.

TABLE 2.3
DAMAGE PRODUCTIVITY OF OVERLAPPING
2.5-keV CASCADES

Number Of Cascades	Cumulative Vacancies	Displacement Efficiency	Displacement Efficiency Decrease, %
1	30	12.0	0
2	56	10.4	13
3	62	2.4	80

quence of maps illustrates the re-arrangement of defects by cascade overlap. The particular trend shown in this sequence is the formation of larger vacancy clusters. Although it appears that the vacancy density increased with each pass, the vacancy density was in fact constant because of a compensating increase in the cumulative collided atom volume. The vacancy cluster size appeared to be limited. The largest observed vacancy cluster size (20 vacancies) is just that observed in the case of isolated cascades. Cascade overlap did not always lead to the aggregation of small clusters to form larger clusters. Rather, the frequency of instances in which clusters were broken up was comparable to that of instances in which larger clusters were aggregated. At this juncture in our study, it appears that overlap tends to increase the relative numbers of single vacancies and clusters containing 10 or more vacancies, relative to the vacancy cluster size distribution produced by isolated cascades.

The overlap of two 5-keV cascades, as it appeared in a (001) slab section two lattice constants thick, is shown in Figure 2.15. The extent of the first cascade collided atom volume in this section is defined by the solid outline. The dashed outline defines the extent of the second cascade collided atom volume. Their overlap region is cross hatched. One observes that the overlap region is also multiply-connected, which explains why highly localized defect re-arrangements were frequently observed. In this instance, the initial directions of both PKA concerned differed by 30 degrees and neither was parallel to [110], yet both collided atom volumes exhibit a marked [110] orientation.

Defect deployment changes were more extensive after the PKA energy was raised to 5 keV than they were at 2.5 keV. This is illustrated by Figures 2.16 and 2.17 which describe the vacancy deployment after the first pass and the second pass, respectively, in the region described by Figure 2.15. All but one of the single vacancies in Figure 2.16 (first pass) were annihilated during the second pass. In addition, the v_4 , v_6 , and v_7 in Figure 2.16 were altered and/or incorporated into new vacancy configurations. Here, v_n denotes a cluster of n vacancies.

A comparison of sections from the cumulative collided atom volumes and cumulative damage distributions for successive stages of cascade overlap yields the following microscopic picture for an approach to a saturated average macroscopic displacement density; viz., a displacement spike is the most simple independent defect produced by an isolated elastic collision cascade. To first order, a cascade produces a high density of "transient" vacancies and interstitials. These transient defects interact as determined by the shape and size of the recombination volume to give a stable deployment of defects – the displacement spike – with the maximum defect density allowed by V_R . The average macroscopic displacement density is increased by a gradual enlargement of the specimen volume in which collision cascades have been initiated. Because the defect density produced by an isolated cascade is saturated "ab initio," cascade overlap cannot contribute to an increased average macroscopic displacement density. Rather, in the region of cascade overlap, the

Fig. 2.15—Overlap of two 5-keV cascade collided atom volumes in a (001) plane section two lattice constants thick (four successive (002) planes). The solid outline defines the extent of the collided atom volume for the first cascade, the dashed line that for the second decade. The overlap region is cross hatched.

old damage distribution is partially annihilated, partially re-arranged and new defects are added, but the local defect density remains unchanged at the level $d_{\max} = V_R^{-1}$.

All other factors being constant, the displacement efficiency of the second of two overlapping cascades could usually be minimized either by making the energy of the second initiating PKA less than that of the first or by making the initial direction of the second PKA the same as that for the first PKA. The first effect is largely a containment effect; i. e., it is possible to attain nearly complete overlap by starting a small cascade in a damaged region sufficiently large to encompass the small cascade. This effect causes the third cascade to always suffer a much larger decrease in displacement efficiency than the second cascade in an overlap sequence. By starting the second PKA parallel to the first, one makes it possible for the $\langle 110 \rangle$ directed cascade branches to experience maximum interference with their counterparts in, and/or penetrate to the center of, the adjacent cascade region, as illustrated in Figure 2.10. In general, the degree of displacement efficiency reduction depended on the separation distance between PKA starting points, the PKA energies, the PKA initial directions, and whether the PKA starting points lay along $\langle 100 \rangle$, $\langle 110 \rangle$, or $\langle 111 \rangle$ directions.

The volume of an anelastic strain region (displacement spike) was close to being simply connected and was about 3.5 times that of the associated, multiply connected collided atom volume at 0°K. Because at least a three-fold overlap of collided atom volumes is

Fig. 2.16 – Damage map associated with the collided atom volume for the first cascade pass in Figure 2.15

Fig. 2.17 – Damage map associated with cumulative collided atom volume for the first and second cascade passes in Figure 2.15. Note alteration of vacancy clusters v_4 , v_6 , and v_7 , of Figure 2.16, produced by the first cascade.

required to give a saturated displacement density, the exposure required to saturate electrical resistivity should be at least an order of magnitude larger than that required to fill the specimen with displacement spikes at 0°K. However, the maximum increase in the lower yield point occurs for a critical dispersion of dislocation barriers. Cottrell²⁰ gives a critical separation distance of about $50a$ (a is lattice constant) for a maximum resistance to dislocation movement. Typical displacement spike diameters range from $10a$ to $30a$ in α -iron, and are, therefore, less than the critical separation distance. Hence, if spikes act as dislocation barriers^{11,21} a spike density a factor of $(50a/20a)^{12}$ smaller than that required to fill the specimen would include maximum hardening. This circumstance, together with the multiple overlap requirement, suggests that the exposure for maximum hardening should be about two orders of magnitude smaller than that for saturation of the electrical resistivity in pure α -iron.

As yet, there is no experimental data which allow a direct comparison with all the computer simulation results outlined above. However, experimental support for the approach adopted for treating irradiation hardening comes from the work of Harries, et al.^{11,22} These investigators measured the increase in the lower yield point of neutron-irradiated ferritic steel produced by each of five different neutron spectra. These spectra ranged from that for a graphite moderated reactor to one nearly as hard as the fission spectrum. The relative standing of the irradiation-induced increases in the lower yield point, among the five neutron spectra, given by the computer simulation method was the same as that observed.

Conclusions

Computer simulations of successive collision cascade traversal of the same local region in pure α -iron indicate that, with one possible exception, no distinction exists between an "unsaturated" and a "saturated" damage state, inside the cumulative collided atom volume, at 0°K. A collided atom volume is the volume of atoms which participate in collisions in a collision cascade. The cumulative collided atom volume is the collection of collided atom volumes produced by all collision cascades during irradiation. The simulations indicate that the displacement density in a displacement spike is saturated "ab initio" in α -iron. In general, the displacement density in a spike is determined by the volume of the vacancy-interstitial recombination volume and the cohesive energy for the material concerned. Because of the saturated displacement density in a spike, the overlap of collision cascades cannot contribute to an increase in the average macroscopic displacement density. The average macroscopic displacement density is increased only by the intrusion of new collision cascades into previously undamaged regions.

The collided atom volume produced by a cascade is multiply connected but the region of anelastic strain produced by the associated displacement spike can be taken to be simply connected and its volume is about 3.5 times the collided atom volume at 0°K in α -iron. The irradiation-produced increase in electrical resistivity is proportional to the cumulative collided atom volume. The electrical resistivity should, therefore, saturate when the cumulative collided atom volume fills the specimens. Irradiation hardening, however, is greatest for a critical separation distance between dislocation barriers. If displacement spike anelastic strain regions are assumed to act as barriers to disloca-

²⁰A. H. Cottrell, *Dislocations and Plastic Flow in Crystals*, Oxford University Press, London, 1953.

²¹R. L. Fleischer, "Solution Hardening by Tetragonal Distortions: Application to Irradiation Hardening in F.C.C. Crystals," *Acta Metallurgica*, Vol. 10, 1962, p. 835; "Effects of Non-Uniformities on the Hardening of Crystals," *Acta Metallurgica*, Vol. 8, 1960, p. 598; *Strength of Metals*, Reinhold Press, New York, 1963.

²²"High-Temperature Materials Program Progress Report No. 50, Part A," GE-NMPO, GEMP-50A, August 20, 1965, pp. 21-32; Harries, Bagley, Bell, Gibson, Gillis, Pfeil and Wright in *Irradiation Behavior of Steel as a Structural and Cladding Material*, Geneva Conference, 1964.

tion movement, then irradiation hardening of a pure metal will be greatest when the displacement spike density is such that this critical separation distance is realized. This circumstance, the fact that the anelastic strain region volume exceeds the collided atom volume, and the at-least three-fold-cascade-overlap condition, for electrical resistivity saturation, combine to give a smaller exposure for maximum hardening than for saturation of electrical resistivity in pure α -iron at 0°K. The exposure for electrical resistivity saturation is predicted to be at least 10^2 times larger than that for maximum neutron-irradiation hardening in pure α -iron at 0°K. During the approach to the saturation of the average macroscopic displacement density, the defect structure of the damage regions exhibited a preferred orientation along $\langle 110 \rangle$ directions.

EQUIPMENT STATUS

A model TT-D-L Instron Universal Instrument was received and installed. The machine possesses reversible load design permitting loading from tension or compression directly into the opposite mode of stress, reversibly, up to the full capacity of the machine. All controls are located on a console unit for remote operation with radioactive specimens. The machine has an X-Y chart drive system for use with extensometers. Preliminary checkout of the machine and training of three operators was conducted by a factory representative. Calibration of the measuring systems and accessory transducers is in progress.

A complete new muffle furnace tube assembly was designed for the intermediate temperature ($T < 1000^\circ\text{C}$) creep-rupture stand. This assembly will provide for lower pressure ($\sim 5 \times 10^{-8}$ Torr) pump down of the test chamber prior to back-filling with gas and prevent the occasional loss of a test due to leaks or contamination of the test atmosphere. All sliding seals have been eliminated in the new design and its all-metal construction will allow bake-out. Vacuum testing at less than 1×10^{-7} Torr should be possible. Design modifications of the high-temperature ($T > 900^\circ\text{C}$) creep-rupture test stands to provide more reliable testing are also in progress.

WORK PLANNED FOR NEXT PERIOD

Tensile testing of control and irradiated Mo-TZM sheet specimens will be initiated. Creep-rupture testing of Mo-TZM specimens will also be continued at 982°C .

Creep-rupture testing of tungsten specimens to determine the dose-dependency of creep rates and rupture lives will be initiated.

Hot-hardness testing of the four measured tungsten single crystals will be repeated following annealing at 1700°C for 1 hour. The hot-hardness tester is to be modified by the installation of a timer to allow conducting of isothermal hot-hardness studies.

Further measurements and analysis of resistivity changes on annealed irradiated molybdenum specimens will be conducted.

Final check-out and calibration of the Instron testing instrument is to be performed.

Computer studies on radiation damage in the bcc metals will be continued.

3. EVALUATIONS OF THE PLASTIC FATIGUE PROPERTIES OF HEAT RESISTANT ALLOYS

(57016)

The objective of this program is to determine the parameters affecting low-cycle fatigue behavior of metals and alloys at elevated temperatures and to generate low-cycle fatigue data for use in the design of structural components of high-performance nuclear reactor systems.

During this fiscal year, uniaxial low-cycle fatigue tests will be performed on Types 304 and 316 stainless steels in the temperature range from 400° to 800°C. In these tests, specimens will be subjected to cyclic loading conditions that are characterized by such parameters as strain amplitude, strain rate, and length of hold time at maximum strain.

METHODS OF TESTING

Modifications by the manufacturer were completed on the servo-controlled hydraulic testing machine to correct drift and phase-shift conditions previously experienced with the electronic control system. The basic features of the machine were discussed in an earlier report.¹ During the current report period tests were made to demonstrate the capabilities of the now completed fatigue testing assembly.

Figure 3.1 shows portions of actual stress and strain records obtained in tests on an aluminum specimen. The tests were performed to verify the ability to control the machine with the aid of an extensometer that monitors the diameter variation in a push-pull type specimen. The extensometer, as well as some preliminary results obtained with it, were discussed in a previous report.² The stress in Figure 3.1 was recorded by means of a load cell that measures the load being applied to the specimen. The diametral strain in Figure 3.1 is the percent change in the specimen diameter, as measured with the extensometer. Type 6061-T6 aluminum was selected because of its observed linear stress-strain relationship in the elastic range of the material. In earlier tests, Type 304 stainless steel exhibited a slightly nonlinear behavior in this range.

Figure 3.1 clearly illustrates that the diametral strain was made to vary in a linear fashion between preset limits. In Case (a) of the figure, the specimen is strain-cycled in the elastic range of the material. The stress in this case must vary in the same triangular mode as the strain, provided the output signal of the extensometer is proportional to the diametral strain, the command signal to the servo-amplifier and the feedback signal from the extensometer have the proper phase relationship, and the recorder has linear response. The stress record shows that these conditions prevailed.

In Case (b) of Figure 3.1, the strain amplitude has been doubled. The strain just exceeds the elastic limits of the material, but the stress variation remains essentially linear.

¹"High-Temperature Materials Program Progress Report No. 50, Part A," GE-NMPO, GEMP-50A, August 20, 1965, pp. 33-37.

²"Fourth Annual Report - High-Temperature Materials and Reactor Component Development Programs, Volume I - Materials," GE-NMPO, GEMP-334A, February 26, 1965, pp. 46-49.

Fig. 3.1 – Stress and strain records obtained in fully-reversed strain-cycling tests with a Type 6061-T6 aluminum specimen

In Case (c), the strain amplitude has been doubled once again. Considerable departure from linearity is now observed in the stress record because of pronounced yielding of the material. Any further increase in strain amplitude results in progressive yielding, as evident in Case (d).

Figure 3.2 shows the corresponding hysteresis loops obtained when stress is recorded versus diametral strain. To present the loops in their customary form, the stress was actually plotted against the negative of the diametral strain. For Case (a) of this figure, the loop is a straight line and a nearly straight line for Case (b). Assuming that the elastic modulus of the material has been measured in a tensile test, the slope of the straight line furnishes Poisson's ratio for elastic deformations. The horizontal width of the loops in Case (c) and Case (d) furnish the plastic portion of the diametral strain range. Subtraction of this portion from the total strain range gives the elastic portion of the diametral strain range. Separation of the diametral strain range into its elastic and plastic parts is necessary for the computation of the elastic and plastic parts of the axial strain range. The hysteresis loop is, therefore, an important piece of information to be recorded in low-cycle fatigue tests with diametral strain control.

Figure 3.3 shows results obtained with three different testing frequencies. In each test, the aluminum specimen was strain-cycled just within the elastic limits. A triangular mode was again selected, so that the performance of the system could be judged by the linearity observed in the stress record. On the basis of the straining rates achieved in these tests, it can be assumed that the extensometer meets the program requirements.

Figure 3.4 shows results obtained on a Type 304 stainless steel specimen in tests at three different temperatures. The strain amplitude is the same in each case, and it can be seen that at 800°C the stress range is less than half the stress range at room temperature. In these tests, induction-type heating was used to bring the gage section of the specimen to the desired temperature level. The induction coil that is placed around the specimen can be seen in Figure 3.5, which shows the test setup with the extensometer removed. The coil geometry was developed to give a flat temperature profile in the hour-glass gage section, while allowing access for the extensometer at the midposition.

Table 3.1 gives the temperature distribution in the specimen at various temperature levels. The measurements were made with chromel-alumel thermocouples welded to the

TABLE 3.1
TEMPERATURE DISTRIBUTION IN INDUCTION-HEATED FATIGUE SPECIMEN
(TYPE 304 STAINLESS STEEL)

	Temperature, °C
403	507	621	723	833	946
	418	525	629	746	858	973
	426	533	646	758	868	982
	427	536	648	762	869	977
	427	538	649	760	871	981
	425	536	648	762	869	979
	422	532	646	760	869	984
	414	523	637	746	863	987
	403	509	621	736	846	972

Legend:

d = minimum diameter = 6.35 mm.

R = profile radius = 38.1 mm.

p = thermocouple spacing = 3.2 mm.

Fig. 3.2—Hysteresis loops recorded in fully-reversed strain-cycling tests with a Type 6061-T6 aluminum specimen

Fig. 3.3 – Stress records for Type 6061-T6 aluminum specimen subjected to fully-reversed strain-cycling at three different frequencies

Fig. 3.4—Hysteresis loops recorded for a Type 304 stainless steel specimen subjected to fully-reversed strain-cycling at three different temperatures

Fig. 3.5 – Test setup with extensometer removed (Neg. P65-11-16B)

specimen surface. In fatigue tests, the temperature is monitored and controlled using a welded thermocouple located approximately 3 mm from the midposition. The uniformity of the temperature distribution in the gage section is also evident from the infra-red photograph shown in Figure 3.6, made with the specimen at 800°C. Outside the coil windings the temperature can be seen to drop off sharply. The temperature rise in the water-cooled fixture is insignificant.

Fig. 3.6 – Infrared photograph of a Type 304 stainless steel specimen heated to 800°C (Neg. P65-11-16A)

WORK PLANNED FOR NEXT PERIOD

Fatigue tests at elevated temperatures will be continued on Type 304 stainless steel specimens. Manufacturing of components and procurement of equipment for use on three additional testing machines will also be continued.

4. EFFECT OF RADIATION ON HEAT RESISTANT METALS AND ALLOYS

(57018)

The objective of this program is to determine the effect of radiation on the time-, temperature-, and stress-dependent properties of heat-resistant metals and alloys at elevated temperatures, to identify the causes of any observed changes in these properties, and to develop remedial measures.

Materials to be investigated in this program include A-286, Hastelloy N (INOR-8), Hastelloy R-235, Hastelloy X, Fe-Cr-Al-Y alloys, Type 304 stainless steel, and ASTM-A350-LF3 pressure vessel steel.

EXPERIMENTAL PROGRAM

Status of Irradiations

A summary of irradiation capsules of current interest is shown in Table 4.1. Capsule GEFP2-152, containing 18 Hastelloy X specimens, is currently being irradiated in the ETR (Cycle 76) at 650°C. Capsule GEFP2-155 was fabricated and shipped to ITS and will be irradiated in the ETR during Cycle 77 at a temperature of 760°C and to a fast neutron dose of approximately 10^{20} nvt ($E_n \geq 1$ Mev). This capsule contains 18 Hastelloy N specimens; nine are in the as-received (mill annealed) condition; the remainder were annealed 4 hours at 870°C in argon.¹

Hastelloy N Creep-Rupture Testing

Creep-rupture testing of irradiated Hastelloy N specimens at a temperature of 650°C is continuing. The results for all specimens tested at 650°C in the as-irradiated condition are shown in Figure 4.1. In evaluating the data, it must be pointed out that several known variables are present:

1. Each capsule was irradiated to a slightly different dose.
2. Three different irradiation temperature conditions are involved:
 - a. 5 hours at 650°C followed by a rapid water quench (leak in capsule) and the remainder of the cycle (702 hours) at reactor ambient temperature (GEFP2-128).
 - b. Approximately 650°C for 80 hours followed by a rapid water quench (due to a leak) and the remainder (320 hours) of the cycle at reactor ambient temperature (GEFP2-139).
 - c. Approximately 650°C for a full reactor cycle (GEFP2-148 and -151).
3. Two different heats of material are involved.
4. Two different vendors manufactured the specimens; hence, surface stresses probably vary. This latter point became evident when it was discovered that specimens from one vendor showed a significant change in concentricity after annealing 4 hours

¹"High-Temperature Materials Program Progress Report No. 52, Part A." GE-NMPO, GEMP-52A, October 20, 1965, pp. 32-33.

TABLE 4.1
IRRADIATION DATA FOR HIGH-TEMPERATURE ALLOY CAPSULES

Capsule	Specimen Data				Facility	Irradiation Conditions			Temperature Data	
						Neutron Data		Fast, nvt ($E_n \geq 1$ Mev)		
						Material	Heat No.		Type ^a	Quantity
ORM-39	A-302B		Res	1	ORR-Rabbit		3.3×10^{18}	4.1×10^{17}	5	Reactor ambient
			H	1						
NRL-1	A-302B		Res	5	IRL ^b			9×10^{17} to 6×10^{18c}		Reactor ambient
	A-302B		H	3						
	310 SS		Res	5						
	Tungsten	Rod J	Res	5						
33MT-99	A-286+0.010B	RV-403-5C	CR	7	ETR-K3	7910	6.5×10^{20}	4.8×10^{19}		Reactor ambient
	A-286+0.0043B	RV-403-5B	CR	7						
	A-286+0.0008B	RV-403-5A	CR	7						
	A-286+0.0065B ^g	RV-451	CR	7						
GEFP2-128	Hastelloy N	5081	CR	18	ETR-E5-NW	5154	5.0×10^{20}	8.8×10^{19}	d	d
GEFP2-139	Hastelloy N	5081	CR	6	ETR-E5-NW	2920	2.4×10^{20}	7.2×10^{19}	e	e
	A-286+0.010B	RV-403-5C	CR	3						
	A-286+0.0043B	RV-403-5B	CR	1						
	A-286+0.0008B	RV-403-5A	CR	4						
	A-286+0.0065B ^g	RV-451	CR	4						
GEFP2-148	Hastelloy N	5081	CR	3	ETR-N14-SE	3348	2.4×10^{20}	4.6×10^{19}	376	~650
	Hastelloy N	5085	CR	6						
	A-286+0.010B	RV-403-5C	CR	3						
	A-286+0.0043B	RV-403-5B	CR	3						
	A-286+0.0008B	RV-403-5A	CR	3						
GEFP2-151	Hastelloy N	5065, 5067	CR(W)	6	ETR-N14-SE	3920	2.9×10^{20f}	5.5×10^{19f}	440	~650
	Hastelloy N	5081	CR	12						
GEFP2-152	Hastelloy X	E-9500	CR	18	ETR-N14-SE			1×10^{20c}		650 ^c
GEFP2-155	Hastelloy N	5081	CR	18				1×10^{20c}		760 ^c

^aCR = Round creep-rupture specimen

(W) = Welded specimen (filler metal is from heat 5101)

Res = Resistivity

H = Hardness

^bIndustrial Reactor Laboratory (Plainsboro, New Jersey).

^cRequested.

^dOperated first 5 hours at ~650°C and the remainder (702 hours) of the cycle at reactor ambient temperature.

^eOperated first 80 hours at ~650°C and the remainder (320 hours) of the cycle at reactor ambient temperature.

^fEstimated dose.

^gB¹¹ isotope - 98.6%

○ Control: Mill-annealed; heat 5081.

Irradiated

□ GEFP2-128: 8.8×10^{19} nvt ($E_n > 1$ Mev);
5 hours at $\sim 650^\circ\text{C}$ plus 702 hours at reactor
ambient temperature; heat 5081.

▽ GEFP2-139: 7.2×10^{19} nvt ($E_n > 1$ Mev);
80 hours at $\sim 650^\circ\text{C}$ plus 320 hours at reactor
ambient temperature; heat 5081.

◇ GEFP2-148: 4.6×10^{19} nvt ($E_n > 1$ Mev);
376 hours at $\sim 650^\circ\text{C}$; heat 5085.

△ GEFP2-151: 5×10^{19} nvt ($E_n > 1$ Mev);
440 hours at $\sim 650^\circ\text{C}$; heat 5081.

Fig. 4.1 – Stress-rupture properties of Hastelloy N at 650°C

at 870°C ; whereas, specimens from the other vendor showed no appreciable change. Specimens from capsule GEFP2-151 and all those in subsequent capsules contain only the low surface stress material; previous capsules contain specimens from both vendors.

In view of the above factors and evaluation of results from further testing of specimens irradiated for a complete cycle at 650°C , it is now believed that reactor ambient and elevated-temperature (650°C) irradiations produce essentially the same reduction in rupture life when tested at 650°C in the as-irradiated condition as shown in Figure 4.1. Note that this is contrary to the preliminary conclusion reported previously² where it was stated that the elevated-temperature irradiations (GEFP2-139) showed a further reduction in the creep-rupture properties compared to the ambient temperature irradiations. Post-test dimensional measurements are not complete; hence, the effect of irradiation temperature on ductility will be discussed in a future report.

A-286 Creep-Rupture Testing

A number of A-286 specimens irradiated in capsule GEFP2-139 were tested at a temperature of 650°C . One specimen from each of the split heats³ containing various amounts

²Ibid., p. 29.

³"Third Annual Report – High-Temperature Materials and Reactor Component Development Programs, Volume I – Materials," GE-NMPO, GEMP-270A, February 28, 1964, p. 78.

of boron was tested at 42.2 kg/mm^2 and 650°C . A comparison of the results from this screening study as well as the results for control specimens and those specimens irradiated in capsule 33MT-99³ are shown in Figure 4.2. It can be seen that both the high- and low-boron content specimens from capsule GEFP2-139 showed significant reduction of rupture life whereas for capsule 33MT-99, only the specimens with low-boron content showed loss of rupture life. However, it should be noted that the low-boron-containing specimens from capsule GEFP2-139 showed a larger decrease in rupture life compared to the high-boron-containing specimens; hence, the results for the two capsules appear to be in qualitative agreement. Further testing of specimens containing various quantities of boron is being performed.

Fig. 4.2 – Stress-rupture properties of A-286 alloy at 650°C

Hastelloy R-235

To obtain further information on the influence of boron on the ductility of irradiated high-temperature alloys, a special split heat containing specimens with the same total boron content but with different isotopic concentrations was prepared. Since it is believed that the elevated-temperature embrittlement, due to neutron irradiation, is more pronounced in the nickel-based precipitation hardening (γ' precipitate) alloys, an alloy with low-cobalt content, Hastelloy R-235, was selected for the study. A similar investigation on an iron-based precipitation hardening alloy (A-286) is also being considered for a later study.

A split heat of commercial-grade (low cobalt) Hastelloy R-235 with carefully controlled boron additions was supplied by the Haynes Stellite Company.⁴ GE-NMPO furnished the

⁴W. D. Manley, Haynes Stellite Company, a Division of Union Carbide Corporation, Kokomo, Indiana.

boron additives in the form of Ni-B buttons. The four heats result in the following nominal boron concentrations:

1. One ingot containing 0.006 weight percent natural boron (19.8% B¹⁰; 80.2% B¹¹).
2. One ingot containing 0.006 weight percent total boron. The boron additive was enriched to 98 percent B¹¹ (i.e., 2% B¹⁰).
3. One ingot containing 0.006 weight percent total boron. The boron additive was enriched to 96 percent B¹⁰.
4. One ingot in which no boron addition was made; hence, the only boron present is that normally present due to melting procedures which is, in this case, about 0.001 weight percent natural boron.

The four materials are currently being fabricated to 1.27-cm-diameter rod and will be machined into creep and tensile specimens for an elevated-temperature irradiation in the ETR, presently scheduled for Cycle 80. An ambient temperature ORR capsule will be fabricated to obtain preliminary data at an earlier date.

Fe-Cr-Al-Y

Concurrent with the work being performed on irradiation effects to heat-resistant alloys as reported herein and for the refractory metals and alloys reported under Job 57004, "Effect of Radiation on Refractory Metals and Alloys," a program to study the effects of irradiation on Fe-Cr-Al-Y alloys is also being performed under Job 57001, "Oxidation Resistant Fuel Element Materials Research," (reported in even numbered progress reports) where this alloy was developed. Studies on this alloy are being performed both at GE-NMPO and in the Metallurgy Division of UKAEA, Harwell, England.⁵

NMPO Results⁶ - The four alloy compositions shown in Table 4.2 were irradiated to 9.25×10^{19} nvt ($E_n \geq 1$ Mev) at reactor ambient temperature. Changes in ductility due to irradiation were determined by bending around a 2t radius, where t is the thickness of the specimen. Prior to irradiation, all alloys exhibited a full 90-degree bend without fracture at a temperature of -79°C. After irradiation, the transition temperature for the 1541 and 2541 alloys was increased to between -79°C and -54°C while the transition for the 0561 and 1041 alloys remained below -79°C.

TABLE 4.2
CHEMICAL ANALYSES OF Fe-Cr-Al-Y ALLOYS IRRADIATED IN GEFP2-220^a

Alloy	Heat No.	Chemical Analyses							
		wt %			ppm				
		Cr	Al	Y	C	P	Si	S	
0561 (Fe - 5Cr - 6Al - 1Y)	M-236	4.71	6.51	0.39	73	3	30	20	
1041 (Fe - 10Cr - 4Al - 1Y)	M-239	9.66	4.63	0.23	61	7	30	10	
1541 (Fe - 15Cr - 4Al - 1Y)	MS-38	14.49	4.48	0.59	30	59	20	16	
1541 FC ^b	MS-41	15.36	4.44	0.43	31	91	85	14	
2541 (Fe - 25Cr - 4Al - 1Y)	MS-36	25.57	4.30	0.41	48	58	29	11	

^aTotal dosage: 9.25×10^{19} nvt ($E_n \geq 1$ Mev) at reactor ambient temperature.

^bFerrochromium used instead of hydrogen-reduced Cr.

⁵Ferrochrome alloys with and without yttrium additions have been supplied to UKAEA from master heats prepared at GE-NMPO. Studies on corrosion, fabricability, irradiations effects, etc., will be performed by the Harwell group and the results made available to GE-NMPO. The radiation effects data will be presented under Job 57018.

⁶"High-Temperature Materials Program Progress Report No. 53, Part A," GE-NMPO, GEMP-53A, pp. 11-22.

Isothermal annealing of irradiated specimens was performed at 250°C and 300°C. All the alloys still exhibited a slight residual hardness after annealing for 1 hour at 250°C. A 0.5-hour heat treatment at 300°C removed all the radiation-induced hardness except for the 1041 alloy which still showed residual hardness values after a 1-hour anneal at this temperature.

UKAEA Results - Preliminary results have been received from the UKAEA (Harwell)⁷ on materials furnished by GE-NMPO. The results of post-irradiation tensile tests performed at elevated temperatures, listed in Table 4.3, show very little changes in yield strength, ultimate tensile strength, and ductility. Of particular significance is the retention of the high-temperature elongation values following irradiation.

TABLE 4.3
RESULTS OF TENSILE TESTING OF CONTROL AND IRRADIATED Fe-Cr-Al-Y^a

Alloy ^b	Specimen	Test Temperature, °C ^c	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Total Elongation, %
1541	Control	650	12,900	15,800	71.4
	Irradiated ^d		11,900	15,800	57.6
	Control	750	8,500	8,500	108.8
			7,900	7,900	103.9
	Irradiated ^d		8,000	8,300	109.0
			7,800	8,100	85.9
	Control	850	4,600	4,600	116.8
	Irradiated ^d		4,400	4,500	108.0
2541	Control	650	16,600	20,000	70.0
	Irradiated ^d		16,200	18,700	73.6
	Control	750	9,600	9,600	110.7
			9,400	9,800	105.9
	Irradiated ^d		8,600	9,800	89.5
			8,800	9,500	118.5
	Control	850	5,100	5,200	150.0
	Irradiated ^d		5,200	5,400	131.7

^aTests and irradiation performed by UKAEA (Harwell).

^bBoth alloys were in the form of cold-rolled sheet and were heat treated 1 hour at 1000°C, rapid air cooled, prior to irradiation or control testing.

^cInitial strain rate 2×10^{-4} sec⁻¹. Just prior to fracture there was a temperature gradient of 20°C (max) along the gage lengths of the specimens exhibiting the highest elongation to fracture.

^dSpecimens were irradiated at 40°C in contact with the heavy water in the DIDO reactor. Approximate neutron doses: 2×10^{20} nvt (thermal); 7×10^{19} nvt (fission).

WORK PLANNED FOR NEXT PERIOD

Creep-rupture testing of Hastelloy N specimens irradiated at 650°C will continue. Testing of irradiated welded specimens will be initiated. Metallographic examination of control and irradiated Hastelloy N specimens will be continued as they become available.

Testing and evaluation of A-286 specimens containing various concentrations of boron and irradiated at 650°C will also continue. These will be compared to other data generated from these same heats of material.

Tensile tests will be performed on the 1541 alloy irradiated at the elevated temperature.

⁷D. H. Harries, Metallurgy Division, Atomic Energy Research Establishment, Harwell, England.

5. RADIATION EFFECTS IN BeO

(57063)

The purpose of this program is to define the irradiation behavior of BeO in terms of the composition and microstructure variables that contribute to extended radiation stability.

IRRADIATION STATUS

The two irradiation tests in progress in the ETR continued to operate satisfactorily. Test GEFP2-134 containing 2.5-cm-diameter by 2.5-cm-long specimens has operated at 900°C for 5 reactor cycles. Test GEFP2-132 is a further irradiation at 1000°C of specimens previously irradiated and examined after dosages of 2 to 4×10^{21} nvt ($E_n \geq 1$ Mev); this test has continued through 3 reactor cycles. Both tests will terminate in January 1966.

MEASUREMENTS OF IRRADIATED BeO

Irradiations at 100°C

Post-irradiation measurements of the changes in the dimensions, volume, open porosity, and strength were continued on specimens irradiated at 100°C. These measurements are part of the studies to evaluate the irradiation behavior of sintered specimens fabricated from different grades of BeO with and without additives and, to some extent, by different production processes. In these evaluations, the most important factor is the resistance to microcracking; i. e., to separation of the grains at the boundaries as a result of strains introduced by anisotropic lattice expansion. Microcracking is indicated by increased expansion and open porosity and by significant decreases in the strength; it is the most important single factor affecting the degradation of BeO materials even though it occurs only at higher dosages in elevated-temperature irradiations. In previous work, grain size has shown the greatest influence on stability with preferential orientation of the c-axis type providing some improvement in a given grain size. Composition differences have not appeared significant in the AOX and UOX grades of BeO, or in the latter material with MgO and ZrO₂ additives. As noted in the previous report,¹ however, a composition containing 0.4 weight percent Al₂O₃ and 0.1 weight percent SiO₂ in UOX² provided the first evidence obtained in this program that compositional variations could improve the resistance to microcracking.

Most of the compositions examined during this period appeared to possess "normal" resistance to microcracking; however, one composition with relatively good and one with poor resistance were observed. The compositions examined included 1 and 2 percent bentonite in UOX, 1 percent Al₂O₃ in UOX, 0.5 percent MgO in Minox AAA, and 3 percent ZrO₂ in UOX. All of the compositions appear to be single phase except the latter,

¹"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, pp. 40-42.

²The specimens examined were fabricated by the Brush Beryllium Company and are identified as Thermalox 995A.

in which a separate zirconia phase occurs. The composition containing 1 percent bentonite is being used in the Experimental Beryllium Oxide Reactor (EBOR); specimens cut from five different EBOR production blocks were examined. Results for the various compositions are summarized in Table 5.1.

The bentonite compositions appeared to possess normal resistance to microcracking, although there were some variations. All of the EBOR materials were microcracked at dosages less than 2×10^{20} nvt ($E_n \geq 1$ Mev). There appeared to be somewhat less grain size dependence than might have been expected; differences in the expansion and strength in specimens of 17- and 28-micron grain size were not appreciable. It may be significant that, although the bulk densities were comparable, the 17-micron grain size material contained more open porosity than the other EBOR materials; there is no separate evidence on this point, however.

The remaining three bentonite compositions listed in Table 5.1 were fabricated at GE-NMPO by extrusion and contain appreciable amounts of preferred orientation of the c-axis of the grains along the longitudinal axis of the specimens. This preferred orientation is considered to account for the somewhat better radiation resistance of these materials than the EBOR specimens of comparable grain size. There was no appreciable difference between the 1- and 2-percent bentonite additions in specimens of 18- and 20-micron grain size. The 1-percent bentonite specimens of 8-micron grain size did not decrease in strength up to the maximum dosage of 2.1×10^{20} nvt ($E_n \geq 1$ Mev), indicating that microcracking was not extensive if it occurred at all. This material may be comparable to the UOX + Al_2O_3 - SiO_2 composition which appeared to possess somewhat superior radiation resistance in previous work.

The UOX + 1 weight percent Al_2O_3 composition appeared very susceptible to microcracking. There were large increases in the open porosity and the strength had decreased to about 60 percent of the unirradiated value at the lowest dosage, about 0.9×10^{20} nvt ($E_n \geq 1$ Mev). The microstructure of the matrix of this material consists of 1- to 2-micron grain size with a more or less uniform distribution of 10 to 20 micron grains at separation distance of 30 to 40 microns. Since the properties of such materials are considered to be governed by the matrix grain size, the poor resistance might be attributed to the Al_2O_3 additive; however, considering that anisotropic lattice expansion is the cause of microcracking, it is plausible that the non-uniformity of the grains is responsible for the poor irradiation behavior.

The remaining two compositions, Minox AAA + MgO and UOX + ZrO_2 , exhibited no unusual features. Both materials decreased in strength at dosages between 1 and 2×10^{20} nvt and, in general, appeared comparable to the EBOR materials and to materials tested previously.

THERMAL DIFFUSIVITY

Measurements of the thermal diffusivity were completed from 300° to 1000°C on two materials in the unirradiated and irradiated condition. The measurements were made following slight modifications of the equipment and further checks on the accuracy and precision of the measurements.

One of the materials examined was UOX + MgO of 4-micron grain size and density of 2.92 g/cm^3 . The irradiated specimens were exposed at 1000°C to 2.5×10^{21} nvt ($E_n \geq 1$ Mev) and, at the end of the exposure, to approximately 2×10^{19} nvt at about 650°C; volume expansion was 2 percent. Both the unirradiated and the irradiated specimens were from the same extrusion mix and sinter and were of the same density. Results

TABLE 5.1
DIMENSION AND STRENGTH CHANGES IN BeO COMPOSITIONS IRRADIATED AT 100°C

Specimen Description			Irradiation Dosage, 10 ²⁰ nvt (E _n ≥ 1 Mev)	Expansion, %			Increase In Open Porosity ^a	Modulus-Of-Rupture At Room Temperature		
Composition	Grain Size, microns	Density, g/cm ³		Length	Diameter	Volume		10 ³ psi	10 ³ kg/cm ³	Percent Of Unirradiated Value
EBOR ^b	26	2.78	0.9	0.1	0.1	0.3	0.1	41.3	2.90	128
B-78-127			1.1	0.2	0.1	0.4	0.2	42.6	3.00	132
			1.1	0.2	0.2	0.5	0.0	37.9	2.66	118
			1.4	0.3	0.2	0.7	0.3	28.8	2.02	89
			1.8	0.4	0.4	1.2	5.0	21.4	1.50	66
			1.8	0.4	0.4	1.2	5.6	21.8	1.53	68
			2.2	0.5	0.4	1.3	3.9	17.7	1.24	55
			2.2	0.5	0.5	1.5	5.1	13.2	0.93	41
EBOR	17	2.79	1.0	0.1	0.1	0.3	0.0	41.5	2.92	106
B-75-93			1.0	0.2	0.1	0.4	0.0	36.4	2.56	93
			1.3	0.2	0.1	0.5	0.0	37.5	2.64	96
			1.4	0.2	0.2	0.6	0.0	31.2	2.19	80
			2.0	0.3	0.3	1.0	2.8	30.8	2.16	79
			2.1	0.4	0.5	1.4	4.3	22.3	1.57	57
EBOR	24	2.80	1.0	0.1	0.1	0.3	0.0	34.4	2.42	109
B-79-100			1.0	0.1	0.1	0.3	0.0	41.4	2.91	131
			1.3	0.2	0.2	0.5	0.0	56.0	3.94	177
			1.4	0.2	0.2	0.6	0.0	43.7	3.07	138
			2.0	0.3	0.4	1.1	4.2	19.4	1.36	61
			2.1	0.4	0.5	1.4	4.6	13.7	0.96	43
EBOR	29	2.80	0.9	0.1	0.1	0.3	0.2	34.1	2.40	128
B-86-47			1.0	0.1	0.1	0.3	0.2	33.6	2.36	126
			1.0	0.1	0.1	0.3	0.0	37.6	2.64	141
			1.1	0.2	0.1	0.4	0.1	36.1	2.54	136
			1.1	0.2	0.1	0.4	0.0	34.6	2.43	130
			1.2	0.2	0.2	0.6	1.0	30.0	2.11	113
			1.3	0.3	0.3	0.9	2.7	19.4	1.36	73
			1.9	0.4	0.4	1.2	3.2	17.1	1.20	64
			2.1	0.4	0.4	1.2	3.7	12.2	0.86	46
			2.2	0.5	0.4	1.3	6.1	14.2	1.00	53
EBOR	26	2.78	0.9	0.1	0.1	0.3	0.0	34.8	2.45	112
B-87-107			0.9	0.2	0.1	0.3	0.0	34.9	2.45	112
			1.0	0.2	0.1	0.3	0.0	39.3	2.76	126
			1.1	0.2	0.1	0.4	0.0	38.5	2.71	124
			1.2	0.2	0.1	0.4	0.0	30.9	2.17	99
			1.3	0.2	0.2	0.6	0.1	39.6	2.78	127
			1.9	0.4	0.4	1.2	4.7	16.8	1.18	54
			2.2	0.5	0.4	1.3	5.6	14.7	1.03	47
UOX + 1% Bentonite	8	2.75	1.0	0.1	0.1	0.3	0.4	—	—	—
1.0			0.1	0.1	0.3	0.1	38.9	2.74	106	
1.2			0.2	0.1	0.4	0.1	39.6	2.78	108	
1.3			0.2	0.2	0.5	0.1	40.6	2.86	111	
1.9			0.3	0.2	0.7	0.1	39.7	2.79	109	
2.1			0.4	0.3	1.0	0.2	42.2	2.97	116	

TABLE 5.1 (Cont.)

DIMENSION AND STRENGTH CHANGES IN BeO COMPOSITIONS IRRADIATED AT 100°C

Specimen Description		Grain Size, microns	Density, g/cm ³	Irradiation Dosage, 10 ²⁰ nvt (E _n ≥ 1 Mev)	Expansion, %			Increase In Open Porosity ^a	Modulus-Of-Rupture At Room Temperature		
Composition					Length	Diameter	Volume		10 ³ psi	10 ³ kg/cm ³	Percent Of Unirradiated Value
UOX+1% Bentonite	18	2.84	1.0	0.1	0.1	0.1	0.3	0.0	40.0	2.81	115
				1.0	0.1	0.1	0.3	0.2	44.1	3.10	127
				1.3	0.2	0.2	0.5	0.0	42.1	2.98	121
				2.0	0.4	0.3	1.0	0.4	38.1	2.68	110
				2.1	0.4	0.4	1.2	2.3	33.4	2.35	96
UOX+2% Bentonite	20	2.75	0.9	0.1	0.1	0.1	0.3	0.6	35.8	2.52	102
				1.1	0.2	0.1	0.4	0.3	37.4	2.63	107
				1.1	0.2	0.1	0.4	0.2	43.6	3.07	125
				1.4	0.3	0.2	0.7	0.5	36.1	2.54	103
				1.8	0.4	0.3	1.0	0.5	33.8	2.38	97
UOX+1% Al ₂ O ₃	2	2.70	0.9	0.1	0.1	0.1	0.3	3.4	26.2	1.84	75
				1.0	0.1	0.1	0.3	10.4	26.8	1.88	58
				1.0	0.2	0.1	0.4	10.0	26.4	1.86	58
				1.1	0.2	0.1	0.4	7.7	26.2	1.84	57
				1.1	0.2	0.1	0.4	9.1	23.2	1.63	50
Minox AAA + 0.5% MgO ^c	18	2.87	1.0	0.1	0.1	0.1	0.3	9.6	26.4	1.86	58
				1.2	0.2	0.2	0.6	10.3	21.1	1.48	46
				1.3	0.3	0.2	0.7	7.5	29.8	2.10	65
				2.1	0.4	0.3	1.0	8.1	31.2	2.19	68
				0.9	0.1	0.1	0.3	0.0	35.9	2.52	150
UOX + 3% ZrO ₂	19	2.97	1.0	0.1	0.1	0.1	0.3	0.0	21.8	1.53	91
				1.0	0.2	0.1	0.4	2.1	19.1	1.34	80
				1.1	0.2	0.1	0.4	0.0	32.3	2.27	135
				1.2	0.2	0.2	0.5	2.4	—	—	—
				1.3	0.2	0.2	0.6	2.8	11.8	0.83	49
			1.7	0.2	0.3	0.3	0.8	2.6	13.7	0.98	57
				1.9	0.3	0.4	1.1	2.9	9.2	0.65	38
				2.1	0.4	0.5	1.4	3.6	4.7	0.33	20
				0.9	0.1	0.1	0.3	0.2	28.6	2.01	117
				1.0	0.2	0.1	0.4	0.6	20.1	1.41	82
			1.0	0.2	0.1	0.1	0.3	0.3	17.4	1.22	71
				1.2	0.2	0.2	0.5	0.7	21.7	1.53	89
				1.3	0.3	0.2	0.7	0.7	19.9	1.40	82
				1.3	0.3	0.2	0.8	1.2	17.3	1.22	71
				1.9	0.4	0.3	1.0	0.7	15.1	1.06	62
			2.2	0.5	0.4	0.4	1.3	0.8	—	—	—

^aValues listed are the difference in percent open porosity before and after irradiation.

^bThe EBOR composition is 1 weight percent Bentonite in UOX-grade BeO. "B-" numbers listed are the numbers of the production blocks from which samples were obtained.

^cMinox AAA grade BeO is a product of the Mineral Concentrates Co., Denver, Colorado.

for this material are summarized in Figure 5.1 in which the diffusivity of the control and the irradiated specimens are compared as a function of temperature.

Fig. 5.1 - Thermal diffusivity changes in BeO irradiated at 1000°C. Specimen was UOX + 0.5 wt % MgO, 4-micron grain size, and density of 2.92 g/cm³; after irradiation at 1000°C to 2.3×10^{21} nvt ($E_n \geq 1$ Mev), density was 2.86 g/cm³.

The curves show that the diffusivity of the as-irradiated specimen decreased by amounts ranging from about 20 percent at 300°C to about 4 percent at 1000°C. Because of the wide diffusivity change in BeO over the measured temperature range, different specimens are required for temperatures above and below 600°C. In general, very good reproducibility was obtained with the different specimens in the several hundred degree range in which both specimens can be measured. In view of this result, the differences observed between two different specimens of the irradiated material used in the 300° to 600°C range, are probably representative of real variations in the irradiated material. The heating curve for the irradiated material shows two small "jumps" which are attributed to thermal annealing during overnight exposure at 600°C and 855°C during the experimental run. The annealing is assumed to occur as a result of the low-temperature exposure at the end of the irradiation. Measurements were also made on specimens maintained overnight (17 hours) at the highest measurement temperature (995°C); these measurements were made during cooling. In this annealed condition, the diffusivity was approximately constant at 96 percent of that of the unirradiated material.

TABLE 5.2

THERMAL DIFFUSIVITY AND CONDUCTIVITY OF BeO BEFORE AND AFTER IRRADIATION AT 1000°C

Specimen Description	Temperature, °C	Thermal Diffusivity, cm ² /sec	Computed Values ^b		
			Density, g/cm ³	Specific Heat, ^c cal/g-°C	Thermal Conductivity, cal/cm-sec-°C
Unirradiated					
UOX+MgO, 4-micron grain size Bulk density 2.92 g/cm ³	300	0.195	2.904	0.397	0.225
	322	0.187	2.902	0.403	0.219
	348	0.177	2.900	0.410	0.211
	371	0.168	2.899	0.416	0.203
	404	0.157	2.896	0.423	0.193
	447	0.143	2.893	0.432	0.179
	501	0.127	2.889	0.441	0.162
	555	0.112	2.885	0.450	0.146
	598	0.103	2.881	0.456	0.135 ^d
	500	0.126	2.889	0.441	0.161
	598	0.102	2.881	0.456	0.134
	698	0.084	2.873	0.468	0.112
	798	0.071	2.864	0.479	0.098
	900	0.060	2.854	0.489	0.083
	996	0.052	2.845	0.498	0.074
Irradiated at 1000°C to 2.3×10^{21} nvt ($E_n \geq 1$ Mev)					
Bulk density 2.86 g/cm ³	300	0.154	2.844	0.397	0.174
	352	0.142	2.840	0.411	0.166
	397	0.132	2.837	0.422	0.156
	448	0.119	2.833	0.432	0.146
	496	0.109	2.830	0.440	0.136 ^d
	406	0.134	2.836	0.424	0.161
	448	0.123	2.833	0.432	0.151
	502	0.111	2.829	0.441	0.138
	550	0.100	2.826	0.449	0.127
	600	0.094	2.822	0.456	0.121
	650	0.086	2.818	0.463	0.112
	700	0.079	2.813	0.469	0.104
	749	0.072	2.809	0.474	0.096
	801	0.067	2.804	0.480	0.090
	855	0.061	2.800	0.485	0.084
	907	0.056	2.795	0.490	0.077
	952	0.052	2.791	0.494	0.073
	996	0.050	2.787	0.498	0.069
Annealed for 17 hours at 995°C after irradiation					
Bulk density 2.86 g/cm ³	303	0.187	2.844	0.398	0.212
	351	0.172	2.840	0.411	0.201
	399	0.156	2.837	0.422	0.187
	452	0.139	2.833	0.433	0.171
	498	0.125	2.830	0.441	0.156 ^d
	496	0.123	2.830	0.440	0.153
	550	0.110	2.826	0.449	0.140
	573	0.106	2.824	0.452	0.135
	594	0.101	2.822	0.455	0.130
	598	0.101	2.822	0.456	0.130
	628	0.095	2.819	0.460	0.123
	647	0.091	2.818	0.462	0.118
	771	0.073	2.807	0.476	0.098
	888	0.058	2.797	0.488	0.080

^aUnirradiated and irradiated specimens were obtained originally from the same extrusion mix and sinter and were of the same density.

^bSee Table 5.3 for conversion relationships.

^cIt is assumed that the specific heat is not changed by irradiation; measurements have established that there is no change (~2% detection limit) in irradiations at 1000°C to 10^{21} nvt ($E_n \geq 1$ Mev).

^dData above and below the lines were obtained from specimens of different lengths.

The measured thermal diffusivity and the computed thermal conductivity data are presented in Table 5.2. The thermal conductivity values shown in Table 5.2 were computed from the diffusivity data by the method shown in Table 5.3.

TABLE 5.3
THERMAL DIFFUSIVITY TO THERMAL CONDUCTIVITY
CONVERSION RELATIONSHIPS

$K = \alpha \rho C_p$ where	K = thermal conductivity, cal/cm-sec-°C
	α = thermal diffusivity, cm ² /sec
	ρ = density, g/cm ³
	C_p = specific heat, cal/g-°C
Density, ρ , was computed from thermal expansion data for randomly oriented BeO using $\rho = \rho_0 / (1 + \Delta L / L)^3$ where ρ_0 = density at room temperature, ΔL = length change, and	
$\% \Delta L = 5.866 \times 10^{-4} (T-25) + 3.87 \times 10^{-7} (T-25)^2 - 7.034 \times 10^{-11} (T-25)^3$ for T in °C.	
Specific heat was computed from the enthalpy equation of Victor and Douglas (<u>Journal of Research of the National Bureau of Standards</u> , Vol. 67A, No. 4, 1963, p. 325). Differentiation of the enthalpy equation gives	
$C_p = \frac{1}{25.012} (11.1084 + 14.249 \times 10^{-4} T - 8.40705 \times 10^5 T^{-2} + 10.6249 \times 10^7 T^{-3})$ for T in °K.	

The second material examined was UOX + MgO of 18-micron grain size and density of 2.93 g/cm³. The irradiated specimen in this case had been exposed to 6.1×10^{20} nvt at 100°C and had a volume expansion of 4.2 percent of which about 2.2 percent was due to voids formed by microcracks. The diffusivity curve (Figure 5.2) for the unirradiated material paralleled that of the curve for the unirradiated 4-micron grain size specimen described in the preceding paragraphs but was about 5 percent lower. The diffusivity of the irradiated specimen decreased markedly – it was less than 10 percent of the unirradiated value at 300°C and decreased slowly with increasing temperature. Relatively large changes in the diffusivity were anticipated in the irradiated sample at the higher measurement temperatures because of annealing; accordingly, the specimen was annealed at 1000°C for 100 hours to avoid transient changes during further measurements. This treatment is sufficient to anneal the lattice expansion but only partially removes the microcracks, as indicated by an annealed density of 2.90 g/cm³. As shown in Figure 5.2, the diffusivity of the irradiated specimen after annealing is about 90 percent of that of the unirradiated specimen. The diffusivity and computed thermal conductivity values are summarized in Table 5.4.

WORK PLANNED FOR NEXT PERIOD

Post-irradiation measurements to determine resistance to microcracking in 100°C irradiations will be made on specimens containing additives of low-, medium-, and high-silica glasses. Thermal diffusivity measurements will be made on specimens irradiated at 1000°C to about 4×10^{21} nvt and at 600°C to about 2.5×10^{20} nvt ($E_n \geq 1$ Mev).

Fig. 5.2 - Thermal diffusivity changes in BeO irradiated at 100°C . Specimen was UOX + 0.5 wt % MgO, 18-micron grain size, density of 2.93 g/cm^3 . After irradiation to $6.1 \times 10^{20} \text{ nvt}$ ($E_n \geq 1 \text{ Mev}$), specimen was extensively microcracked; density of 2.81 g/cm^3 .

TABLE 5.4

THERMAL DIFFUSIVITY AND CONDUCTIVITY OF BeO BEFORE AND AFTER IRRADIATION AT 100°C

Specimen Description ^a	Temperature, °C	Thermal Diffusivity, cm ² /sec	Computed Values ^b		
			Density, g/cm ²	Specific Heat, ^c cal/g-°C	Thermal Conductivity, cal/cm-sec-°C
<u>Unirradiated</u>					
UOX + MgO, 18-micron grain size					
Bulk density 2.93 g/cm ³	304	0.185	2.913	0.398	0.215
	354	0.167	2.910	0.412	0.201
	401	0.151	2.906	0.422	0.185
	446	0.138	2.903	0.432	0.172
	502	0.122	2.899	0.441	0.156
	554	0.107	2.894	0.450	0.139
	597	0.097	2.891	0.456	0.128
	649	0.088	2.887	0.463	0.117
	701	0.079	2.882	0.469	0.107
	748	0.073	2.878	0.474	0.100
	804	0.065	2.873	0.480	0.089
	850	0.060	2.869	0.484	0.083
	901	0.056	2.864	0.489	0.078
	1006	0.048	2.854	0.498	0.068
<u>Irradiated at 100°C to 6.1×10^{20} nvt ($E_n \geq 1$ Mev), Volume expansion 4.2 percent, Open porosity 3.5 percent</u>					
Bulk density 2.81 g/cm ³	303	0.016	2.794	0.398	0.018
	354	0.015	2.791	0.412	0.017
	399	0.014	2.787	0.422	0.017
	499	0.013	2.780	0.441	0.016
	602	0.012	2.772	0.456	0.015
	700	0.012	2.764	0.469	0.015
<u>Annealed for 100 hours at 1000°C after irradiation</u>					
Bulk density 2.90 g/cm ³	705	0.068	2.852	0.469	0.091
	707	0.068	2.852	0.469	0.091
	792	0.060	2.845	0.479	0.082
	896	0.051	2.835	0.489	0.071
	984	0.045	2.827	0.497	0.063

^aUnirradiated and irradiated specimens were obtained originally from the same extrusion mix and sinter and were of the same density.

^bSee Table 5.3 for conversion relationships.

^cIt is assumed that the specific heat is not changed by irradiation; measurements have established that there is no change (~2% detection limit) in irradiations at 1000°C to 10^{21} nvt ($E_n \geq 1$ Mev).

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6. FISSION PRODUCT TRANSPORT PROCESSES IN REFRACTORY-METAL FUEL SYSTEMS

(57070)

The purpose of this program is to study fission product transport processes in refractory-metal fuel systems at temperatures exceeding 1700°C in inert and reducing atmospheres, and to determine the effects of fission product impurity atoms on the mechanical properties of refractory metals.

As part of the overall evaluation of refractory metals for reactor applications, fission product migration and the effect of fission product impurities on the mechanical properties of refractory metals and their alloys are being investigated. The current work is divided into three major areas: (1) diffusion of rare gases (Kr⁸⁵) in refractory metals, (2) transport of fission products in refractory metals and (3) effect of fission product impurity atoms on mechanical properties of refractory metals.

DIFFUSION STUDIES

As stated previously,¹ the transport of krypton in polycrystalline tantalum at elevated temperatures (to 1730°C) exhibits only short range mobility (~1000Å), apparently along high diffusivity paths. No volume diffusion was detected. These data were obtained in short-time (15-to-30 minutes) experiments using a concurrent loading-diffusion glow discharge technique. Since a fuller understanding of the transport of the rare gases in various refractory metals and alloy systems is sought, information regarding the mechanism of krypton transport in tantalum (bulk and grain boundary diffusion coefficients, their structural correlation, and time and concentration effects) as well as experimental procedures for a submicron sectioning technique for other refractory metals and alloys needs to be determined.

Krypton in Tantalum

The lack of observable volume diffusion in tantalum may result from physical characteristics more favorable to grain boundary diffusion in (1) the temperature range employed, (2) the concentration of krypton relative to the concentration of lattice vacancies, and (3) the amount of grain and subgrain structure. The apparent short-range transport of krypton may result from the short time of the experiments or from the immobilization of krypton atoms at defect sites in the sample; it is important to demonstrate if long-range mobility can be obtained from samples subjected to longer times at temperature in the glow discharge environment to clarify this point. Work was continued on loading krypton into tantalum using system parameters more favorable for observing volume diffusion and using longer times for detecting longer range transport.

At higher sample temperatures and lower krypton concentrations, volume diffusion might be observable. Previously, the temperature limit for adequate loading of fine-

¹"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, p. 43.

grained arc-cast samples was about 1730°C. However, it was found that by lowering the krypton pressure, samples can be loaded to just measurable levels at 1830°C, and possibly at higher temperatures. The planned use of a source 25 times more concentrated in Kr⁸⁵ will facilitate the detection of krypton in the samples loaded at higher temperatures. The more concentrated Kr⁸⁵ source will also permit detectable loading at lower total krypton concentrations at lower temperatures. The present levels of loading could emphasize grain boundary migration since the concentration of krypton in the tantalum samples is about equal to or greater than the vacancy concentration ($\sim 10^{-4}$ mole fraction) at 1730°C.

The ratio of volume to grain boundary diffusion is also greatly affected by the structure of the sample. The greater the grain boundary, subgrain boundary, and dislocation density, the more likely grain boundary diffusion kinetics will predominate at a given temperature. The changes in structure caused by heat treatment are being examined. Samples were annealed for 1 hour at 1730°, 1830°, 1930°, 2030°, and 2130°C under dynamic vacuum (10^{-5} to 10^{-6} Torr), etched for various times, and specific areas photographed (100X) after each etching. The results obtained are to be used in conjunction with the development of a computer program for grain-boundary diffusion.

To reduce the amount of grain and subgrain boundaries and dislocations, both large-grained and single-crystal samples of tantalum are being prepared for loading in the glow discharge apparatus. Samples of arc-cast polycrystalline tantalum, 0.025- to 0.050-cm-thick are being heat-treated in dynamic vacuum at 2000° to 2100°C to stabilize the grain size. A 7.6-cm length of 1.0-cm-diameter triple-pass, zone-refined, single-crystal tantalum of 012 orientation was cut into plate specimens (~ 0.04 by 2.54 by 0.7 to 0.95 cm) and the individual plates electron-beam-welded to polycrystalline tabs.

The length of time at which samples can be held at temperature under glow discharge conditions to allow observation of long-range mobility has been limited by concurrent outgassing of the vacuum system which results in chemical contamination of the samples. The degree of contamination determines the thickness of the first section which can be removed and it is desirable that this section be 100 to 200 Å in thickness. To lessen outgassing, the vacuum system was overhauled, thoroughly cleaned and provided with a Cryosorb² liquid nitrogen trap. Run times were extended from 20 minutes to more than 200 minutes at a sample temperature of 1730°C. The first section of 200 Å thickness could not be completely removed during the mechanical stripping operation; however, it does appear that this will be possible with additional improvements in the outgas rate of the vacuum system.

Krypton in Other Refractory-Metal Systems

The tungsten, molybdenum, and W – 30Mo – 25Re systems are also being investigated in reference to the transport of krypton. A submicron sectioning procedure has been established for these materials (anodization in a bath of acetone saturated with nitrate ion) which must be calibrated in terms of the thickness of metal oxidized for a given current and time. The radiochemical and analytical techniques for calibration of the sectioning procedure are now being developed for molybdenum and W – 30Mo – 25Re alloy. Such a procedure, using an aqueous nitrate ion bath, is already known for tungsten.

High-purity, arc-cast tungsten was received and high-purity, arc-cast molybdenum was ordered for krypton transport experiments.

²Registered trademark of Granville-Phillips Co., Boulder, Colorado.

TRANSPORT OF FISSION PRODUCTS IN REFRACTORY METALS

Determination of the migration and release of selected fission products (other than the rare gases) in refractory metals during irradiation at temperatures up to 2000°C was continued in two phases: (1) determination of the fission product distribution in refractory metals used to contain fuel during irradiation at high temperatures and (2) determination of the extent of release of fission products from refractory-metal-canned fuel samples by measuring the amount deposited on the in-pile test assembly adjacent to the canned sample and in the exit gas line.

Determination of Fission Product Distribution

Tantalum cans of the same dimensions as those used to contain uranium fuel during test irradiations were neutron-activated and employed as control specimens during remote preparation, sectioning, and sampling procedures, to parallel conditions anticipated for sampling of the test fuel cans. The specimens, each fitted with an electrode, were mounted in plastic and the mounts ground and polished as metallographic samples. The initial grind and polish was made to dispose of excess plastic mounting material and to just expose the surface of the bottom plate of the cylindrical can to establish the zero sampling depth.

Subsequent grind and polish operations were made at successively greater depths into the bottom plates to expose fresh sampling surfaces for obtaining counting samples. The smallest increment of the plates which can be ground and polished away to expose fresh sampling surfaces is limited to about 100 microns due to the inability to measure more precisely than ± 12 microns with present techniques in the remote facility. This size increment is sufficient for observing the distribution profile of fission products across the approximately 1.1-mm-thick bottom plate of the irradiated fuel can although it negates the possibility of defining the approximately 4-micron-thick recoil region adjacent to the plate – fuel interface.

Counting samples were obtained by (1) anodization of the mechanically polished surface followed by mechanical stripping of the oxide film and (2) by hand-lapping and then collecting the debris in a drop of water. Anodization – mechanical stripping removed a thickness of 650 ± 100 Å of tantalum at 90 volts potential. Complete mechanical stripping of the oxide was not attained; thus, a comparison of the quantity of Ta^{182} in the oxide film removed to the known activation value of the base tantalum was required to establish actual sample size. Samples obtained by the hand-lapping method, using 400-grit alumina paper, were generally smaller than those from the anodization method. The hand-lap method will be used in the future as a backup to the anodization method to ensure that the anodization method does not selectively leach fission products.

Although considerable effort was exerted to maintain cleanliness during remote handling, contamination from handling of previously irradiated fuel was present in all samples obtained. Circumvention of this contamination is being attempted; a scheduled shutdown of the remote handling facility to permit alterations will present an unusual opportunity to do the work in a clean facility. Examination of both the tantalum and the W-Mo-Re fuel cans is scheduled for the period immediately following the shutdown.

Determination of Fission Product Release

Leaching of selected fission products from the internal surfaces of the hardware from the in-pile test cartridge used during the 2000°C, 1015-hour irradiation of a W-Mo-Re canned fuel sample was completed. The current data, obtained from the final 12.2 meters of off-gas ducting, are shown in Table 6.1 along with those data previously obtained from

TABLE 6.1

DISTRIBUTION OF FISSION PRODUCTS IN IN-PILE TEST CARTRIDGE^a WHICH
CONTAINED A W-Mo-Re ALLOY FUEL CONTAINER DURING IRRADIATION AT 2000°C

Isotope	Distribution, % of total recovery			Total Activity (D/M)	
	W-Mo-Re Can And Adjacent Area	Off-Gas Lines		Recovered Experimentally	Predicted From Rare Gas Measurements
		First 76.2 cm	Remaining 12.2 m ^b		
Ce ¹⁴⁴	14	77	9	1.7×10^5	$0.7-2 \times 10^5$
Ba ¹⁴⁰	66	34	— ^c	9.8×10^5	$5-10 \times 10^5$
Cs ¹³⁷	16	74	10	2.8×10^4	$0.3-0.8 \times 10^4$
I ¹³¹	8	92	<1	5.2×10^5	$4-10 \times 10^5$
Zr ⁹⁵	90	8	2	2.7×10^5	$3-10 \times 10^5$
Sr ⁸⁹	77	13	10	4.9×10^5	$3-10 \times 10^5$

^aIncludes off-gas ducting.

^bOff-gas lines leading from irradiation zone to rare-gas sampling station.

^cNo recovery due to lengthy decay time.

the high-temperature zone adjacent to the fuel can and the first 76.2 cm of exit ducting which contained a ceramic fiber particulate filter.

Considering the relative volatility of the listed fission products, and of their probable oxides and precursors, the apparent high mobility of cerium-144 is unaccountable. The low recovery of iodine-131 from the off-gas ducting is apparently due to adsorption on the particulate filter. With the exit ducting inventory included, the release of the solid fission products with the possible exception of cesium-137 is still no greater than was expected from recoil release from surface uranium contamination of the test specimen. As concluded previously,³ there is no indication that release by diffusion processes occurred for these fission products.

EFFECT OF FISSION PRODUCTS ON MECHANICAL PROPERTIES

To determine the effect of fission products on the mechanical properties of refractory metals and their alloys at high temperatures, creep measurements will be made on specimens impregnated with fission products at maximum in-pile temperatures of 1600° to 2200°C. The first irradiation cartridge, GEFP2-28, will provide 28 powder-metal-lurgy tungsten specimens for post-irradiation creep testing to determine whether creep is affected by the presence of fission product impurities, and the magnitude of the effect, if any. Creep tests will be run on three types of tungsten specimens: (1) unirradiated, (2) irradiated at various temperatures with no fission products present, and (3) irradiated at various temperatures with fission products recoiled and, at higher temperatures, diffused into the specimens. Succeeding cartridges will provide specimens of other refractory metals and alloys for creep testing.

During this report period, additional preparations for the first irradiation were made and assembly of the irradiation cartridge components was begun. Figure 6.1 illustrates one of the centerless-ground creep specimens which were received from the vendor. These were wound with 0.635-mm tungsten wire at each end of the reduced section of the specimens to serve as fiducial marks.

Final design of the high-temperature furnace and creep stand was completed and detail shop drawings begun. Delivery of the ultra-high vacuum system and the optical extensometer is scheduled for the next report period.

³"High-Temperature Materials Program Progress Report No. 50, Part A," GE-NMPO, GEMP-50A, August 20, 1965, p. 64.

Fig. 6.1 – Tungsten creep specimen with 0.635-mm-diameter tungsten wires as fiducial marks (Neg. P65-11-5A, $\sim 20\times$)

WORK PLANNED FOR NEXT PERIOD

Work on grain boundary and volume diffusion in polycrystalline and single-crystal tantalum will continue and the work on the tungsten system will commence. Calibration of the anodization – stripping process for molybdenum and W – 30Mo – 25 Re alloy will be completed.

Work will continue on the determination of the distribution profile of fission products in tantalum and W-Mo-Re fuel containers. Fabrication of components and assembly of the irradiation cartridge for tungsten creep specimens will be completed and assembly of the high-vacuum creep apparatus will begin.

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7. PHYSICO-CHEMICAL STUDIES OF CLAD UO_2 IN POTENTIAL MELTDOWN ENVIRONMENTS

(57075)

The objective of this program is to determine the behavior of Zircaloy- and stainless steel-clad UO_2 fuel elements in steam and steam plus air from 1200°C to the melting point of UO_2 and includes the measurement, where necessary, of the physical and mechanical properties of the claddings, the oxidized cladding, and UO_2 to their respective melting points.

REACTION MECHANISMS AND KINETICS

Zirconium-Base Alloy Reaction with Water Vapor (Steam)

Studies of the corrosion of Zircaloy were continued to explore its behavior in steam when (1) heated to the melting point at a temperature rise of either 0.3°C or 3°C per second and (2) isothermally tested at 1200°C and at 1400°C . The compositions of the Zircaloy-4 and Zircaloy-2 metal used in these tests as compared to the LOFT specifications for Zircaloy-4 are shown in Table 7.1.

Testing with Dynamic Heating - Tests were continued using the apparatus and sample configuration described previously.¹ The sample was altered slightly by reducing the width of a short center section of the sample so that the reaction would be restricted to the sighting range of the sight glass and the optical pyrometer. The sample material was provided by GE-APED and is reported to have a composition as shown for the Zircaloy-4 LOFT specifications given in Table 7.1.

Temperature measurements in both steam and argon atmospheres were checked with a Pt / Pt - 10Rh thermocouple attached to the back of the sample. Temperatures up to 1200°C agreed within the accuracy of the optical measurements. Above 1200°C the thermocouple was unreliable, probably as a result of alloying with the sample.

The sample breakage in steam atmosphere at about 1300°C which was noted previously² was eliminated by improving the attachment of the flexible electrodes. Sample failure by melting has been achieved in steam but, due to the localized nature of the failure and the averaging aspect of the optical temperature measurement, the indicated sample test temperature was only about 1700°C at the time of failure.

The tests in steam included runs at heating rates of both 0.3°C rise per second and 3°C rise per second. Although these data are tentative, about 65 percent of the metal remained after testing at 3°C per second as compared to about 35 percent remaining from the 0.3°C per second rate. These tests, of course, do not accurately reflect the most desirable simulation of the conditions expected when the fuel element undergoes a tem-

¹"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, pp. 47-48.

²Ibid., p. 47.

TABLE 7.1

LOFT SPECIFICATIONS FOR ZIRCALOY-4 COMPARED TO COMMERCIAL LOTS OF ZIRCALOY-4 AND ZIRCALOY-2

Element	LOFT Specifications ^a		Zircaloy-4 (Commercial Lot) ^b				Zircaloy-2 (Commercial Lot) ^b	
	Composition (Range), wt %	Maximum Impurities, ppm	Ingot Analyses, wt %		Ingot Impurities, ppm		Ingot Analyses, wt %	Ingot Impurities, ppm
			Top	Bottom	Top	Bottom		
Major Constituents								
Cr	0.005 – 0.15		0.09	0.10			0.091	
Fe	0.18 – 0.24		0.20	0.19			0.132	
Sn	1.2 – 1.7		1.45	1.46			1.49	
Ni	—		—	—			0.043	
Zr			Bal	Bal			Bal	
Impurities								
C		270			190	210		192
H		25			3.7	5		16
N		80			28	25		21
O		1400			1200	1200		945
Al		75			34	27		43
B		0.5			<0.2	<0.2		<0.3
Ca		—			—	—		—
Cd		0.5			<0.2	<0.2		<0.3
Co		15			<5	<5		<10
Cu		50			<10	<10		<20
Hf		6200			63	64		<125
Mg		20			<10	<10		<10
Mn		50			<10	<10		<25
Mo		50			<10	<10		<10
Na		20			—	—		<10
Nb		—			<100	<100		—
Ni		70			10	10		—
Pb		130			<5	<5		<20
Si		100			86	62		52
Ta		—			<200	<200		—
Ti		50			<15	<15		20
V		50			<5	<5		<10
W		50			<25	<25		<20
Zn		—			<50	<50		—

^aPreliminary specification F-6-LOFT, December 15, 1964.^bAnalyses provided by the vendor.

perature excursion after a loss of coolant accident since the sample is corroded on both sides and the type of failure is strongly influenced by the corrosion film thickness. An example of the appearance of the melted end of the corroded Zircaloy-4 sample heated at 3°C per second in a steam atmosphere is shown in Figure 7.1 (this type of melting behavior is to be compared to that shown for Type 304 stainless steel in Figure 7.6). Electron microprobe analysis showed that the metal network seen in the ZrO_{2-x} layer is a tin-rich alloy and this alloy is more concentrated near the oxide - metal interface.

Reaction Rate Measurements - Thermogravimetric studies of the reaction kinetics of steam with Zircaloy at fixed temperatures were continued. These studies have been on both ring-type samples and spherical samples. These two types of samples are used to determine if the appearance of a "breakaway" rate of corrosion, as reported previously,³ is dependent on testing a thin sheet of metal as suggested by Pemsler.⁴

The ring samples were made from Zircaloy-2 tubing. These rings were 1.25 cm ID and 0.75 cm long with a wall thickness of 0.08 cm. The spherical samples, 0.625 cm in

³Ibid., p. 50.⁴J. P. Pemsler, "Studies on the Oxygen Gradients in Oxidizing Metals, III. Kinetics of the Oxidation of Zirconium at High Temperatures," *Journal of Electrochemical Society*, Vol. 112, No. 5, 1965, pp. 477-484.

Fig. 7.1 – Photomicrograph showing failure portion of Zircaloy-4 heated at 3°C per second in steam (Neg. 6566, 75X)

diameter, were made from a Zircaloy-4 rod; the chemical analyses for both alloys are shown in Table 7.1. The results of these studies, shown in Figure 7.2 along with data from Lemmon⁵ and Bostram,⁶ indicate the same rate behavior at 1200°C and 1400°C.

About 80 percent of the ring-type specimen was converted to oxide after heating in steam for 25 minutes at 1400°C. Approximately 20 volume percent air was added during the last 5 minutes of the tests and no increases in reaction rate, the so-called "break-away" phenomenon, occurred as was previously noted at 1200°C when air was added to the steam.⁷ Although large cracks usually form in the oxide during corrosion, (see Figure 7.3c) the coatings remained essentially intact upon cooling, even when cooled from 1400°C which is above the transformation point of ZrO_2 .

Photomicrographs of cross sections of oxidized samples are shown in Figure 7.3. Figure 7.3a shows a ring sample heated at 1200°C for 34 minutes, 7.3b a spherical sample heated for 63 minutes at 1200°C, and 7.3c a spherical sample heated for 50 minutes at 1400°C. The large voids seen in all three photomicrographs probably resulted from polishing "pull-out." There are lines of metal particles in the oxide layer and parallel to the metal – oxide interface clearly shown in all of these microstructures. Electron microprobe analysis of these particles showed them to be tin or a tin-rich alloy. A secondary phase is shown in the central region of the metal. This phase was identified by electron microprobe analysis to be iron and chromium enriched.

A comparison of Zircaloy metal microstructures is shown in Figure 7.4. The original microstructure, Figure 7.4a, is to be compared to samples heated for about 30 minutes at 1200°C. The structure shown in Figure 7.4b is the typical β -zirconium structure after heating in a vacuum (1.6×10^{-5} Torr). Figure 7.4c shows the structure after heating in

⁵A. W. Lemmon, "Studies Relating to the Reaction Between Zirconium and Water at High Temperatures," Battelle Memorial Institute, BMI-1154, January 3, 1957.

⁶W. A. Bostram, "The High Temperature Oxidation of Zr-2 in H_2O ," Westinghouse-APD, WAPD 104, March 19, 1954.

⁷"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, p. 50.

Fig. 7.2—Steam corrosion of Zircaloy

a steam atmosphere. This latter structure is the oxygen containing α -modification as expected in agreement with the zirconium-oxygen phase equilibrium diagram.⁸

Metallographic examination of the ring of Zircaloy tested at 1400°C in steam showed varied effects as can be seen in Figure 7.5. As shown in Figure 7.5a, the lines of tin are especially clear on both sides of the metal in this area; while in Figure 7.5b, there is oxide separation or there are apparently three lines of tin on one side of the sample. Figure 7.5c illustrates the effect of one of the large cracks mentioned above showing the oxide layer protruding into the metal.

Type 304 Stainless Steel Reaction with Water Vapor (Steam)

Tests were continued on the behavior of Type 304 stainless steel heated at 0.3°C and 3°C per second in steam. Analyses of the metal used in this testing are shown in Table 7.2. The microstructure of samples after test are shown in Figures 7.6 and 7.7. It can be seen that the bloating behavior when heating at a rate of 0.3°C per second is much greater than that for a heating rate of 3°C per second. However, the sample heated at 0.3°C per second shows much greater oxide penetration in comparison with the one heat-

⁸R. F. Domagala and D. J. McPherson, "System Zirconium-Oxygen," *Journal of Metals*, Vol. 6, 1954, pp. 238-246.

a. Ring sample tested for 34 minutes at 1200°C (Neg. 6424)

b. Spherical sample tested for 63 minutes at 1200°C (Neg. 6471)

c. Spherical sample tested for 50 minutes at 1400°C (Neg. 6592)

Fig. 7.3—Photomicrographs of cross sections of Zircaloy samples corroded in steam (100X)

a. Original metal structure (Neg. 6726)

b. After 30 minutes at 1200°C in 1.6×10^{-5} Torr vacuum (Neg. 6727)

c. After 30 minutes at 1200°C in a steam atmosphere (Neg. 6729)

Fig. 7.4—Comparison of Zircaloy microstructures before and after thermal treatments in vacuum and in steam atmosphere (Polarized light, 250X)

Fig. 7.5—Photomicrographs of polished cross sections of a Zircaloy ring heated for 30 minutes at about 1400°C in flowing steam (75X)

a. Microstructure showing corrosion film (Neg. 6696, 250X)

b. Appearance of the melted end of the sample (Neg. 6694, 8X)

Fig. 7.6 – Type 304 stainless steel heated at 0.3°C per second in steam

a. Microstructure showing corrosion film (Neg. 6697, 250X)

b. Appearance of the melted end of the sample (Neg. 6695, 8X)

Fig. 7.7 – Type 304 stainless steel heated at 3°C per second in steam

TABLE 7.2
TYPE 304 STAINLESS STEEL COMPOSITIONS

Element	Tubing, ^a wt %	Sheet, ^b wt %
C	0.020	0.024
Cr	18.67	18.00
Co	0.26	0.05
Cu	0.26	0.28
Ni	9.42	10.95
Mn	1.22	1.69
Mo	0.30	0.16
P	0.022	0.024
S	0.012	0.011
Si	0.34	0.70

^aHardness of tubing ranged from R_B 65 to 71.

^bHardness of annealed sheet was R_B 72.

ed at 3°C per second. This oxide penetration for the latter sample is also much greater than that reported previously⁹ for the same test conditions. Oxidation in both stainless steel samples is considerably less than that described above for the Zircaloy.

WORK PLANNED FOR NEXT PERIOD

The power source for inductive heating of clad UO₂ pins has been installed, and the inductive heating arrangement will be developed and transient temperature steam corrosion tests started using this equipment.

Isothermal tests at different temperatures will be continued with emphasis on determining the behavior of Zircaloy where corrosion has been noted to depart from the parabolic rate.

Strength measurements of Zircaloy at temperatures above 700°C will be run using 0.38-cm-thick stock.

⁹"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, p. 50.

8. PHYSICO-CHEMICAL STUDIES OF Fe-Cr-Al-CLAD FUEL SYSTEMS

(57076)

The objective of this program is to define and to understand the reactions which can occur between oxidation-resistant Fe-Cr-Al alloys and UO_2 in the temperature region of 500° to 1200°C .

In early attempts to apply the Fe-Cr-Al alloys as claddings for UO_2 or metal - UO_2 cermet cores, an undesirable result was the appearance of uranium as surface contamination. The tentative explanation of this result is that reduction of UO_2 by Al produced U which diffused to the surface. The present program, aimed at quantitative understanding of the problem has been divided into three parts: phase studies, diffusion studies, and thermodynamic investigations.

PHASE STUDIES

The first point to be elucidated is the mechanism of uranium production. Available thermodynamic information indicates that the reaction of pure Al with UO_2 to give Al_2O_3 and free U is not favored, but any reliable prediction must take into account the actual chemical potentials of Al and U. To do this, it is necessary to examine the Fe-Cr-Al-U-O system for stable U-bearing phases. A thorough investigation of this type would require a major effort. A simplified approach, which is being tried first, is to form diffusion couples between representative Fe-Cr-Al alloys and U metal. If U compounds form, metallographic examination of heat-treated capsules should show bands of constant composition. If no compounds form, a continuous gradient of U concentration should be observed.

The first nine representative iron base alloys contain 5, 15, and 25 weight percent Cr, and at each Cr level the Al contents are 4, 7, and 10 percent.

Capsules of Fe - 15Cr - 4Al-clad uranium, whose fabrication has been previously described,¹ were heat treated at 600° , 800° , 1000° , and 1200°C in air. The results of the preliminary metallographic examination are summarized in Table 8.1. Specimens held at 600°C for times up to 750 hours were almost completely devoid of any core - cladding interaction; whereas, at temperatures above the melting point of the Fe-U eutectic (725°C), extensive reaction and compound formation occurred. In the as-fabricated capsules, the interface between the U core and the Fe-Cr-Al cladding was a 0.318-cm-diameter cylindrical surface. The diameter of the interfacial surface between Fe-Cr-Al and the U-rich core increased by 10 percent in tests at 800°C and by 25 and 30 percent at 1000°C . The reactions taking place between core and cladding appear to be fairly uniform and to be influenced more by temperature than by extended time. At 1200°C (above the melting point of uranium), reaction occurs very rapidly; after 49 hours the diameter of the interfacial surface increased 23 percent and the reaction zone was fairly uniform. After 80 hours at 1200°C , the U-bearing core material completely penetrated the 0.478-cm-thick cladding wall. The final stage of cladding penetration appears to

¹"High-Temperature Materials Program Progress Report No. 52, Part A," GE-NMPO, GEMP-52A, October 20, 1965, p. 54.

progress by movement along the grain boundaries and leads to isolation of large sectors of unattacked Fe-Cr-Al cladding. Identification of the phases present in these samples will be accomplished by electron microprobe examination.

TABLE 8.1
RESULTS OF HEAT-TREATING Fe - 15Cr - 4Al-ENCAPSULATED
U-METAL CORES IN AIR

Temperature, °C	Time At Temperature, hr	Increase In Core Diameter, %	Core - Cladding Reaction
600	300	0.0	Trace at interface
600	750	0.0	Trace at interface
800	150	10.2	Multiphase core area
800	1000	9.6	Multiphase core area
1000	150	24.7	Multiphase core area
1000	500	29.5	Multiphase core area
1200	49	22.5	Multiphase core area
1200	80	>50.0	Complete grain boundary penetration of cladding

A survey of the low-uranium portion of the Fe-Cr-Al-U phase space was started. Alloys of Fe - 25Cr - 10Al and Fe - 5Cr - 4Al, each plus 1 percent and 5 percent U, were prepared. Samples of each of these four alloys will be heat-treated at various temperatures, quenched, and examined metallographically. These samples should show either a single-phase Fe-Cr-Al-U solid solution (unsaturated in U) or various compounds (such as UFe_2) plus an Fe-Cr-Al-U solid solution of the composition representing U saturation at the soaking temperature. Microprobe analysis of solid solutions of the latter type should give the desired limiting solubility of U as a function of temperature and composition.

DIFFUSION STUDIES

Results from the phase studies indicate that Fe-Cr-Al versus U couples will not be convenient for measurement of diffusion coefficients of U. After the solubility limit of U in Fe-Cr-Al alloys is determined as outlined in the previous section, couples of the type Fe-Cr-Al-U (single phase) versus Fe-Cr-Al will be used in determining diffusion coefficients of U.

Another problem to be investigated is the possibility that diffusion in these alloys may depend on the gradient of chemical potential rather than on the gradient of concentration. Fe-Al and Fe-Cr alloys were prepared, and diffusion couples of the type Fe - 10Al versus Fe - 25Cr - 10Al are being studied to determine, for example, if there is preferential migration leading to a discontinuity of Al concentration at the couple interface. These tests are to be followed by examination of Fe-Cr-Al versus Fe couples to determine the rate of influx of Al to the zone of reduction of UO_2 .

THERMODYNAMIC INVESTIGATIONS

Measurement of the emf of electrochemical cells with molten salt electrolytes was chosen as the most promising method for determining the chemical potential of components of alloy solutions. Apparatus similar to that described by Radcliffe, Averbach, and Cohen² is being assembled. The first experiments will consist of repeating some of the measurements of these workers on Fe-Al alloys to assure that the method is reproducible.

WORK PLANNED FOR NEXT PERIOD

Testing of additional diffusion couples and identification of phases formed in tested couples will be continued.

Assembly of equipment to measure the emf of cells with molten salt electrolytes will continue.

²S. V. Radcliffe, B. L. Averbach, and M. Cohen, "Relative Thermodynamic Properties of Solid Iron-Aluminum Alloys," *Acta Metallurgica*, Vol. 9, March 1961, pp. 169-176.

9. APPENDIX

The "High-Temperature Materials Program Progress Reports" previously issued in this series are listed below. The first two reports were each issued as one document, containing both the classified and unclassified portions. The subsequent reports were issued as two documents; part A, the unclassified portion and part B, the classified portion.

<u>Report No.</u>	<u>Report Period</u>	<u>Publication Date</u>
GEMP-1	May 1961 - June 30, 1961	July 15, 1961
GEMP-2	July 1, 1961 - July 31, 1961	August 15, 1961
GEMP-3, A and B	July 1, 1961 - August 31, 1961	September 15, 1961
GEMP-4, A and B	August 1, 1961 - September 30, 1961	October 15, 1961
GEMP-5, A and B	August 15, 1961 - October 15, 1961	November 15, 1961
GEMP-6, A and B	September 15, 1961 - November 15, 1961	December 15, 1961
GEMP-7, A and B	October 15, 1961 - December 15, 1961	January 15, 1962
GEMP-106, A and B (First Annual Report)	Calendar Year 1961	February 28, 1962
GEMP-9, A and B	January 1, 1962 - February 15, 1962	March 30, 1962
GEMP-10, A and B	January 1, 1962 - March 15, 1962	April 16, 1962
GEMP-11, A and B	February 15, 1962 - April 15, 1962	May 15, 1962
GEMP-12, A and B	March 15, 1962 - May 15, 1962	June 15, 1962
GEMP-13, A and B	April 15, 1962 - June 15, 1962	July 31, 1962
GEMP-14, A and B	May 15, 1962 - July 15, 1962	August 15, 1962
GEMP-15, A and B	June 15, 1962 - August 15, 1962	September 15, 1962
GEMP-16, A and B	July 15, 1962 - September 15, 1962	October 15, 1962
GEMP-17, A and B	August 15, 1962 - October 15, 1962	November 15, 1962
GEMP-18, A and B	September 15, 1962 - November 15, 1962	December 14, 1962
GEMP-19, A and B	October 15, 1962 - December 15, 1962	January 25, 1963
GEMP-177, A and B (Second Annual Report)	Calendar Year 1962	February 28, 1963
GEMP-21, A and B	January 1, 1963 - February 15, 1963	April 23, 1963
GEMP-22, A and B	January 1, 1963 - March 15, 1963	April 30, 1963
GEMP-23, A and B	February 15, 1963 - April 15, 1963	May 31, 1963
GEMP-24, A and B	March 15, 1963 - May 15, 1963	June 28, 1963
GEMP-25, A and B	April 15, 1963 - June 15, 1963	July 31, 1963
GEMP-26, A and B	May 15, 1963 - July 15, 1963	August 16, 1963
GEMP-27, A and B	June 15, 1963 - August 15, 1963	September 30, 1963
GEMP-28, A and B	July 15, 1963 - September 15, 1963	November 11, 1963
GEMP-29, A and B	August 15, 1963 - October 15, 1963	November 29, 1963
GEMP-30, A and B	September 15, 1963 - November 15, 1963	December 31, 1963
GEMP-31, A and B	October 15, 1963 - December 15, 1963	January 24, 1964
GEMP-270, A and B (Third Annual Report)	January 1, 1963 - January 31, 1964	February 28, 1964

<u>Report No.</u>	<u>Report Period</u>	<u>Publication Date</u>
GEMP-34, A and B	February 1, 1964 - March 15, 1964	April 15, 1964
GEMP-35, A and B	February 1, 1964 - April 15, 1964	May 28, 1964
GEMP-36, A and B	March 15, 1964 - May 15, 1964	June 19, 1964
GEMP-37, A and B	April 15, 1964 - June 15, 1964	July 31, 1964
GEMP-38, A and B	May 15, 1964 - July 15, 1964	August 19, 1964
GEMP-39, A and B	June 15, 1964 - August 15, 1964	September 30, 1964
GEMP-40, A and B	July 15, 1964 - September 15, 1964	October 15, 1964
GEMP-41, A and B	August 15, 1964 - October 15, 1964	November 30, 1964
GEMP-42, A and B	September 15, 1964 - November 15, 1964	December 18, 1964
GEMP-43, A and B	October 15, 1964 - December 15, 1964	January 29, 1965
GEMP-334, A and B (Fourth Annual Report)	January 31, 1964 - January 31, 1965	February 26, 1965
GEMP-46, A and B	February 1, 1965 - March 15, 1965	April 15, 1965
GEMP-47, A and B	February 1, 1965 - April 15, 1965	May 28, 1965
GEMP-48, A and B	March 15, 1965 - May 15, 1965	June 25, 1965
GEMP-49, A and B	April 15, 1965 - June 15, 1965	July 28, 1965
GEMP-50, A and B	May 15, 1965 - July 15, 1965	August 20, 1965
GEMP-51, A and B	June 15, 1965 - August 15, 1965	September 30, 1965
GEMP-52, A and B	July 15, 1965 - September 15, 1965	October 20, 1965
GEMP-53, A and B	August 15, 1965 - October 15, 1965	November 26, 1965

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