BNWL-1349-1
UC-25

4-
4-70

QUARTERLY PROGRESS REPORT
JANUARY, FEBRUARY, MARCH, 1970
REACTOR FUELS AND MATERIALS
DEVELOPMENT PROGRAMS
FOR FUELS AND MATERIALS BRANCH
OF
USAEC DIVISION
OF REACTOR DEVELOPMENT AND TECHNOLOGY
April 1970

AEC RESEARCH & DEVELOPMENT REPORT

REVISION	DATE	BY	FILED ROUTE (DATE)

BNWL-1349-1

BATTELLE

BATTELLE MEMORIAL INSTITUTE

NORTHWEST

PACIFIC NORTHWEST LABORATORIES

BATTELLE BOULEVARD, P. O. BOX 999, RICHLAND, WASHINGTON 99352

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

3 3679 00061 9231

BNWL-1349-1

UC-25, Metals,
Ceramics and Materials

QUARTERLY PROGRESS REPORT
JANUARY, FEBRUARY, MARCH, 1970
REACTOR FUELS AND MATERIALS DEVELOPMENT PROGRAMS
FOR
FUELS AND MATERIALS BRANCH
OF
USAEC DIVISION OF REACTOR DEVELOPMENT AND TECHNOLOGY

Edited by
W. F. Sheely

April 1970

FIRST UNRESTRICTED
DISTRIBUTION MADE

MAY 27 '70

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

PREVIOUS QUARTERLY REPORTS

<u>Quarter</u>	<u>Year</u>	<u>Report Number</u>
	<u>1966</u>	
First		BNWL-CC-694
Second		BNWL-CC-761
Third		BNWL-CC-821
Fourth		BNWL-CC-957
	<u>1967</u>	
First		BNWL-435
Second		BNWL-473
Third		BNWL-658
Fourth		BNWL-668
	<u>1968</u>	
First		BNWL-768
Second		BNWL-842
Third		BNWL-919
Fourth		BNWL-971
	<u>1969</u>	
First		BNWL-1058
Second		BNWL-1144
Third		BNWL-1223
Fourth		BNWL-1279

CONTENTS

SUMMARY	1.1
FAST-REACTOR CERAMIC FUELS RESEARCH	2.1
High Temperature Creep of Unirradiated Fast-Reactor Oxide Fuels - O. D. Slagle.	2.1
In-Reactor Creep of Mixed Oxide Fuels - J. A. Basmajian	2.7
Thermal Conductivity of (Pu,U)O ₂ - R. L. Gibby	2.9
Properties of Molten Fast-Reactor Oxide Fuels - R. P. Nelson, J. J. Rasmussen and O. D. Slagle.	2.19
Hot-Pressing of Fast-Reactor Ceramic Oxides - P. E. Hart.	2.25
Lattice Expansion of UO ₂ -PuO ₂ - P. G. Pallmer	2.27
Phase Studies - C. E. McNeilly.	2.34
Determination of Oxygen/Metal Ratios for Oxide Fuels - R. E. Woodley	2.35
IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS	
J. A. Christensen	3.1
In-Reactor Thermal Performance of UO ₂ -PuO ₂ Fuel J. A. Christensen	3.1
Fuel-Clad Gap Conductance Measurements - L. A. Lawrence, J. A. Christensen and G. R. Horn.	3.1
Molybdenum Distribution in Irradiated Oxide Fuels - L. A. Lawrence and J. A. Christensen.	3.8
Fission Product Migration in Oxide Fuels - L. A. Lawrence and J. A. Christensen.	3.11
Effects of Burnup on Melting Heat Rating - C. R. Hann	3.14
Evaluation of Alternate Irradiation Facilities - C. R. Hann and L. A. Lawrence	3.20
Fast Reactor Experiments - C. R. Hann	3.21
Fuel-to-Cladding Gap Conductivity Analyses - G. R. Horn G. R. Horn.	3.21
Irradiation of Hypostoichiometric UO ₂ -PuO ₂ Fuel - J. E. Hanson	3.27
High Burnup Thermal Flux Irradiations - R. D. Leggett, J. W. Weber, L. A. Pember, D. Christensen, E. D. Jenson and D. C. Bullington	3.27

Fuel Swelling and Analytical Modeling - K. R. Merckx and J. A. Christensen	3.50
Material Characterization and Swelling Model Evaluation - K. R. Merckx, J. L. Daniel, S. J. Mayhan, W. L. Nicholson and H. A. Triebs	3.50
In-Reactor Swelling Measurements - F. E. Panisko.	3.58
Migration Behavior in Mixed Oxides - C. A. Hinman	3.62
IRRADIATION DAMAGE TO REACTOR METALS.	4.1
Irradiation Behavior of Structural Materials - R. P. Marshall.	4.1
Irradiation Facilities Operation - A. L. Ward	4.1
In-Reactor Measurements - E. R. Gilbert and N. E. Harding	4.1
Fracture Analysis - C. L. Hellerich	4.9
The Stability of Voids in Neutron-Irradiated Nickel - G. L. Kulcinski, B. Mastel, H. E. Kissinger and T. K. Bierlein	4.23
Grain Boundary Stabilization - G. L. Guthrie and T. K. Bierlein	4.35
Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels - C. W. Hunter and J. A. Williams	4.37
Fuel Subassembly Materials - T. T. Claudson	4.55
EBR-II Irradiations - P. K. McDaniel.	4.55
Damage Analysis - J. L. Straalsund, J. F. Bates and H. R. Brager.	4.56
Mechanical Testing of Fuel Pin Cladding - R. L. Fish	4.67
Fast Reactor Supporting Studies	4.72
Acceptance Testing and Clad Characterization - M. M. Paxton.	4.72
Tensile Studies of Types 304 and 316 Stainless Steels - A. L. Ward	4.73
Uniaxial Creep-Rupture Studies of Types 304 and 316 Stainless Steel - A. J. Lovell	4.82
Weldment Studies - A. L. Ward and A. J. Lovell	4.83

FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS . . .	5.1
Damage Functions and Data Correlation - W. N. McElroy, R. L. Simons, and L. D. Blackburn . . .	5.1
SAND-II Solution Accuracy Study for EBR-II Spectra - R. L. Simons and W. N. McElroy . . .	5.5
Fast Reactor Materials Dosimetry Center - J. A. Ulseth and J. L. Jackson . . .	5.11
Computer Simulation of Radiation Damage - D. G. Doran . . .	5.15
Computer Simulation of Irradiation Processes - G. E. Russcher . . .	5.23
Computer Tabulation of Dounreay Fast Reactor Flux and Reaction Rate Data - R. E. Dahl, Jr., R. D. Bourquin, and N. J. Graves . .	5.23
Computer Simulation Studies of Dislocation Core Structure - R. E. Dahl, Jr., and R. D. Bourquin . . .	5.24
Fluence Determinations Using Time-Varying Flux - R. D. Bourquin . . .	5.28
FAST REACTOR ABSORBER MATERIALS - G. L. Tingey . .	6.1
Radiation Effects on Boron Carbide and Tantalum - A. L. Pitner and D. E. Baker . . .	6.1
NUCLEAR GRAPHITE - G. L. Tingey . . .	7.1
Dimensional Changes of Graphites at High Neutron Fluences and High Temperatures - A. L. Pitner and W. J. Gray . . .	7.1
Computer Calculations of Graphite Oxidation in HTGR - R. C. Giberson . . .	7.5
Radiolysis of H ₂ -CO ₂ -He Mixtures - D. E. Clark and G. L. Tingey . . .	7.7
NONDESTRUCTIVE TESTING . . .	8.1
Eddy Current Methods - H. L. Libby . . .	8.1
Ultrasonic Methods . . .	8.8
Phase Measurements of Reflected Ultrasonic Waves Near the Rayleigh Critical Angle - F. L. Becker . .	8.8
Ultrasonic Measurement of Cold Work - R. L. Trantow . . .	8.15
Flaw Identification and Characterization - N. E. Dixon . . .	8.19

Thermal Methods - D. R. Green	8.28
New Techniques	8.35
Holographic Methods - F. R. Reich	8.35
ART WATER LOOP OPERATION AND MAINTENANCE - R. S. Hope	9.1

QUARTERLY PROGRESS REPORT
JANUARY, FEBRUARY, MARCH
REACTOR FUELS AND MATERIALS DEVELOPMENT PROGRAMS
FOR
FUELS AND MATERIALS BRANCH
OF
USAEC DIVISION OF REACTOR DEVELOPMENT AND TECHNOLOGY

Edited by
W. F. Sheely

1.0 SUMMARY

(2.0) FAST-REACTOR CERAMIC FUELS RESEARCH

High Temperature Creep of Unirradiated Fast-Reactor Oxide Fuels

A study was undertaken to determine if the creep rates of UO_2 in bending exhibit the same nonreproducible behavior as has been observed in UO_2 -20 wt% PuO_2 . Although a similar behavior was observed, no single cause could be determined for the effect.

An analysis has been carried out to determine the effect of a nonlinear stress-dependent strain rate on the evaluation of stresses and strains in four-point bending.

In-Reactor Creep of Mixed Oxide Fuels

The second test capsule has been irradiated in the K-West Reactor for 80 hr. The problems encountered with the first capsule were eliminated by design modifications. One of the two test specimens broke 1 hr after irradiation began, and the other broke at some undetermined time. Breakage appeared to be related to microcracks in the sample.

Thermal Conductivity of $(\text{Pu},\text{U})\text{O}_2$

The thermal conductivities of stoichiometric UO_2 - PuO_2 solid solutions containing 0, 5, 12, 20, 25, and 30 wt% PuO_2

were calculated from measured thermal diffusivity values. Additions of PuO_2 to the UO_2 lattice resulted in a slight but measurable decrease in thermal conductivity at all temperatures within the range investigated (100 to 1200 °C).

Properties of Molten Fast-Reactor Oxide Fuels

Measurements Facility

The fluoroscopic observation of the molten UO_2 - PuO_2 fuels has been improved by the installation of an X-ray image intensifier.

Density

A gas pycnometer technique and a gamma attenuation technique are being evaluated as methods to improve the accuracy of measurement of the volume expansion on melting and liquid density of UO_2 - PuO_2 fuels.

Surface Tension

A new furnace has been constructed to measure surface tension of UO_2 - PuO_2 fuels by the pendant drop technique. The precision of surface profile techniques for surface tension measurement is limited to $\pm 10\%$ by the technique used to mechanically measure the profile.

Hot-Pressing of Fast-Reactor Ceramic Oxides

High density $(\text{U}_{0.75}\text{Pu}_{0.25})\text{O}_2$ fuels have been fabricated by hot pressing. Specimens are being evaluated for physical property measurements.

Lattice Expansion of UO_2 - PuO_2

A lattice thermal expansion of $9.8 \times 10^{-6}/^\circ\text{C}$ has been measured for $(\text{U}_{0.75}\text{Pu}_{0.25})\text{O}_2$ to 1600 °C. Temperatures were determined from measurement of the lattice expansion of molybdenum powder additions.

Phase Studies

The present micro-thermobalance apparatus is being modified to extend the temperature range for measurements of O/M ratios of mixed oxides as a function of oxygen potential.

Determination of Oxygen/Metal Ratios for Oxide Fuels

The existence of CO₂ sources other than that arising from the carbon reduction of the oxide fuel sample has continued to plague the development of a method of O/M determination on milligram quantities of oxide fuels. It may prove necessary to eliminate any exposure of the powdered graphite employed as a fuel reductant to gases which are subsequently released as oxides of carbon.

(3.0) IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS

In-Reactor Thermal Performance of UO₂-PuO₂ Fuel

Irradiation of eight UO₂-25 wt% PuO₂ fuel pins with variable fuel clad gaps and Xe/He ratios in the fill gas was completed. Initial postirradiation examination showed substantial gap closure for nominal FTR gap and peak heat rating conditions during a 100-hr test. Fuel with a nominal FTR gap operated with significantly higher fuel temperatures than did fuel with a minimum initial gap. Changing Xe/He progressively from 0 to 1 caused a commensurate and consistent increase in fuel temperature for both large- and small-gap pins.

Fission product molybdenum, zirconium, cerium, and ruthenium distributions were measured in UO₂-25 wt% PuO₂ fuels with various time and heat rating histories. Studies of high, fast-flux burnup effects on UO₂-25 wt% PuO₂ fuel melting heat rating were begun.

A survey and analysis of existing fuel-clad gap conductance data clearly showed that present knowledge is not adequate for fuel design purposes.

Irradiation of Hypostoichiometric UO_2 - PuO_2 Fuel

Extensive precipitation of fission-product-metal inclusions and nuggets was observed in UO_2 -25 wt% PuO_2 fuels irradiated to 75,000 to 100,000 MWd/MTM. Migration of plutonium toward the fuel center was also detected. Compositions of a number of metal and oxide fission product inclusions was studied with the shielded microprobe. Radial distributions of molybdenum, ruthenium, neodymium, cerium, zirconium, and xenon were measured. Iron-, nickel-, and, in some cases, chromium-bearing inclusions in the fuel were interpreted in terms of fuel-clad-fission product chemical interactions. The clad was found to contain both fuel and manganese inclusions.

Terminal gamma scans of fuel pins previously scanned at midlife showed gross redistribution of central, fission-product nuggets.

Fuel Swelling and Analytical Modeling

Scanning electron microscope examination of replicas from several EBR-II irradiated fuel pins is in progress. The same instrument is being used to characterize oxide starting materials. Results are being used to quantitatively evaluate fission gas bubble migration models. Model development was focused on characterizing size and distribution of fission gas bubbles trapped on grain boundaries.

Fabrication of fuel and components for the first in-reactor basic oxide swelling experiment was completed. The first assembly, which will contain 27 specimens with various fissile atom densities and geometries, will be irradiated in April to evaluate analytical predictions of specimen temperatures and fission rates.

Migration Behavior in Mixed Oxides

The major thermal-gradient-induced forces responsible for oxygen migration in fuels are being investigated theoretically and experimentally. These are the electronic component, characterized by the electronic thermo-electric power (electronic heat of transport) and the ionic component, characterized by the ionic heat of transport.

(4.0) IRRADIATION DAMAGE TO REACTOR METALS

Irradiation Facilities Operation

Operation of the ETR G-7 pressurized hot water loop has been discontinued. The pressure tube was removed in November 1969. A total of 402 corrosion, tensile, and creep-rupture specimens were discharged from the loop at the conclusion of cycles 103 and 104.

In-Reactor Measurements

In-reactor creep measurements in a Hanford K Reactor are compared with published data. Irradiation-induced creep strain data can be described as the sum of a fluence-dependent transient strain and a steady-state creep term. The irradiation-induced transient strain is larger in 20% cold-worked Type 316 SS than in solution treated Type 304 and 316 SS. Irradiation-induced creep decreases with increasing temperatures over the range of 43 to 370 °C. The greater amount of irradiation-induced creep obtained in the tensile test compared to the torsion test is attributed to the influence of the hydrostatic pressure component of the tensile stress on swelling.

Fracture Analysis

The results of the fractographic examination of Type 316 SS creep-rupture specimens are reported. Determinations are given for the effects of aging temperature, test

temperature, and irradiation on the fracture behavior (fracture mode and mechanism). Correlation between fracture mode and ductility are established and microstructural examination reveals the presence of a platelet form of precipitate at grain boundary microcracks. Increased crack propagation rates in irradiated material (potential flaw sensitivity) are demonstrated fractographically.

The Stability of Voids in Neutron Irradiated Nickel

Some additional information supplementary to that reported previously has been derived from experiments on the characteristics and stability of voids in nickel during postirradiation annealing treatments. The applicability was assessed of several small angle X-ray scattering methods for determining the distribution of void sizes in nickel irradiated to 3×10^{21} n/cm² ($E > 0.1$ MeV) at 450 °C as a function of annealing. The stability of the voids during annealing is attributed in part to the presence of some gas atoms in the voids.

Grain Boundary Stabilization

Grain size stabilization of small grain nickel powder compacts (~ 10 μ grain size) has been achieved in ex-reactor annealing studies. Stabilization accomplished by a technique using helium-atom pinning of boundaries required a volume concentration of helium of only 0.3 ppm.

The effect of variation of injection parameters on the quantity and binding depth of the trapped gas has been investigated.

Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels

Planning, engineering, and procurement has been completed for the installations of electrical heaters in the ETR M-3 loop to provide greater temperature flexibility during specimen

irradiation. Postirradiation annealing of A533-B pressure vessel steel specimens revealed that the reduction of irradiation hardening commenced above 540 °F and was 70% complete by 650 °F. A fluence increase from 2 to 4.4×10^{19} n/cm² (E > 1 MeV) produced only a modest increase in strength. The fatigue crack behavior indicated the propensity for crack propagation hindrance.

EBR-II Irradiations

Pins BNW B41 and B42 are being consolidated with BNW Subassembly 5. The fourth in a series of five swelling experiments (Pin BNW 25) is being assembled for placement in EBR-II at the beginning of Run 41. A revised summary of current irradiation is presented.

Damage Analysis

Density measurements were made to determine the extent of irradiation swelling in a pin which was used to contain tensile specimens in Row 7 in the EBR-II. Both the total amount of swelling and the gradient in swelling along the axial length of the pin were less than that predicted by the most recent empirical swelling equation.

Empirical equations are presently being developed to describe the effects of neutron exposure and irradiation temperature on void size and concentration in Types 304 and 316 SS.

Mechanical Testing of Fuel Pin Cladding

Previously reported burst data on PNL-1 series fuel pins revealed a very low strength and ductility region above the reactor midplane. The low strength is thought to be due to grain boundary attack during testing of sensitized cladding.

Additional fueled cladding specimens from above the midplane of PNL 1-16 and 1-18 have revealed intergranular cracks

during room temperature leak testing. The orientation of these cracks suggests that the pins were subjected to a bending stress during storage.

Electron microprobe results, ring test results, and cellulose acetate replication findings are presented, all of which help narrow down the possible causes for the observed behavior.

Acceptance Testing and Clad Characterization

The characterization of PNL tube lots E, F, G, and H is essentially complete except for long-term stress-rupture data, and testing is continuing on PNL tube lots N-1 and N-2. A data handbook for each lot of tubing is now being assembled.

Tensile Studies of Types 304 and 316 Stainless Steels

Tensile test results from AISI Type 316 SS specimens irradiated in Row 2 of the EBR-II have been combined with previously reported data from the EBR-II 5C3 control rod thimble (Type 304), in order to clarify the roles of helium embrittlement and displacement damage in elevated temperature ductility losses. These materials represent helium concentrations in the range from 1 to 16 ppm (atomic). The study has shown that post-irradiation annealing at 2000 °F for 1 hr produces nearly complete restoration of the pre-irradiation tensile ductility at test temperatures below $T_m/2$, but only partial recovery at higher temperatures.

Uniaxial Creep-Rupture Studies of Types 304 and 316 Stainless Steels

A comparison of Types 304, 316, and 348 SS shows that the creep-rupture properties of the three alloys at 1000 °F are similar after irradiation to 1.2×10^{22} n/cm² (total) at ~900 °F, even though the alloys exhibited quite different properties prior to irradiation.

Weldment Studies

Results are reported for all-weld specimens representing the four weld processes included in the FTR vessel weldment program. These data show that tensile properties are essentially unaffected by irradiation to 1×10^{21} n/cm² (total) at 700 and 800 °F, while substantial strengthening and ductility loss are observed following irradiation to 3.8 and 4.4×10^{21} n/cm² (total) at 800 °F, except in the case of the stick-electrode weld where ductility is apparently unaffected by the higher fluence irradiation.

Tensile test results are also reported for weldment and base metal specimens of a modified design. The study was to determine whether a larger diameter (0.250-in. gage diameter) would provide an advantage over the 0.125-in. gage diameter miniature specimen used in the vessel weldment irradiation experiment.

Creep-rupture test results for all-weld specimens from two of the weld processes are reported. These results show that the submerged arc weldment and the TIG weldment respond differently to an irradiation to 8×10^{21} n/cm² (total) at 1100 °F.

(5.0) FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS

Damage Functions and Data Correlation

A preliminary form of a damage function has been obtained for a change in yield strength to 60 ksi in Type 304 SS irradiated and tested at ~450 °C. The derived damage function is used to evaluate a parameter in an equation that relates the measured property change to total fluence. This allows the application of available test reactor data to other spectral environments of interest in the development of Liquid Metal Fast Breeder Reactors.

SAND-II Solution Accuracy Study for EBR-II Spectra

A study has been initiated to more precisely define the accuracy of integral fluxes determined by the SAND-II code for the EBR-II run 31F dosimetry test. Accuracy limits due to uncertainties in cross sections and reaction rate measurements will be defined by a computerized error propagation technique. From the initial steps in the study, uncertainties due to lack of solution uniqueness are estimated to be no more than about 4% (1σ) in the core and bordering areas and about 25% (1σ) in the blanket regions.

Fast Reactor Materials Dosimetry Center

The Fast Reactor Materials Dosimetry Center data file has been expanded by the addition of unfolded neutron spectra. Axial flux distribution for any particular radius above or below any specified energy, among other data, can now be obtained in computer plotted or tabulated form by request. Aside from data applicable to past experimentation, the center can also provide data estimates applicable to future EBR-II irradiations.

Computer Simulation of Radiation Damage

The overlap of 20 keV displacement cascades in α -Fe was simulated with the CASCADE-CLUSTER and HAP-4 computer codes. The result was a noticeable increase in both maximum and average cluster sizes. A parameter study with HAP-4 indicates that, for short term annealing simulations, a 112-site isotropic recombination region may be equivalent to the 62-site anisotropic region presently employed.

Computer Tabulation of Dounreay Fast Reactor Flux and Reaction Rate Data

DFR flux and reaction rate data have been incorporated into a computer library and analysis program. Highly detailed

flux and linear heat generation profiles can now be obtained for any pin or subassembly irradiated within the DFR core or blanket.

Computer Simulation Studies of Dislocation Core Structure

Dislocation structures have been modeled using the GRAINS program. This modeling is a necessary step in determining the parameters and mechanisms for creep and swelling in structural materials.

Fluence Determinations Using Time-Varying Flux

A computer program, TIMH, has been written to use reactor power history data to determine flux and fluence by foil activation analysis. This means of flux and fluence calculation avoids excessively large errors by accounting for the effects of reactor cycling.

(6.0) FAST REACTOR ABSORBER MATERIALS

Irradiation in a Thermal Flux

Irradiation experiments have been completed on B_4C in a thermal flux. B_4C powders and 65, 80 and 99% TD pellets were irradiated to a maximum exposure of 15×10^{20} captures/cm³ at temperatures ranging from 500 to 1200 °F. Gas release fractions increased with temperature from about 10% at 500 °F to 25% at 1200 °F.

Fast Reactor Irradiation

Postirradiation examination of samples in the tantalum irradiation capsule, TA-5, has been completed. Annealing of the silicon carbide temperature monitor in the capsule indicated a maximum temperature of 2100 °F, considerably higher than the calculated temperature of 1650 °F. Swelling measurements show density changes of ~0.5% for a sample irradiated at 2100 °F to an exposure of 8.6×10^{21} nvt ($E > 0.1$ MeV) and a change of 0.8% for a sample irradiated at 1300 °F to an exposure of 3×10^{21} .

(7.0) NUCLEAR GRAPHITE

Dimensional Changes of Graphites at High Neutron Fluences and High Temperatures

Irradiation data have been obtained on a variety of conventional and experimental graphites at temperatures up to 1400 °C and fluences up to 1.4×10^{22} n/cm² (E > 0.18 MeV). Post-turnaround expansion of at least some graphites is less rapid at temperatures above 1200 °C than at temperatures of 900 to 1200 °C. Graphites manufactured by POCO Graphite Inc. exhibit smaller dimensional changes and less rapid expansion at high fluences than do conventional graphites.

Computer Calculations of Graphite Oxidation in HTGR

A steady state of H₂/H₂O ratio of 28.3 is calculated for normal operation of the 1100 MW_e HTGR based on current estimates of the leak rate and purification factor as obtained from Gulf General Atomic.

Radiolysis of H₂-CO₂-He Mixtures

A study of the radiolysis of a mixture of carbon dioxide and hydrogen in helium shows that the radiolytic rate decreases with a decrease in the relative concentrations of CO₂ and H₂. At relative concentrations of H₂ and CO₂ greater than 1000 ppm the 100 eV yield of products is about 1.8 molecules/100 eV which decreases slowly with relative concentrations to a value of less than 1.0 molecule/100 eV at 50 ppm. At concentrations in an HTGR the product yield would have a very low value.

(8.0) NONDESTRUCTIVE TESTING

Eddy Current Methods

A multiparameter eddy current tester has been used to solve a practical tube testing problem. This technique makes it possible to test for flaws both on the inner and outer

surface of a tube even though the flaws are under simulated tube supports. The advantages of using multiple frequencies for practical testing problems are presented in this study.

Ultrasonic Methods

Phase measurements of reflected ultrasonic waves incident near the Rayleigh critical angle are reported. These measurements substantiate the predictions of our model which differ substantially from previous theories. The critical angle is sensitive to changes in material properties such as elastic modulus and density.

Research into the selection and development of alloys for use in the core assembly of the Fast Flux Test Facility (FFTF), has indicated that for a material to mechanically survive the high fluence environment anticipated, its structural state (e.g., grain size and cold work) is of critical importance. To ensure that the specified requirements of structural state have been met, nondestructive evaluation of the core material, especially in fuel pin tubing and fuel bundle ducting, is mandatory. A testing methods program is described making use of critical angle ultrasonic techniques.

A series of measurements have been made to identify flaw size and shape by the use of wide bandwidth ultrasonic pulses and appropriate spectrum analysis techniques. Differences in the wide bandwidth spectrum are indicated for a series of flat disks, spheres, and ribbons of different sizes. Further study is required before practical implementation of a flaw identification ultrasonic system can be designed and built.

Thermal Methods

A full scale experimental thermal image transducer is described which was designed to test conical specimens. Also, an emittance-independent, noncontacting thermal-transducer

technique is presented. Results are presented to show the sensitivity that can be expected from this method.

Holographic Methods

Interferometric holography has been applied in a non-destructive manner to a number of sample configurations. These applications include double exposure holograms of pressurized Fast Flux Test Fuel reactor cladding pin weld samples and thermal conductivity gradients in honeycomb construction panels. Weld defects and annealed areas were resolved in the tubing. Bond discontinuities and void regions in the honeycomb panels correlated with results from a previous thermographic test.

(9.0) ATR WATER LOOP OPERATION AND MAINTENANCE

The ATR accumulated 6800 MWd at 185 MW during Cycle 1 ending in February. Cycle 1B ended in March after an accumulation of 3700 MWd. The 1D-N loop, which operated during these periods at design conditions, contained stainless steel tensile specimens and Zircaloy corrosion coupons.

2.0 FAST-REACTOR CERAMIC FUELS RESEARCH

HIGH TEMPERATURE CREEP OF UNIRRADIATED FAST-REACTOR OXIDE FUELS

O. D. Slagle

Considerable difficulty has been experienced in obtaining reproducible strain rates as a function of stress for UO_2 -20 wt% PuO_2 under four-point bending.^(1,2) One possible cause is microstructural alteration produced when large stresses are applied which subsequently change the strain rates obtained for low stresses.

Because of difficulties induced in obtaining suitable samples of UO_2 -20 wt% PuO_2 , tests were initiated on stoichiometric UO_2 to determine if a similar effect could be observed. The UO_2 samples had a density of 96.5% TD and a grain size of 14 μ . A series of measurements was carried out at 1495 °C in which loads were varied between stresses of 2100 psi and 8320 psi. All stresses and strains refer to the calculated values for the area under maximum tensile stress. The initial steady-state strain rate of 2100 psi was $3.4 \times 10^{-4} \text{hr}^{-1}$. After applying a load of 8320 psi, little change occurred in the strain rate at 2100 psi. At a total accumulated strain of 1.6%, a series of loads: 8320, 6960, 5270, 3690, and 2100 psi were applied. The strain rate for 8320 psi dropped 25% from the previous measurement; however, a plot of log strain rate versus log strain (Figure 2.1) for these five stresses fell on a line with a slope of 2.08. The strain rate at 2100 psi after this series of tests and an accumulated strain of 2.5% was $1.21 \times 10^{-4} \text{hr}^{-1}$ or about half that at 1.6% strain. Further straining to 4% strain resulted in an initial decrease in strain rate to $0.85 \times 10^{-4} \text{hr}^{-1}$, then an increase to $1.23 \times 10^{-4} \text{hr}^{-1}$, and finally a decrease to $0.89 \times 10^{-4} \text{hr}^{-1}$. These tests gave no firm conclusion as to what is responsible for changing creep rates during testing.

FIGURE 2.1. Plot of Maximum Strain Rate Versus Maximum Stress for UO_2 at 1495°C

A second sample was used to determine the activation energy for creep of stoichiometric UO_2 for comparison with existing data. A series of strain rates were measured for a stress of 3420 psi at temperatures of 1495 °C and 1410 °C. The least amount of scatter in the data occurred for strains between 2.3% and 3.1%, and strain rates versus time decreased evenly at both temperatures. The data is listed in Table 2.1. The activation energy was calculated from an average of the strain rates observed, and was found to be 106 kcal/mole, in good agreement with existing values.

TABLE 2.1. Comparison of Maximum Strain Rates at 1495 °C and 1410 °C for a Stress of 3420 psi to Determine Activation Energy. Data Were Taken in the Order Listed. Strain Rates Are in Units of 10^{-4}hr^{-1} .

<u>Maximum Strain Rate</u>	<u>Temperature, °C</u>	<u>Maximum Strain Rate, %</u>
3.87	1495	
	1410	0.80
3.09	1495	
	1410	0.70
2.84	1495	
	1410	0.52
2.55	1495	

From the tests on these two samples, it is evident that obtaining reproducible strain rates as a function of stress, strain, or temperature in four-point bending is a problem common both to UO_2 and UO_2 -20 wt% PuO_2 .

Calculation of maximum stress and strain rate for creep in four-point bending is commonly done by the use of expressions derived for elastic behavior which assume both small deflections and stress proportional to strain. Creep

measurements in mixed oxides have indicated that creep rates are not proportional to the applied stress. Therefore, an analysis has been carried out for the case where strain rate ($\dot{\epsilon}$) is proportional to stress (σ) raised to the nth power or

$$\dot{\epsilon} = B \sigma^n \quad (1)$$

where B is a proportionality constant.

The previous work of Popov⁽³⁾ has been extended to relate push rod deflection rate ($\dot{w}$) and the applied load (P). The resulting expression is

$$\dot{w} = B P^n \left[\frac{2(2 + 1/n)c}{bh^2} \right]^n \left[\frac{2c(L - c) + nc(L - 2c)}{(n + 2)h} \right]. \quad (2)$$

The different symbols are defined in Figure 2.2. For $n = 1$, this expression reduces to

$$\dot{w} = B P \frac{6c}{bh^2} \frac{c(3L - 4c)}{3h}. \quad (3)$$

Equation (3) agrees with elastic theory when compared with the expressions for maximum strain rate ($\dot{\epsilon}_m$) and stress (σ_m)

$$\dot{\epsilon}_m = \frac{c(3L - 4c)}{3h} (1/w) \quad (4)$$

and

$$\sigma_m = \frac{6c}{bh^2} P. \quad (5)$$

The importance of Equation (2) may be realized by comparing the values of B which are computed using Equation (2) and those which would result from inserting Equations (4) and (5) into Equation (1) or

$$\dot{w} = B P^n \left(\frac{6c}{bh^2} \right)^n \frac{c(3L - 4c)}{3h}. \quad (6)$$

- P = LOAD ON EACH KNIFE EDGE
 h = HEIGHT OF SPECIMEN
 b = WIDTH OF SPECIMEN (NOT INDICATED)
 L = DISTANCE BETWEEN LOWER KNIFE EDGES
 c = HORIZONTAL DISTANCE FROM UPPER KNIFE TO NEAREST LOWER KNIFE EDGE
 w = RELATIVE VERTICAL DISPLACEMENT BETWEEN UPPER AND LOWER KNIFE EDGES

FIGURE 2.2. Parameters Used in Analysis of Creep Under Four-Point Bending

Denoting B calculated for the creep case in Equation (1) as B_{cr} and for the elastic case in Equation (6) as B_{el} , then for given measured values of $\dot{w}$ and P the ratio of B_{cr}/B_{el} can be calculated as a function of n . The results of this calculation are plotted in Figure 2.3. For example, for the UO_2 data obtained in this quarter which had an n value of 2.08, the actual value of B would be 60% higher than that calculated using the elasticity relations.

FIGURE 2.3. Ratio of the Proportionality Constant Calculated Using Equation (2) to That Calculated Using Equation (6) Plotted Versus Stress Exponent

IN-REACTOR CREEP OF MIXED OXIDE FUELS

J. A. Basmajian

The second in-reactor creep test of UO_2 -20 wt% PuO_2 fuel was completed during this quarter. Capsule modifications were incorporated in this test which eliminated problem areas found in the initial test. An internal view of the modified creep test capsule is shown in Figure 2.4.

In order to eliminate the spilling problem caused by rotation of the capsule during insertion, a tilt mechanism was developed and used. This device signaled the technicians when the capsule was rotated approximately 5° in either direction from the vertical. The second improvement was the use of a gas bellows to apply the load to the specimens and thus prevent the bouncing of the previously-used dead weights. This technique allowed the capsule to be loaded into the reactor with essentially no load on the samples. The third change involved moving the heater out of the tin to prevent mass transfer and also resulted in less electric power being needed to maintain the temperature at 900°C .

The experiment ran in the K-West Reactor for 4 days. The temperatures recorded by the two thermocouples were between 875 and 900°C during the entire run. The load on the bellows was kept at 200 psi gage which resulted in a load of 450 g on one sample and 800 g on the other. The maximum stresses on the two samples were 413 psi and 735 psi, respectively.

The sample breakage signal was observed on the sample with the 413 psi load approximately 1 hr after insertion. The second signal was received approximately 2 hr later, but was found to have resulted from a short in the system. When the capsule was examined in the hot laboratory, both samples were found to be broken.

Neg 700300-1

FIGURE 2.4. General View of Four-Point Bending Creep Capsule

Recent out-of-reactor creep tests have resulted in some mixed oxide samples breaking after very short times under light loads. Investigation of the cause of this problem is in progress. Microstructural examination of untested samples has indicated the presence of crack networks which are likely to be extended by stress and result in failure during testing.

THERMAL CONDUCTIVITY OF (Pu,U)O₂

R. L. Gibby

The purpose of this study was to determine the effect of PuO₂ content on the thermal conductivity of stoichiometric (Pu,U)O₂ solid solutions. These data are required to estimate what effect plutonium redistribution during irradiation might have on the thermal conductivity of the fuel element. The thermal diffusivities of samples of UO₂, UO₂-5 wt% PuO₂, UO₂-12 wt% PuO₂, UO₂-30 wt% PuO₂, and UO₂-45 wt% PuO₂, all prepared by coprecipitation from a nitrate solution, have been previously reported.⁽⁴⁾ The thermal diffusivities of UO₂-20 wt% PuO₂, UO₂-25 wt% PuO₂, and PuO₂ have now been measured to complete the solid solution range of interest. During the last quarter, the thermal conductivities of each of the above oxides have been calculated in the temperature range 100 to 1200 °C.

Thermal conductivity was calculated using the relationship

$$\lambda (\text{W/cm-}^\circ\text{C}) = 4.186 \alpha \rho C_p \quad (1)$$

where

α = thermal diffusivity, cm²/sec

ρ = density, g/cm³

C_p = heat capacity, cal/g-°C.

Since thermal diffusivity was the only parameter measured as a function of temperature, it was necessary to assign values for

density and heat capacity. The density at room temperature was measured by immersion in FC-75 (an organic liquid). The samples were first coated, however, with a thin layer of collodion to assure that bulk density was determined. The variation of density with temperature was calculated from thermal expansion data.⁽⁵⁾ Because of thermal expansion, it was also necessary to recalculate previously reported thermal diffusivity data since it was found that neglecting changes in sample thickness due to thermal expansion would result in as much as 6% error in thermal diffusivity at 1200 °C. Heat capacity data for the mixed oxides were calculated using heat capacities reported for UO_2 ⁽⁶⁾ and PuO_2 ,⁽⁷⁾ assuming a Kopp's law relationship.

Calculated thermal conductivities are plotted as a function of temperature in Figures 2.5 through 2.11. The data for each of the different samples obey the relationship

$$\lambda = (A + BT)^{-1} \quad (2)$$

which is as expected for dielectric solids. Values of A and B, determined from a least squares analysis, are shown in Table 2.2.

The results of this study for UO_2 are compared in Figure 2.5 with the UO_2 thermal conductivity data of Godfrey et al.⁽⁸⁾ which was measured directly using a radial heat flow steady-state technique. The data are in reasonable agreement considering the possible differences in samples such as impurity content, microstructure, and stoichiometry.

FIGURE 2.5. Thermal Conductivity of UO_2

FIGURE 2.6. Thermal Conductivity of $\text{U}_{0.95}\text{Pu}_{0.05}\text{O}_2$

FIGURE 2.7. Thermal Conductivity of $U_{0.88}Pu_{0.12}O_2$

FIGURE 2.8. Thermal Conductivity of $\text{U}_{0.80}\text{Pu}_{0.20}\text{O}_2$

FIGURE 2.9. Thermal Conductivity of $\text{U}_{0.75}\text{Pu}_{0.25}\text{O}_2$

FIGURE 2.10. Thermal Conductivity of $\text{U}_{0.70}\text{Pu}_{0.30}\text{O}_2$

FIGURE 2.11. Thermal Conductivity of PuO_2

TABLE 2.2. Values of A and B Determined from a Least-Squares Fit of UO₂-PuO₂ Thermal Conductivity Data to the Relationship $1/\lambda = A + BT$, $T = ^\circ K$

Sample No.	% TD	% PuO ₂	A cm-°C/W	B cm/W	Std. Dev. for Single Mea.	Remarks
TR-0	96	0	3.08 ± 0.04	0.02294 ± 0.00004	±0.9006	
TR-5	97	5	3.04 ± 0.09	0.0239 ± 0.0001	±0.8043	
TR-12	97	12	2.20 ± 0.03	0.02714 ± 0.00003	±0.7665	
TR-20	98	20	3.09 ± 0.13	0.0261 ± 0.0001	±1.2279	
TR-25	97	25	3.13 ± 0.10	0.0260 ± 0.0001	±1.1557	
TR-30	97	30	5.30 ± 0.09	0.0252 ± 0.0001	±0.9222	Corrected to 97% TD from 92% TD
GCW-10	97	100	0.46 ± 0.18	0.0283 ± 0.0002	±1.670	Corrected to 97% Td from 93.3% TD

The data for $(\text{U,Pu})\text{O}_2$ solid solutions and PuO_2 are compared with the data for UO_2 in Figures 2.6 through 2.11. These comparisons make it readily apparent that PuO_2 substitution into the UO_2 lattice has a small but measurable effect on thermal conductivity. The decrease in thermal conductivity resulting from addition of PuO_2 to the UO_2 lattice is small in comparison, however, to decreases resulting from deviations from stoichiometry in mixed oxide compositions.⁽⁹⁾ It has been shown⁽⁹⁾ that increases in thermal resistance arising from deviations from stoichiometry are associated primarily with the term A in Equation (2). This term can be associated with thermal resistance arising from the presence of lattice defects which scatter phonons. As shown in Table 2.2, the slight increases in thermal resistance resulting from addition of plutonium to the lattice in this study appear, on the other hand, to be associated with the term B in Equation (2), which is related to the thermal resistance contribution arising from phonon-phonon interactions called Umklapp processes.

It appears at this time that the changes observed in the Umklapp resistance with plutonium content can be explained theoretically and, in fact, are related to changes in melting temperature which also occur on addition of plutonium.

PROPERTIES OF MOLTEN FAST-REACTOR OXIDE FUELS

R. P. Nelson, J. J. Rasmussen and O. D. Slagle

Measurements Facility

Improvements have been made in the facility (Figure 2.12) which will be used to measure volume expansion on melting and liquid density, surface tension, viscosity, and compressibility of UO_2 - PuO_2 fuels.

A high resolution, high sensitivity (brightness) X-ray image intensifier tube has been substituted for the fluorescent screen previously used in fluoroscopy of molten samples. This

FIGURE 2.12. Facility for Measurement of Properties of Molten $\text{UO}_2\text{-PuO}_2$ Fuels

change will facilitate the observation of melting and the condition of the liquid in density, surface tension, and viscosity measurements.

Provisions have been made in the glove box to install a new furnace (Figure 2.13) for measurement of surface tension by the pendant drop technique. This furnace will be operated alternately with the tungsten-mesh resistance furnace. Both furnaces will use a common power supply and radiographic system.

A rapid advance roll-film cassette has been designed and will replace the single sheet cassettes used previously. The cassette has been designed to be used in conjunction with the image intensifier, permitting simultaneous fluoroscopy and film radiography. This arrangement will be particularly useful in rapidly obtaining radiographs of UO_2 - PuO_2 pendant drops which are inherently unstable due to the high density of the molten oxides.

Density

Two techniques are being investigated which may lead to an improvement in the accuracy of measurements of the volume expansion on melting and liquid density of UO_2 - PuO_2 fuels. Liquid density of refractory oxides is currently determined by using radiography to measure the height of a liquid column of known mass and diameter.⁽¹⁰⁾ The accuracy of the radiographic technique is limited to about $\pm 30\%$ by the accuracy of determining the average column height.

One technique under consideration will employ a high-temperature gas pycnometer.⁽¹¹⁾ A tube extending from a refractory metal capsule containing the oxide sample will connect to a limited-volume gas system outside the furnace. Melting will result in a change in pressure of an inert cover gas as measured on a sensitive capacitance-type pressure gage,

FIGURE 2.13. Furnace for Surface Tension Measurements by Pendant Drop Technique

which may be related to the volume expansion. Since it is anticipated that rapid measurements may be made, this technique is potentially useful for measuring compositions such as PuO_2 which react to some extent with the refractory metal crucible. The tungsten-mesh furnace in the measurements facility can be adapted for this measurement.

The second technique for determining precise density will involve measuring the attenuation of a gamma ray beam in the sample above and below the melting point. Measurements of molten metal densities using this method have been reported.^(12,13) The tungsten-mesh furnace can easily be adapted for this measurement. A ^{60}Co source and a simple electronic counting system consisting of a scintillation detector, single-channel analyzer, and scaler/timer will be employed.

Both techniques will be tested for measurement precision using Al_2O_3 and UO_2 . Equipment has been assembled and construction of the systems has been initiated.

Surface Tension

A new furnace has been built to measure the surface tension of UO_2 - PuO_2 (Figure 2.13) by the pendant drop technique (Figure 2.14).⁽¹⁴⁾ This furnace is a double-shelled aluminum furnace which is completely water cooled. The unit has a D-shaped cross section which allows the radiographic film to be placed within 2-1/2 in. of the capsule center. A smaller capsule-film distance will reduce the geometric uncertainty which has been observed previously.

The surface profile computer program error was analyzed by rounding off sessile drop data reported by Butler and Bloom⁽¹⁵⁾ which had been measured to 0.00001 in. As long as the data was accurate to 0.001 in. or better, the error was less than 1%. If the data was only accurate to 0.01 in.,

FIGURE 2.14. Capsule Design for Pendant Drop Measurement of Surface Tension

the error jumped to 17%. In current studies, scribe marks used to outline the meniscus or the pendant drop profiles on enlarged prints made from radiographic negatives can be read to ± 0.001 in. Since the standard deviation of the calculated UO_2 surface tension is about 10%, the greatest source of error comes from not being able to accurately place the scribe marks on the photographic enlargements. Preliminary tests indicate that a commercially available microdensitometer can be used to obtain the profile data to 0.001 in. optically.

HOT-PRESSING OF FAST-REACTOR CERAMIC OXIDES

P. E. Hart

Mixed oxide fuel specimens, containing 25 wt% PuO_2 and having densities in excess of 95% TD, have been fabricated via hot-pressing for use in in-reactor and out-of-reactor creep tests.

Specimens are being fabricated from coprecipitated plutonium hydroxide and ammonium diuranate powders calcined for 8 hr at 850 °C under flowing Ar-8% H_2 . Hot pressing was previously performed at temperatures up to 1300 °C. However, specimens did laminate on reheating to 1600 °C.⁽¹⁾ Consequently, pressing temperatures have been increased to 1600 °C. The present calcining technique yields powders with O/M's that fall between 2.01 and 2.02. Higher initial O/M's yield pellets with lower densities. The dependence of densities on O/M probably results from O_2 release during reheating. Due to the relatively short time at elevated temperatures during hot-pressing, not enough strength is developed to prevent laminating when O_2 is released during reheating. By increasing the pressing temperature, the specimen strength has been increased and the excess O_2 concentration reduced.

Density versus temperature curves for hot pressing of 25 wt% PuO_2 specimens are shown in Figure 2.15. Autoradiographs

FIGURE 2.15. Influence of Pressing Temperature and Powder Preparation on Density of Hot-Pressed UO_2 -25 wt% PuO_2 ; Pressure = 5000 psi

of the unmilled powder specimens indicate that some areas contain high Pu concentrations. Calcined powders ball milled in ethyl alcohol yield reduced hot-pressed densities; PNL HP-86 density equals 10.60 g/cm^3 . The influence of milling on plutonium distribution has not been determined yet. The deactivation may be due to changes in surface adsorbed ions. Microstructures of this hot-pressed pellet are being examined.

As previously reported,⁽²⁾ microstructures of hot-pressed UO_2 powders, initially having high O/M (above 2.2) exhibited two phases. Pressed pellet O/M ratios were approximately 2.04. This O/M corresponds to about 2 mole % U_4O_9 . The pellets exhibited microstructures typical of U_4O_9 in a UO_2 matrix as reported by Schaner.⁽¹⁶⁾

Density of UO_2 specimens hot-pressed at low temperatures strongly depends on powder initial O/M. As shown in Figure 2.16 for specimens pressed at 900°C , small changes in O/M cause significant changes in final density. These results reflect like influence of O/M on creep behavior of UO_2 .

A typical microstructure of hot-pressed UO_2 specimens is shown in Figure 2.17. Specimen density is 10.90 g/cm^3 , O/M = 2.01 and grain size is $14 \text{ }\mu\text{m}$. The specimen bulk is free of cracks. However, pressing defects appear on pellet surfaces.

LATTICE EXPANSION OF $\text{UO}_2\text{-PuO}_2$

P. G. Pallmer

A lattice thermal expansion of $9.8 \times 10^{-6}/^\circ\text{C}$ has been measured for $(\text{U}_{0.75}\text{Pu}_{0.25})\text{O}_2$ to 1600°C . The temperatures were obtained from the lattice expansion of added molybdenum powder. The measured values were corrected for misalignment by extrapolation to $180^\circ\text{-}2\theta$ versus the function $\cos\theta \cot\theta$. The extent of misalignment resulting from expansion of the ribbon appeared to vary uniformly with temperature. The same

FIGURE 2.16. Influence of Powder O/M on Hot-Pressing Behavior of UO_2 at 900 °C;
Pressure = 4000 psi

250X

250X

FIGURE 2.17. Microstructure of Hot-Pressed UO_2 ;
Density = 10.90 g/cm^3 (PNL HP-55)

correction was used for the single molybdenum reflection used as a temperature reference. The temperatures were read from curves fitted to published parameter values.^(17,18) The values are listed in Table 2.3, and plotted in Figure 2.18.

TABLE 2.3. Lattice Expansion of $(U_{0.75}Pu_{0.25})O_{2.00}$

Lattice Parameter		Temperature, ^(a) °C
Molybdenum, Å	Oxide, Å	
3.1466	5.4530	25
3.1548	5.476	505
3.1612	5.492	825
3.1650	5.498	995
3.1700	5.513	1215
3.1740	5.523	1390
3.1744	5.524	1410
3.1800	5.539	1640

a. Temperatures derived from the expansion of molybdenum from 25 °C.

Powdered samples of the coprecipitated stoichiometric dioxide were affixed to an aligned tungsten ribbon heater. After the vertical tungsten ribbon is aligned in the diffractometer, as shown in Figure 2.19, the stage is moved to a horizontal position for application of the powders as a slurry. Alignment is recovered with the aid of auxiliary clamps and shims, as shown in Figure 2.20. A temperature gradient in the oxide powder layer influences the temperature of the admixed molybdenum powder and its normally well-resolved lines were also broadened. To minimize thermal gradients, an additional U-shaped ribbon was positioned in front of the sample ribbon to function as a heat shield. The diffracted beam was occulted from about 130 to 140° 2θ and though resolution was improved, line broadening was still present above 1600 °C. The

FIGURE 2.18. Lattice Expansion of $(U_{0.75}Pu_{0.25})O_{2.00}$

Neg 700669-5

X-Ray Diffractometer with Alignment Gage for Tungsten Ribbon

FIGURE 2.19

Neg 700669-4

FIGURE 2.20. Vacuum Chamber for High Temperature Lattice Parameter Measurements on the X-Ray Diffractometer

thickness of the applied powder layer now appears to have the greatest effect on line broadening at high temperature. The broadening results from a temperature gradient arising because of the low absorptivity of the oxide.

Further improvement in the accuracy of the thermal expansion values may benefit from more direct temperature measurements. Nominally corrected optical pyrometer readings as well as temperatures indicated by tungsten lattice thermal expansion are somewhat lower than the molybdenum temperatures. A helium atmosphere did not lead to differing expansion values. The helium cools the ribbon but apparently does not efficiently transfer heat to the sample, although some reduction in line broadening was noted. Tungsten-rhenium alloy thermocouple wires have been spot welded to the back of the tungsten ribbon to provide a more direct measurement of the temperature.

PHASE STUDIES

C. E. McNeilly

Modification of the micro-thermobalance apparatus for use at higher temperatures is in progress in order to extend the measurement of O/M ratio of mixed oxides as a function of oxygen potential to a temperature of at least 1500 °C from the present limit of about 1100 °C. The present upper temperature limit is determined by the maximum temperature to which the alumina reaction tubes can be heated in hydrogen atmospheres containing low oxygen potentials (< -110 kcal/mole O_2). We have been attempting to replace the alumina with thoria tubes, but this has proved extremely difficult since thoria tends to fail in the present experimental arrangement after only a few heating-cooling cycles.

This problem will be circumvented by protecting the alumina from the reducing atmosphere by means of a close-fitting liner made from 0.001-in. thick molybdenum. The liner

has been fabricated and will be installed and tested and, if satisfactory, experiments should be started soon.

In order to cover the stoichiometry range of interest ($1.90 < O/M < 2.00$) in the temperature interval of 1100 to 1500 °C, the oxygen potential must be controlled over the range of about -100 to -140 kcal/mole O_2 . This region of oxygen potentials, coupled with the temperature dependency in the equilibrium constant for the H_2/H_2O equilibrium, requires wide variations in the controlled parameters for the sample atmosphere. The two parameters most amenable to control are the partial pressures of hydrogen and water vapor. Hydrogen content is controlled manometrically whereas the water vapor partial pressure is varied by controlling the temperature of a water (ice) trap through which the hydrogen gas circulates. The most severe case [$\Delta\bar{G}(O_2) = -140$ kcal/mole O_2 at 1200 °C] calls for an ice bath temperature of -51.4 °C. This temperature is attained by means of a dry-ice-boosted, mechanically refrigerated cryostat. By using a thermistor temperature controller with the thermistor sensing element cemented to the cooling coils, the cryostat temperature can be controlled to better than ± 0.1 °C.

DETERMINATION OF OXYGEN/METAL RATIOS FOR OXIDE FUELS

R. E. Woodley

The ability to determine oxygen/metal ratios on very small samples of oxide fuels, on the order of 1 mg, would allow the measurement of oxygen distributions across the diameters of fuel elements. The reduction of a fuel sample of this magnitude by carbon yields an amount of CO_2 which may be readily analyzed chromatographically. However, the precision of the determination depends upon the elimination of extraneous sources of CO_2 . In the present study, the graphite crucible initially employed was replaced by a tungsten crucible because

of outgassing difficulties with the graphite crucible. Even the small amount of powdered graphite currently employed is not readily relieved of its oxides of carbon prior to reduction of the fuel sample. Furthermore, any minute air leakage into the experimental system, upon reaction with the heated graphite, yields significant quantities of CO_2 .

Several measurements made during the past quarter to determine the extent of this extraneous CO_2 were performed using the following procedure or slight variation thereof: About 10 mg of finely powdered, pure graphite, which is more than sufficient to reduce 1 mg of UO_2 , was inserted in the tungsten crucible which, in turn, was installed in the experimental system. Following room temperature evacuation, the crucible was heated inductively to about 2000 °C for at least 1 hr at which time the pressure was $<10^{-5}$ torr. The crucible was then returned to room temperature, the system brought to atmospheric pressure with pure helium, and the crucible plus contents were exposed to air for up to 10 min to simulate the loading of the UO_2 sample. After reassembling the system and evacuating, the crucible plus graphite were heated to about 1200 °C for 1 to 2 hr under either vacuum or a stream of pure helium. At 1200 °C, outgassing of the graphite occurs without reduction of the fuel sample. Following this outgassing period, the crucible temperature was increased to about 1700 °C and maintained at this temperature for a period sufficient to reduce 1 mg of UO_2 . During the simulated fuel reduction, released gases were swept through a heated tube of CuO , where CO is oxidized to CO_2 , and into a trap immersed in liquid nitrogen where the evolved CO_2 was collected for subsequent analysis.

The smallest amount of CO_2 collected, 0.18 micromole, is equivalent to $\Delta(\text{O/M}) = 0.056$ for 1 mg of UO_2 . This amount of CO_2 is considered excessive, and future work on this method of

O/M determination will concentrate on its elimination or reduction to an acceptable value. Although it would not be as convenient, it may prove necessary to eliminate any exposure of the powdered graphite to air following the initial high temperature outgassing by loading the fuel sample into the crucible in an inert atmosphere.

To improve the accuracy of the CO_2 analysis, the gas chromatograph employed in this investigation has been calibrated for CO_2 concentrations in the range from 0 to 1%. Four separate analyses at each of nine different CO_2 concentrations within this concentration range were averaged to construct the desired calibration curve.

REFERENCES

1. Quarterly Progress Report, July, August, September 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-1223. Battelle-Northwest, Richland, Washington, November 1969.
2. Quarterly Progress Report, October, November, December 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-1279. Battelle-Northwest, Richland, Washington, February 1970.
3. E. P. Popov. "Bending of Beams with Creep," J. Appl. Phys., vol. 20, pp. 251-256. 1949.
4. R. L. Gibby. "Thermal Diffusivity of $(\text{U,Pu})\text{O}_2$," Quarterly Progress Report, January, February, and March 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-1058, p. 5.8. Battelle-Northwest, Richland, Washington, May 1969.
5. J. Roth and E. K. Halteman. "Thermal Expansion of Coprecipitated $(\text{U,Pu})\text{O}_2$ Powders by X-Ray Diffraction Techniques," NUMEC-2389-9. Nuclear Materials and Equipment Corp., Apollo, Pennsylvania, October 1965.

6. J. B. Conway and A. D. Feith. An Interim Report on a Round Robin Experimental Program to Measure the Thermal Conductivity of Stoichiometric Uranium Dioxide, GEMP-715. General Electric Company, Cincinnati, Ohio, October 1969.
7. Owen L. Kruger and Howard Savage. "Heat Capacity and Thermodynamic Properties of Plutonium Dioxide," J. of Chem. Phys., vol. 49, no. 10, pp. 4540-4544. 1968.
8. T. G. Godfrey, W. Fulkerson, T. G. Kollie, J. P. Moore, and D. L. McElroy. "Thermal Conductivity of Uranium Dioxide from -57 °C to 1100 °C by a Radial Heat Flow Technique," J. Am. Cer. Soc., vol. 48, no. 6, pp. 297-305. 1965.
9. R. L. Gibby. The Effect of Oxygen Stoichiometry on the Thermal Diffusivity and Conductivity of $U_{0.75}Pu_{0.25}O_{2-x}$, BNWL-927. Battelle-Northwest, Richland, Washington, 1969.
10. Quarterly Progress Report, July, August, September 1968, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-919, p. 5.2-5.3. Battelle-Northwest, Richland, Washington, November 1968.
11. J. O'M. Bockris, A. Pilla, and J. L. Barton. "Densities of Solid Salts at Elevated Temperatures and Molar Volume Change on Fusion," J. Phys. Chem., vol. 64, pp. 507-508. 1960.
12. A. S. Basin and A. N. Solov'ev. "Investigation of the Density of Liquid Pb, Cs, and Ga by the Gamma Method," Zh. Prikl. Mekh. Tekh. Fiz., no. 6, pp. 83-87. 1967. (In Russian) (English translation: ANL-Trans-677.)
13. I. G. Dillion. Density Determinations Using a Gamma Radiation Attenuation Technique, ORO-3404-3. Oak Ridge Operations Office, Oak Ridge, Tennessee, September 30, 1968.
14. Quarterly Progress Report, April, May, June 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-1144, p. 5.1-5.5. Battelle-Northwest, Richland, Washington, August 1969.

15. J. N. Butler and B. N. Bloom. "Curve-Fitting Method for Calculating Interfacial Tension from the Shape of a Sessile Drop," Surface Science, vol. 4, pp. 1-17. 1966.
16. B. E. Schaner. "Metallographic Determinations of the UO_2 - U_4O_9 Phase Diagram," J. Nucl. Mater., vol. 2, p. 110. 1960.
17. J. W. Edwards, R. Speiser, and H. L. Johnston. "High Temperature Structure and Thermal Expansion of Some Metals as Determined by X-Ray Diffraction Data. I. Platinum, Tantalum, Niobium, and Molybdenum," J. Appl. Phys., vol. 22, p. 424. 1951.
18. R. G. Ross and W. Hume-Rothery. "High Temperature X-Ray Metallography," J. Less-Common Metals, vol. 5, p. 258. 1963.

3.0 IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS

J. A. Christensen

IN-REACTOR THERMAL PERFORMANCE OF $\text{UO}_2\text{-PuO}_2$ FUEL

J. A. Christensen

Fuel-Clad Gap Conductance Measurements

L. A. Lawrence, J. A. Christensen and G. R. Horn

The high heat fluxes and large fuel-clad gaps characteristic of FTR fuel pins result in large calculated temperature drops across the fuel-clad interface. The associated decrease in allowable heat rating may be 30 to 40% of the rod powers permissible in rods with closely mating fuel and clad. Closure of the fuel-clad gap has been observed and is anticipated early in life for FTR. Object of the present study is to measure the rate of gap closure and associated change in ΔT as a basis for specifying reactor startup schedules.

Capsule FP-37, designed to study fuel-clad gap closure kinetics, was successfully irradiated for 100 hr in a Hanford K Reactor. Fuel characteristics and test conditions are listed in Table 3.1. The test variables were fuel-clad gap and fill gas. The fill gas contained various amounts of xenon to simulate effects of fission gas contamination of the initial helium bond on gap conductance. The two fuel-clad diametral gaps were 0.002 in. (the practical fabrication lower limit) and 0.011 in. The latter is chosen to maintain the gap-to-fuel diameter ratio expected in nominal FTR fuel pins.

The capsule was equipped with upstream and downstream thermocouples to measure individual rod powers. The assembled capsule was flow tested prior to irradiation to determine the coolant flow in the four channels. The flow data along with the temperature differential measurements provided a continuous heat rating record for each pin during irradiation.

TABLE 3.1. Fuel Characteristics and Test Conditions for FP-37A. Fuel

Form	Cold pressed and sintered pellets
Composition	UO ₂ -25 wt% PuO ₂
Oxygen/Metal	1.96 ± 0.01
Density	95% TD
²³⁴ Pu/Pu	87%
²³⁵ U/U	Normal
Diameter	0.320 in.
Cladding	Type 304L SS
Cladding OD	0.458 in.
Impurity Content	<400 ppm cation impurity

B. Test Conditions

Rate of Power Increase During Charging	60% per hr
Rate of Power Decrease During Discharge	3% per sec
Time at Steady-State Power	100 hr

C. Capsule

<u>Fuel Pin Number</u>	<u>Fuel-Clad Gap, in.</u>	<u>Fill Gas^(a)</u>
FP-37-1A	0.002	He
1B	0.011	He
2A	0.002	He-41.2 Xe
2B	0.011	He-41.2 Xe
3A	0.002	He-75.6 Xe
3B	0.011	He-75.6 Xe
4A	0.002	Xe
4B	0.011	Xe

a. Preirradiation analysis

Following irradiation, the four fuel pins were removed from the capsule assembly and sampled for fission gas. Each of the eight fueled segments was sectioned at the fuel mid-plane and two adjacent samples removed--one for ceramography and the other for radiochemical burnup analyses based on ^{95}Zr , ^{141}Ce , and $^{140}\text{Ba-La}$. Fuel Pin 3A was radially sampled for the fission products ^{95}Zr , ^{99}Mo , and ^{141}Ce by selectively removing small amounts of fuel (approximately 0.5 mg) with a 0.025 in. diameter high speed twist drill and vacuum sniffer. Ceramography samples were impregnated with epoxy resin and polished through a 600 grit. Macrographs and alpha and beta-gamma autoradiographs were taken of these surfaces (Figures 3.1 through 3.4).

Postirradiation structures are consistent with the increase in fuel-clad gap and xenon content of the fill gas. Pins 1B, 2B and 3B show cracking of the unrestructured fuel some of which probably occurred during the postirradiation cutting operation. All of the "B" fuel pins have an approximately cylindrical central void which increases in size with xenon content of the fill gas. Pins 3B and 4B show fuel melting to 10% and 35% of the fuel radius, respectively. Only these pins show any plutonium relocation on the alpha-autoradiographs, with a depleted band corresponding roughly to the pore free zone on the macrographs. Preliminary examination of the fuel-clad gaps indicates only a decrease in the initial 0.011 in. fuel-clad diametral gap after 100 hr of irradiation to 0.004 to 0.006 in. Precise measurements of the postirradiation fuel-clad gaps is in progress.

Calculations to determine relative values for the fuel-clad gap conductance for the eight pins by matching observed fuel structures with temperature profiles are in progress. These computations are performed using the SINTER program operational on the Battelle-Northwest Hybrid Computer.

Neg 701157-3

FIGURE 3.1. Postirradiation Structures of Fuel Pins 1A and 1B

Neg 701157-5

FIGURE 3.2. Postirradiation Structures of Fuel Pins 2A and 2B

Neg 701157-4

FIGURE 3.3. Postirradiation Structures of Fuel Pins 3A and 3B

Neg 701157-1

FIGURE 3.4. Postirradiation Structures of Fuel Pins 4A and 4B

Molybdenum Distribution in Irradiated Oxide Fuels

L. A. Lawrence and J. A. Christensen

The distribution of fission products affects fuel life-time and safety. Fission product molybdenum is of particular interest in that the oxygen balance in irradiated oxide fuels hinges upon the oxidation state of the molybdenum. The relative location of the molybdenum is thus important in considerations of the radial O/M profile in irradiated oxide fuels. In addition, estimations of the contributions of solid fission products to the swelling of oxide fuels requires a knowledge of their redistribution during irradiation. As part of the postirradiation examination of several UO_2 -25 wt% PuO_2 fuel pins, radial distributions of ^{99}Mo were measured.

Because of the relative short half-life of the only gamma active molybdenum isotope present (^{99}Mo , $\tau_{1/2} = 67$ hr) the elapsed time from sample discharge to postirradiation examination must be less than 1 week. The close proximity of the special irradiation facilities at the Hanford reactors and the Radiometallurgy Laboratory permits this short elapsed time. The fuel characteristics and test conditions of the two fuel pins of interest are listed in Table 3.2.

The radial distribution of molybdenum was measured by removing from selected positions across the fuel radius small amounts of fuel using a high speed, 0.025 in. diam twist drill and a vacuum sniffer. The samples (approximately 0.5 mg) were radiochemically analyzed for ^{99}Mo and plutonium. To normalize the molybdenum activity to a constant sample size it was assumed that no plutonium relocation occurred during irradiation. This should be a valid approximation as the alpha-autoradiographs (Figure 3.5) show no indication of any plutonium movement. The similarity of the postirradiation structures of the two pins (Figure 3.5) indicate that they operated at approximately the same heat rating, the only

TABLE 3.2. Fuel and Test Characteristics and Results

A. Preirradiation

Form	Cold pressed and sintered pellets
Composition	UO ₂ -25 wt% PuO ₂
Oxygen/Metal	1.96 ± 0.01
Density	95% TD
²³⁹ Pu/Pu	87%
²⁴⁰ Pu/Pu	11%
²³⁵ U/U	Normal
Average Fuel-Clad Gap	0.003 in.
Cladding	Type 304L SS

B. Irradiation

<u>Capsule/Pin</u>	<u>Irradiation Time</u>
FP-35 35-3A	10 hr
FP-37 37-3A	100 hr
Rate of Power Increase During Charging	60% per hr
Rate of Power Decrease During Discharge	3% per sec

C. Postirradiation

<u>Pin/Sample</u>	<u>R/R₀, (a) %</u>	<u>Activity (b)</u>
35-3A 1	20	0.180
2	26	0.198
3	50	0.325
4	67	0.415
5	87	0.778
37-3A 1	9	0.174
2	21	0.196
3	42	0.216
4	68	0.554
5	92	0.892

a. Sample width 13% of fuel radius.

b. Normalized to 1.00 at the fuel surface.

Neg 701157-2

FIGURE 3.5. Postirradiation Structures of Pins 35-3A (10 hr Test)
and 37-3A (100 hr Test)

difference being the length of irradiation. Radiochemical burnup determination based on ^{95}Zr , ^{141}Ce , and $^{140}\text{Ba-La}$, showed that the 10-hr and 100-hr samples operated at linear power densities within 10% of each other.

The molybdenum activity of the microdrilled samples was normalized to a value of 1.00 at the fuel surface. Results (Figure 3.6) show no difference between the 10-hr and 100-hr test suggesting no significant relocation of the molybdenum in the first 100-hr of irradiation at heat ratings below incipient melting.

Fission Product Migration in Oxide Fuels

L. A. Lawrence and J. A. Christensen

Five identical UO_2 -25 wt% PuO_2 fueled pins, irradiated for a short period of time at carefully controlled conditions, were radially sampled for the fission products ^{95}Zr , ^{103}Ru , and ^{141}Ce . Fuel characteristics and test conditions are listed in Table 3.3.

Radial fission product distributions were obtained by selectively drilling polished fuel cross-sections at several preselected positions using a high speed, 0.025-in. diam twist drill and collecting the drillings with a vacuum sniffer. The microdrilled samples were radiochemically analyzed for ^{95}Zr , ^{103}Ru , ^{141}Ce , and plutonium. Plutonium content was obtained from an alpha spectrum analysis. From the specific activity of the fuel and plutonium activity of the microdrilled samples, sample size was calculated.

Fission product activity was normalized to a constant sample size (Figures 3.7 through 3.11).

The resulting fission product concentration profiles show no major relocation of ^{95}Zr and ^{141}Ce except for a slight minimum associated with partial fuel melting in Pins 3, 4, and 5.

Neg 701168-5

FIGURE 3.6. ^{99}Mo Distribution

TABLE 3.3. Fuel and Test Characteristics and ResultsA. Preirradiation

Composition	UO ₂ -25 wt% PuO ₂
Oxygen/Metal	1.96 ± 0.01
Density	95% TD
²³⁹ Pu/Pu	87%
²³⁵ U/U	Normal
Diameter	0.320 in.
Average Fuel-Clad Gap	0.003 in.
Cladding	Type 304L SS

B. Irradiation

Rate of Power Increase During Charging	60% per hr
Rate of Power Decrease During Discharge	3% per sec
Time at Steady-State Power	10 hr
Calculated Fuel Surface Temperature at Steady-State	950 °C
In-Reactor Calorimetry Data	

Heat Rating, kW/ft

<u>Pin No.</u>	<u>Peak Value During 10 hr</u>	<u>Value at Time of Withdrawal</u>	<u>Average Value for 10 hr</u>
1	22.0	21.8	21.2
2	29.3	29.0	28.4
3	*	*	*
4	30.7	29.7	31.3
5	33.0	32.3	31.6

* Thermocouple failed during irradiation.

TABLE 3.3. (contd)C. Postirradiation

Pin No.	Maximum Extent of Fuel Melting* $\frac{R_{\text{melt}}}{R_0}, \%$	Minimum ^{103}Ru Concentration $\frac{R}{R_0}, \%$
1	0	---
2	0	20
3	25	30
4	43	50
5	47	45

* Identified as the outer boundary, moving outward of the pore free zone.

Migration of plutonium can be seen on the alpha-autoradiographs from these sections. The autoradiographs are not quantitative, thus some estimate of plutonium relocation is necessary. Plutonium redistribution has been observed in mixed-oxide fuels operating at comparable heat ratings.⁽¹⁾ The observed redistribution was on the order of 5% which is consistent with the minimum observed in the ^{95}Zr and ^{141}Ce data.

The ^{103}Ru concentration profiles show gross relocation. The ^{103}Ru concentration minimum corresponds approximately to the observed maximum extent of fuel melting. This correlation suggests that a zone refining process is responsible for the ruthenium redistribution.

Effects of Burnup on Melting Heat Rating

C. R. Hann

The accumulation of fission products in mixed-oxide fuel decreases the thermal conductivity and melting point, thereby reducing the allowable heat rating of high burnup fuel rods.

α - AUTORADIOGRAPH $\beta\gamma$ - AUTORADIOGRAPH

MICROGRAPH

Neg 701168-3

FIGURE 3.7. Fission Product Distribution and Postirradiation Structures of Pin 1

α - AUTORADIOGRAPH

βγ - AUTORADIOGRAPH

MICROGRAPH

Neg 701168-6

FIGURE 3.8. Fission Product Distribution and Postirradiation Structures of Pin 2

α - AUTORADIOGRAPH $\beta\gamma$ - AUTORADIOGRAPH

MICROGRAPH

Neg 701168-2

FIGURE 3.9. Fission Product Distribution and Postirradiation Structures of Pin 3

α - AUTORADIOGRAPH

$\beta\gamma$ - AUTORADIOGRAPH

MICROGRAPH

Neg 701168-4

FIGURE 3.10. Fission Product Distribution and Postirradiation Structures of Pin 4

α - AUTORADIOGRAPH

$\beta\gamma$ - AUTORADIOGRAPH

MICROGRAPH

Neg 701168-1

FIGURE 3.11. Fission Product Distribution and Postirradiation Structures of Pin 5

Comparative irradiations of fast-flux-irradiated, high-burnup fuels and their unirradiated counterparts will be used to determine the magnitude of the heat rating reduction.

High burnup (to 47,000 MWd/MTM), EBR-II-irradiated specimens will be available by the first of April. Estimated activity and gamma radiation levels of the test specimens are about 700 curies and about 600 R/hr at 3 ft, respectively. These high levels require an irradiation assembly that is compatible with hot-cell encapsulation requirements and handling techniques available at the irradiation testing facilities. Suitable closure techniques have been designed for the capsule that will contain the irradiated fuel; tests will be conducted on mockup capsules prior to encapsulation. Hazard analyses for reactor charging operations are in progress. Preliminary work indicates shielding problems will be difficult but not impossible. Efforts are continuing on methods to accommodate instrumentation leads in the shielding facilities.

The capsules will be irradiated for short times at heat ratings that result in small amounts of center melting. The effects of burnup on melting heat rating will be determined by measuring the extent of melting and compensating for changes in fissile atom and fission product inventories.

Evaluation of Alternate Irradiation Facilities

C. R. Hann and L. A. Lawrence

The thermal irradiations are scheduled for the remaining Hanford K Reactor. Alternate facilities at the National Reactor Testing Station in Idaho were evaluated in the event the existing facilities at Hanford were phased out. No serious obstacles were foreseen in transferring tests to the Engineering Test Reactor, and some test positions in the Advanced Test Reactor probably could be used with appropriate shielding. However, neither the ETR nor the ATR provides the

precise experimental conditions available in the K reactors, nor are they as convenient and economical to use.

Fast Reactor Experiments

C. R. Hann

Thermal irradiation facilities have been used in this program thus far because they permit more precise study of certain selected variables. Not only are the facilities more accessible than the existing fast test space, but thermal irradiations are simpler, less time consuming, and less expensive. One of the main goals of this program has been to identify, through the use of thermal test facilities, critical experiments for EBR-II. Such tests now being planned include:

- Fuel swelling measurements.
- Measurement of fission gas release rate during irradiation.
- A study of the effects of power cycles on fuel-cladding mechanical interactions.

These experiments are being scoped for preliminary discussions with the ANL EBR-II Project staff prior to submitting the Approval-in-Principle documentation.

FUEL-TO-CLADDING GAP CONDUCTANCE ANALYSES

G. R. Horn

Fast reactor fuel pins operate with twice the surface heat flux characteristic of thermal reactor fuel rods. A commensurately more precise knowledge of the heat transfer coefficient at the fuel-to-clad interface is required for fuel temperature calculations. Several investigators have attempted to measure fuel-clad gap conductance but, as will be shown, the results are not sufficiently precise.

Gap conductance data published by Bain,⁽²⁾ Duncan,⁽³⁾ Baily, et al.,⁽⁴⁾ and Craig, et al.⁽⁵⁾ were analyzed for this

study and are summarized in Figure 3.12. Conditions represented in each experiment are summarized in Table 3.4. Figure 3.12 shows that a correlation between conductance and initial diametral gap does exist. A least squares fit of the data yields the functional relationship:

$$h_g = 1873e^{-67.6G} \quad (1)$$

where

h_g = gap conductivity, Btu/hr-ft²-°F

G = cold diametral gap, in.

The imprecision of this relationship is so great (as indicated by the 95% confidence band on Figure 3.12) as to render it virtually useless. For example, for a fuel pin having a cold diametral gap of 0.006 in., Equation (1) predicts (with 95% confidence) that the gap conductivity will be between 540 and 2850 Btu/hr-ft²-°F. As will be shown, this range is too broad for design purposes.

Attempts to develop more sophisticated correlations involving other parameters such as heat rating, fuel diameter, fill gas thermal conductivity, etc., have so far provided no better correlation than the one shown in Figure 3.12. Results of some individual experimenters have shown, however, that improved correlations should exist. For example, Bain⁽²⁾ showed that, in fuel pins with constant cold fuel-to-clad gaps, gap conductance increased with increasing heat rating. Bain also showed that gap conductance in helium-filled pins was higher than that for similar, argon-filled pins. The latter conclusion was confirmed by Duncan, but Craig et al. concluded that fill gas had no significant effect on the heat transfer coefficient.

The large uncertainties that exist in correlating fuel-to-clad heat transfer coefficients with fuel pin fabrication

FIGURE 3.12. Gap Conductivity as a Function of Cold Diametral Gap (From Data of Bain², Duncan³, Baily, et al.⁴ and Craig et al.⁵)

TABLE 3.4. Summary of Conditions Embodied
in Measurements of Gap
Conductivity

Experimenter	Cold Diametral Gap, in.	Fuel Diameter, in.	$\int_{T_s}^{T_c} c_k dT,$ W/cm	Fill Gas	Measured Gap Cond.
Bain ⁽²⁾	0.003 to 0.0331	0.660 to 0.740	38 to 89	Argon Mostly, Some W/He	116 to 2460
Duncan ⁽³⁾	0.002 to 0.012	0.334 to 0.432	40 to 54	Helium	635 to 5100
Baily et al. ⁽⁴⁾	0.002 to 0.010	0.218	40.5	Argon	600 to 2000
Craig et al. ⁽⁵⁾	0.0008 to 0.0101	0.218	32 to 48	Argon or Helium	520 to 3000

or operating parameters result primarily from uncertainties associated with individual gap conductance measurements. Although none of the investigators have reported error limits for their data, it must be realized that the experimental error in all of the measurements is very large. Craig et al.⁽⁵⁾ for example, reported a gap conductance of about 1000 Btu/hr-ft²-°F for a fuel pin having an initial cold diametral gap of 0.0041 in. If one assumes that power generation was known to ±5%; coolant temperature was known to ±5 °F; material properties and conditions of heat transfer between coolant and cladding were known to ±5%; fuel thermal conductivity was known exactly; the temperature at which columnar grains form was known to ±50 °C; and dimensions of structural features observed during postirradiation examination were known to ±0.001 in.; the calculated gap conductance varies from 408 to 2935 Btu/hr-ft²-°F. This magnitude of uncertainty is typical for all of the reported gap conductivity values.

The importance of knowing precise values for gap conductivity is illustrated by Figure 3.13. This figure was constructed for a hypothetical fuel pin having the following characteristics:

FIGURE 3.13. Allowable Heat Rating in a Hypothetical Fast Reactor Fuel Pin as a Function of Gap Conductivity

Cladding OD	0.250 in.
Fuel OD	0.212 in.
Fuel Density	93% TD

It was assumed that the pin was cooled with 750 °F sodium and that thermal conductivity of fuel was that published by Baily et al.⁽⁶⁾ It was further specified that fuel melting defined the maximum allowable heat rating. With these assumptions, Figure 3.13 shows how allowable heat rating (i.e., the heat rating which would just cause melting) varies with changing gap conductance. Obviously, if gap conductances in fast reactor fuels are high (above ~ 3000 Btu/hr-ft²-°F), the precise knowledge is not important to fuel pin design (increasing gap conductivity from 3000 to 4000 increases allowable heat rating by less than 2%). Alternatively, if gap conductances are characteristically low (below ~ 1600 Btu/hr-ft²-°F), precise knowledge of conductances is important. As an illustration, suppose a conductance of 600 is used for design when the value actually is 1600. The pin would be limited to a heat rating of ~ 11.5 kW/ft instead of the 14.7 kW/ft actually allowable. In this case, the heat rating could be increased 30% if gap conductance was more precisely known.

These results illustrate the importance of continuing to collect gap conductance data, particularly those data obtained by more accurate experimental methods than ordinarily employed. Search for a data correlation which reduces the error bands in Figure 3.12 is continuing.

IRRADIATION OF HYPOSTOICHIOMETRIC $\text{UO}_2\text{-PuO}_2$ FUEL

J. E. Hanson

High Burnup Thermal Flux Irradiations

R. D. Leggett, J. W. Weber, L. A. Pember, D. Christensen,
E. D. Jensen and D. C. Bullington

This task includes irradiation testing of mixed-oxide fuel specimens of different stoichiometrics to high burnups in a thermal flux. Substoichiometric mixed-oxides are expected to exhibit better fuel cladding-sodium compatibility at high burnups than the nearly stoichiometric oxides currently specified for the FTR. System compatibility, swelling, and gas release characteristics of $\text{UO}_2\text{-PuO}_2$ fuel as a function of stoichiometry and fuel form must be determined at high burnup before a maximum performance fuel can be designed.

Status of Thermal Flux Irradiation Tests

The general status of the fuel pins being examined is summarized in Table 3.5. The reported burnup values were derived from ^{148}Nd analyses of sections removed from the center of the fuel pins. The burnup values for those pins not yet examined were estimated from the reported fluence, assuming a linear relationship between fluence and burnup. BNW 1-10 and 1-12 were discharged on November 9 at the end of ETR Cycle 104. These two capsules have been gamma scanned at the ETR. When the MTR reaches approximately 12 MW, expected early in February, they will be neutron radiographed. A decision will be made then whether to return the capsules to the ETR for further irradiation or terminate the irradiation and start examinations.

TABLE 3.5. Summary Status of Irradiation Experiments

Test Designation	Description ^(a) Fuel Form	O/M	Facility	Status			
				Goal Exposure, MWd/MTM	Actual Exposure, MWd/MTM	Discharge Date	Status
BNW 1-1	Solid	1.93	MTR	100,000	(Est)120,000	2-69	Being Exam. at BNW
BNW 1-2	Solid	1.93	ETR	50,000	(Est) 70,000	2-69	Being Exam. at BNW
BNW 1-3	Solid	1.96	MTR	100,000	(Est)150,000	2-69	Being Exam. at BNW
BNW 1-4	Solid	1.96	MTR	50,000	71,000	6-68	Exam. Completed at BNW
BNW 1-5	Solid	2.00	MTR	100,000	(Est)160,000	2-69	Being Exam. at BNW
BNW 1-6	Solid	2.00	MTR	50,000	75,200	10-68	Exam. Completed at BNW
BNW 1-7	Annular	1.93	MTR	50,000	<1,000		Lead Tube Failed. Exam at BNW
BNW 1-8	Annular	1.93	MTR	100,000	29,000	1-68	Failed. Exam. at BNW
BNW 1-9	Annular	1.96	MTR	50,000	81,800	6-68	Exam. Completed at LASL
BNW 1-10	Annular	1.96	ETR	100,000	(Est) 92,200	(Est)2-70	(b)
BNW 1-11	Annular	2.00	MTR	50,000	87,400	8-68	Exam. Completed at LASL
BNW 1-12	Annular	2.00	ETR	100,000	(Est) 96,500	(Est)2-70	(b)

a. Fuel

25% PuO₂-75% UO₂
 Pellets 0.212 in. OD (0.052 in. ID for annular pellets)
 Smeared density - 87.5% TD
 Pellet density - solid pellet 93% TD, annular pellet 95% TD
 Fuel column length 13.5 in.

Cladding

OD - 0.250 in.
 Wall thickness - 0.016 in.
 Type 304 SS

b. Discharged 2-69 for interim exam. Recharged 5-69. Discharged 11-9-69 for second interim.

Postirradiation Examination of BNW 1-1, 1-2, 1-3, 1-5

Metallographic Examination of BNW 1-1 and 1-2. Fuel pins BNW 1-1 and 1-2 were sectioned for metallographic and burnup samples. Specimens for cladding burst tests were also obtained. The burnup samples have been submitted for chemical analysis. Rough grinding is in progress on all of the longitudinal and transverse metallography samples. All metallographic preparation is being done using nonaqueous lubricants which react minimally with the fuel and fuel-cladding interface. Between grinding steps, the metallographic specimens are stored in an evacuated system. Each burst test and spare fuel pin section is stored in individual aluminum capsules marked with sample identification and orientation. These capsules are in turn stored in other capsules under approximately 25 psig, moisture-free argon.

Preparations are being made to section BNW 1-3 and 1-5 for metallographic examination. Grinding through 600 grit, macrophotography, and α and β - γ autoradiography have just been completed on samples 1-2A (longitudinal bottom of pin) and 1-2D1 and 1-2F (transverse and longitudinal respectively from the middle of the fuel column).

Sample 1-2A has a fission product ingot located in the central void approximately 3/8 in. from the original bottom of the fuel column. The ingot is located at the lower end of a mass of fuel that appears to have melted and moved down the central void. This ingot has been identified from beam analysis of gamma ray spectra data as primarily niobium. The β - γ autoradiographs show penetration of fission products into cracks in the lower insulator pellets to a depth of about 0.2 in. The α and β - γ autoradiographs reveal no evidence of extensive reaction of the fuel pellets and insulator pellets.

Sample 1-2F also shows a large ingot approximately 0.25 in. long filling the central void. Analysis of the gamma ray spectra data showed this ingot to be comprised primarily of ruthenium.

Preliminary measurements of void and columnar grain region diameters on sample 1-2D1 showed 0.075 and 0.175 in., respectively, for the 0.220 in. diameter pellet. There are many small metallic ingots in the columnar grains, as well as a concentration of these inclusions at the columnar grain terminus. Examination of these three metallography samples will continue to establish the conditions of the fuel and the movements of the fission products.

Shielded Electron Microprobe Examination

E. D. Jenson

Fuel pin BNW 1-6, Section E, has been partially examined in the shielded electron microprobe. This section was 6.8 in. from the lower end cap and had an average burnup of 75,200 MWd/MTM (38,000 MWd/MTM at centerline and >200,000 MWd per MTM at the surface) at an average power of 16.3 kW/ft. The fuel surface temperature was 1100 °C and the centerline temperature 2700 °C (calculated). It had cooled 12 months before microprobe examination. Figure 3.14 shows the surface examined with indications of areas and points examined.

Uranium and plutonium are known to undergo a redistribution during high temperature irradiations, with plutonium concentration increasing near the central void. Quantitative data on the concentration changes that occur are necessary for increased understanding of the processes involved. The concentration of uranium and plutonium was determined along four radii, as shown in Figure 3.14. Path R1 was analyzed using a 5 μ diameter beam at 10 μ intervals. The results, corrected for background and specimen current drift, are shown in

Neg 700504-2

25X

FIGURE 3.14. Mosaic of Fuel Pin BNW 1-6, Section E, Examined in the Shielded Electron Microprobe. R1, R2, R3, and R4 Are Paths of Radial Analysis of Uranium and Plutonium Distribution. R5, R6, and R7 Are Paths of Radial Analysis for Molybdenum and Ruthenium in the Fuel Matrix and in Inclusions

Figure 3.15. The concentration values are relative, being uncorrected for absorption, fluorescence, backscatter, and detector dead time. The net effect of these corrections is expected to be small. In the horizontal portion of the plots, an observed standard deviation of $\pm 1.2\%$ is applicable to the data. The sharp drops in both uranium and plutonium concentration are due to pores, voids, cracks, and fission product ingots. The largest dips in the curves are voids or cracks. Smaller dips may be either voids or fission product inclusions. To identify which, a simultaneous analysis for ruthenium (the most abundant element in the ingots) must be run. Both plots show depletion of uranium and plutonium in the outer fuel areas, with a decrease of uranium near the central void and a simultaneous increase of plutonium near the central void.

The columnar grain region of this section shows numerous metallic inclusions. Their composition (uncorrected) averages around 40% ruthenium, with under 10% molybdenum. Ruthenium, rhodium, technetium, molybdenum, and a very small amount of either palladium or silver were identified (in order of decreasing concentration). Large enough samples were analyzed to completely eliminate the beam penetration. No uranium or plutonium lines were excited, showing that these elements are not present in the inclusions. A plot of ruthenium and molybdenum concentrations in the inclusions against radial fraction is given in Figure 3.16. The inclusion nearest the unrestructured fuel, but still in the columnar grains, shows only half as much ruthenium as inclusions further into the columnar grains. The five inclusions examined in the unrestructured area showed no ruthenium, but did have 15 to 30% molybdenum. Four of these five inclusions did not appear as lustrous particles on the mosaic, but were found associated with voids or short cracks. This indicates they may be underneath the fuel surface or they may be a different physical form, such as an oxide.

FIGURE 3.15. Uranium and Plutonium Concentration Across Fuel Pin BNW 1-6, Section E, Path R1. A standard deviation of $\pm 1.2\%$ applies to each element.

FIGURE 3.16. Molybdenum and Ruthenium Composition of Inclusions in BNW 1-6, Section E

The results of analysis for molybdenum in the fuel matrix are shown in Figure 3.17. Molybdenum concentrations in the fuel matrix of 1 to 4% are observed in the unrestructured fuel, dropping to 0 to 1% in the columnar grains. Ruthenium analysis across a radius indicated no measurable ruthenium (under 0.15%). However, up to 0.3% ruthenium was found in pores in the unrestructured material.

Xenon was detected and its relative distribution across line R3 determined. The xenon counts (in 100 seconds) in the outer 700 μ is shown as part of Figure 3.18. Between 700 μ and the central void, the xenon count was zero. Since a xenon standard is rather difficult to examine under vacuum, counts per 100 seconds are plotted in place of percent in Figure 3.18. The outer two maxima observed are within the unrestructured fuel while the broad, but lower maximum appears to be just outside the unrestructured region. The drop off in counting rate out to 650 μ from the fuel OD corresponds to the fuel region showing numerous pores or voids. Within the area of the columnar grains showing well developed lenticular voids, the xenon count rate is essentially zero. Counting times of 100 seconds for each point, and also the background, with 0.23 microamp specimen current were used. A statistical counting error of about 150 counts is applicable to this data (standard deviation of 75 counts).

Analysis of neodymium, zirconium, and cerium across the radius (R3) was also completed. The distribution of these elements is also shown in Figure 3.18. Each of these elements shows highest concentration in the unrestructured fuel area where the burnup was highest. The concentration drops to a minimum about half-way across the fuel, then increases near the central void and approaches the count rate observed in the unrestructured fuel, with a few isolated points exceeding the unrestructured area. The form of these plots near the central

FIGURE 3.17. Molybdenum Concentration in the Fuel Matrix of BNW 1-6, Section E.

FIGURE 3.18. Neodymium, Cerium, Zirconium, and Xenon Distribution Across Fuel Pin BNW 1-6, Section E. The xenon count rate beyond 800 μ was zero.

voids appears similar to the plutonium distribution in Figure 3.15. The distance coordinates do not appear to be the same, but after allowing for the increased path length of R3 due to not passing through the center (see Figure 3.14), the increase of plutonium, neodymium, and zirconium begin at nearly the same radial fraction, and cerium begins just before plutonium increases. Considering the interval of analysis and the variation of the plot, all four components probably increase in the same area. The increase near the central void for each element, compared to the area half-way across the radius, is given in Table 3.6.

TABLE 3.6. Increase of Plutonium and Fission Product Concentration near Central Void Relative to Mid-Radius

<u>Element</u>	<u>% Increase</u>
Plutonium	~40
Neodymium	~40
Zirconium	~30
Cerium	~50

The correlation of position and concentration increase suggests that the increased fission products near the central void are simply due to fission of the increased plutonium at that location rather than transport of fission products. From the calculated burnup at the outer edge and center of the fuel (>200,000 and 38,000 MWd/MTM, respectively), one would expect a ratio of an indicator fission product at these locations of >5:1. The observed ratio of ~1:1 has not yet been explained.

At least 20 fission product elements have been identified in this pin. Table 3.7 lists the elements and the observed count rate (in the outer fuel area using a specimen current of approximately 10 microamps).

TABLE 3.7. Fission Products Identified in Fuel Pin
BNW 1-6, Section E

<u>Element</u>	<u>Count Rate for $L\alpha_1$ Line*</u> <u>(Approximately Proportional</u> <u>to Concentration)</u>
Strontium	200
Yttrium	100
Zirconium	1400
Molybdenum	3000
Technetium	600
Ruthenium	1000
Rhodium	300
Palladium	1000
Tellurium	100
Iodine	(a)
Xenon	(b)
Cesium	3000
Barium	1500
Lanthanum	1100
Cerium	3400
Praseodymium	2000
Neodymium	4500
Promethium	(b)
Samarium	1500
Europium	(b)
Gadolinum	(b)

- a. Calculated from $L\beta_1$ or other lines for lines interfering with the $L\alpha_1$ line. Counting rates above and below iodine are not comparable; different detectors were used in these regions.
- b. These elements were identified using point-by-point counting with high specimen current and long counting times (up to 100 seconds).

A pore near the cladding showed a lining of iron and nickel, with the interior of the pore nearest the cladding containing barium and cerium, as illustrated in Figure 3.19. The iron and nickel in the lining of the pore show the pattern of maximum iron concentration, then maximum nickel concentration with distance from the cladding. The maximum concentration of both iron and nickel observed was about 30%. Barium and cerium in the pore show some separation in the direction perpendicular to the pin radius. Barium shows a much higher concentration on one side of the pore than on the other, while the cerium concentration is highest on the side with low barium, as shown in Figure 3.20. Along the centerline of the pore (parallel to the pin radius) barium and cerium concentrations increase in an approximately parallel manner toward the center of the pin. The temperature in the area of the pore was about 1200 °C (calculated). The barium-cerium deposits do not show a metallic luster, and previous samples have shown an indication they are nonmetallic. The barium and cerium fission products should be present in the same ratio whenever they appear, unless one deposit is older than the other. The different ratios of barium to cerium may indicate deposition on one side of the pore early during irradiation, then deposition on the other side during the remainder of the irradiation, allowing that first deposited to decay and change the relative amounts of barium and cerium. Vapor phase transport is not likely since the analysis was made along what should be an isothermal line, i.e., along an arc approximately centered at the pin center.

An iron-nickel streamer extending from the cladding into the fuel is shown in Figure 3.21. The specimen current display shows a gray area indicating something unusual. The iron and nickel deposits were found bordering this area. The gray area contained the following elements in order of the X-ray

Neg 700504-4

FIGURE 3.19. Specimen Current, Iron, Nickel, Barium, and Cerium Distribution in a Void near the Fuel-Cladding Interface of BNW 1-6, Section E. Isolated Dots and Lines on Iron and Cerium Displays Are Due to Background Noise. Dark Area in Lower Left-Hand Corner of 300X Specimen Current Display Is the Cladding (1000X Is 80×80 Microns)

FIGURE 3.20. Barium and Cerium Counting Rates (Proportional to Composition) Across a Deposit of Gray Phase in BNW 1-6, Section E.

Specimen Current

Iron

Nickel

Cesium

Neg 700504-1

FIGURE 3.21. Specimen Current, Iron, Nickel, and Cesium Distribution on a Streamer Extending from the Cladding into the Fuel of BNW 1-6, Section E (80 x 80 Microns)

$L\alpha$ or $K\alpha$ line intensity: iron, nickel, palladium, tellurium, cadmium, molybdenum, barium, silver, cesium, silicon, and possibly potassium.

Chromium migration from the cladding onto the fuel surface was observed in this section. Figure 3.22 shows the separation of chromium from the cladding and deposition on the fuel. No iron or nickel was found associated with the chromium removed from the cladding. Many examples of iron and nickel migration from the cladding into the fuel have been seen, but very little chromium has been associated with the iron and nickel. The most likely explanation for the difference in migration behavior of these elements appears to be the large difference in the vapor pressure of their iodides. Fission product iodine reacting with the cladding forms iron, nickel, and chromium iodides, which then diffuse through the gas phase down the gradient of chemical potential of the metal iodide into the fuel, and then decompose in the hotter regions. Chromium iodide does not migrate very far since its vapor pressure is about a factor of 300 less than iron and nickel, giving a smaller gradient of chemical potential along the path into the fuel. From the above model of iodide migration, one would expect chromium to migrate only a very short distance (compared to iron and nickel) due to the variations in the gradient of chemical potential with distance. This deposit of chromium lends support to the above mechanism of transport of cladding components into the fuel.

The cladding of this section showed bright and dark inclusions. The bright inclusions were fuel, probably deposited during the polishing operation. The dark inclusions were found to be rich in manganese. Figure 3.23 is a concentration profile across one such inclusion, showing iron, nickel, chromium, and manganese. Small amounts of molybdenum, titanium, copper, and vanadium were also identified.

Neg 700504-3

FIGURE 3.22. Specimen Current, Iron, Nickel, and Chromium Distribution in a Void near the Fuel-Cladding Interface of BNW 1-6, Section E. Arrow in Iron Display Indicates Traverse in Figure 3.23
(260 × 260 Microns)

FIGURE 3.23. Concentration of Iron, Manganese, Nickel, and Chromium Across a High Manganese Inclusion in the Cladding of BNW 1-6, Section E. Fuel-Cladding Interface is at $\sim 125\mu$.

Gamma Scanning of Irradiated Pins BNW 1-10 and 1-12

Gross gamma scan traces of BNW 1-10 and 1-12 each show several peaks indicating concentrations of gamma emitting isotopes. It is interesting to compare these gamma scans made at an estimated 92,200 and 96,500 MWd/MTM for BNW 1-10 and 1-12, respectively, with the gamma scans made on these two capsules during an interim examination in March 1969 at approximately 55,000 MWd/MTM (estimate) burnup for both capsules. At the lower exposure BNW 1-10 showed a high narrow peak at 1-3/4 in. from the top of the fuel. This peak has disappeared completely on the scans at 92,200 MWd/MTM (estimate). A number of smaller peaks shown on 1 to 10 at the lower exposure also do not show up at the proper axial positions on the scans at higher exposure. Three peaks at 0 to 1.5 in. from the bottom of the fuel can be seen at corresponding positions on both gamma scans. Examination of neutron radiographs made at 55,000 MWd/MTM (estimate) on 1 to 10 showed no observable concentration of materials in the central void of the fuel.

A large peak is located at the bottom of the fuel column on both gamma scans of BNW 1-12. At 96,500 MWd/MTM (estimate) the peak is broader than is found on the scan at 55,000 MWd/MTM (estimate). Three other broad activity peaks at 5, 6.5, and 8.5 in. from the bottom of the fuel correlate as to axial position on both gamma scans. There are two activity peaks at the top of the fuel at the higher exposure that do not show on the scan at the lower exposure.

The disappearance of some peaks found on gamma scans of BNW 1-10 at 55,000 MWd/MTM (estimate) and the appearance of other peaks on the gamma scan at 92,000 MWd/MTM (estimate) and the broadening of peaks on BNW 1-12 shows that significant migrations of fission products has occurred in the fuel. Detailed gamma spectra data collected at several axial positions are needed before these capsules are returned to the ETR

for further irradiation to establish which fission products are migrating and where they migrate.

Analysis of LASL Profilometer Data

To obtain an estimate of the error on diameter measurements made by LASL of irradiated BNW fuel pins, an analysis was made on data from a special profilometer run conducted by LASL at the request of BNW. The measurements were made on a profilometer standard having five steps.

<u>Step</u>	<u>Diameter Range, in.</u>
1	0.3019 - 0.3020
2	0.2996 - 0.2998
3	0.3050 - 0.3051
4	0.3022 - 0.3023
5	0.3022 - 0.3023

The diameter range for each step was determined and certified as traceable to NBS standards by a measurement and inspection group at LASL. The following measuring procedure was used. The standard was positioned in the profilometer instrument where it remained for the duration of the measurements. The two profilometer linear transducer styluses at 90° to one another were positioned on the bottom step, and the two recording instruments were brought to zero. Ten profiles were obtained for both upward and downward travel of each stylus, with the instruments being brought to zero each time on the bottom step. This is the LASL standard operating procedure. Two diameter measurements are then obtained simultaneously making a total of 40 profiles for each step on the standard.

This data was analyzed statistically by T. M. Beetle of the BNW Mathematics Department to determine if significant variations occur in diameter measurements for:

- Runs made up or down.

- Different transducers and recorders.
- Different steps on the standard.
- Different measurement runs or profiles.
- Determination of random variation for all of the measurements.

Although there were statistically significant variations for these parameters the maximum was 0.00003 in. The variation in the measurements due to random error was determined to be about 0.00006 in. Thus each diameter selected from a LASL profilometer trace has a standard deviation of 0.00006 in. assuming that the profilometer has been calibrated with an accurately known standard. The error for any fuel pin diameter at the 95% confidence level is ± 2 standard deviations or ± 0.00012 in.

Modifications to BNW 324 East Cell Profilometer

The remote profilometer for diameter measurements of irradiated fuel pins is being modified. All wiring for the electric motor controls and drive for vertical movement and rotation of the fuel pins has been completed. Startup and testing of the drive has been curtailed until a misaligned bearing on the pin rotation chuck can be replaced. Two new matched linear displacement transducers have been obtained. These are expected to improve the capability of the electronic measuring system to measure diameters to greater than ± 0.0001 in. accuracy.

Two diameter standards for use in calibration of the profilometer were received. These standards were machined to give diameters of 0.245, 0.250, 0.255, 0.233, 0.238 and 0.243 in., accurate to ± 0.00002 in. They have been certified by the BNW Secondary Standards Laboratory with secondary standards traceable to the National Bureau of Standards.

FUEL SWELLING AND ANALYTICAL MODELING

K. R. Merckx and J. A. Christensen

Material Characterization and Swelling Model EvaluationK. R. Merckx, J. L. Daniel, S. J. Mayhan, W. L. Nicholson
and H. A. Triebs

Work is in progress to develop the quantitative metallographic techniques required to characterize the structure of both unirradiated and irradiated mixed oxide fuel materials. These structural characterizations are formulated for use in interpreting the effects of irradiation in terms of existing calculational models for the restructuring, bubble migration, and the swelling of mixed-oxide materials. The present studies are directed toward evaluating the swelling mechanisms associated with bubble formation within the grains, the subsequent migration of the bubbles to the grain boundary, and the growth of the grain boundary bubbles.

Detailed scanning electron microscope examination of fast reactor fuel materials in cooperation with the FFTF Fuels Evaluation and Special Products Fabrication Sections has continued with the examination of a second replica from an irradiated fast reactor fuel pin (PNL 1-17) and direct examination of an unirradiated mixed-oxide fuel pellet (MEE-63-4). In addition, specifications have been written for the preparation of specimens from four irradiated fuel pins, PNL 1-19, PNL 2-19, PNL 59-5, and PNL 59-7. These specimens are being prepared under closely controlled conditions for comparison of microstructure in fuels irradiated in a fast neutron flux with similar specimens irradiated in a thermal neutron flux.

A second replica from an irradiated fuel pin (PNL 1-17), prepared by LASL for routine metallographic examination, was examined and micrographs taken at preselected locations. These included ten positions along one radius at 0.25 mm intervals

and also at the cladding edge, midradius, and center axis along additional radii. As in the first replica studied previously, the microstructure and porosity appear consistent on each axis. However, comparison of grain size and porosity distribution show marked differences between the first replica (PNL 1-17-E) from the high flux region at the center of the fuel pin. This series is being evaluated by the FFTF Fuels Evaluation Section. Grain growth and accumulation of larger pores in the grain boundaries with depletion in the adjacent zone, and clusters of smaller pores in the grain interior (Figure 3.24) is evident. These observations support the relevance of the grain boundary swelling model selected for evaluation by this task.

Since primary data will be provided by the micrographs obtained from direct examination of plastic replicas in the scanning electron microscope, carefully controlled and documented preparation of samples is necessary to ensure accurate quantitative evaluation and comparison of void sizes and distribution in specimens irradiated under different conditions. Specimens irradiated to similar burnup levels in both a fast and a thermal neutron flux are available as shown in Table 3.8.

TABLE 3.8. Specimens Irradiated in Both a Fast and a Thermal Neutron Flux

<u>Capsule</u>	<u>Burnup</u>	<u>Linear Power Rating</u>	<u>Reactor</u>
PNL 1-19	8,700 MWd/MTM	9 kW/ft	EBR-II
PNL 2-19	47,000 MWd/MTM	9 kW/ft	EBR-II
PNL 59-5	9,500 MWd/MTM	10 kW/ft	GETR
PNL 59-7	48,000 MWd/MTM	10 kW/ft	GETR

These specimens are being prepared in the PNL Radiometallurgy laboratory. Close control, frequent visual inspections, and documentation of all preparation procedures will provide a guide for subsequent examinations of irradiated fuel specimens.

Plastic Replica; Irradiated Fuel
Pellet. Scanning Electron
Micrograph.

Neg 3416

1500X

FIGURE 3.24. Micrograph of Area Midway Between Cladding
Edge and Center Axis of Pellet from Irradi-
ated Fuel Pin (PNL 1-17-E). Negative PVC
Plastic Replica Has Been Coated with an
Evaporated Gold Film

The reference materials from which these fuel pins were fabricated, MEE-12 and ME-22, is also being characterized. A review of the standard characterization techniques is being made to determine if specialized characterization techniques will be required for the model evaluation studies to be conducted by this task.

An unirradiated mixed oxide fuel pellet (MEE-63-4) has been examined for the Special Products Fabrication Section by both electron microprobe and scanning electron microscopy. Initial examination in the microprobe included traces along three axes and micrographs taken at ten locations along each axis. Since the magnification limit (1000X) and the resolution of the microprobe are insufficient for identification and characterization of submicron pores, examination was continued in the scanning electron microscope. There was close agreement between micrographs from both instruments but the higher magnification and better resolution of the SEM still showed no submicron pores, indicating either their absence or the need for etching to reveal the fine surface structure. Comparison of scanning electron micrographs taken in the same area before and after etching in dilute nitric acid-cerium nitrate solution, shows many submicron pores and details of the fine surface structure (Figures 3.25b and c). Figure 3.25 also shows the comparison between micrographs made in each instrument (Figure 3.25a). The effect of different conducting thin films evaporated on the specimen surface is also being evaluated to determine if the use of gold or carbon contributes to the differences in resolution.

The characterizations carried out on this project are providing the background for the specialized developments required for future quantitative evaluations of microstructural features. The photomicrographs from these studies are the observational input for developing new quantitative

1000X

As-Polished

a. Micrograph from Electron Microprobe

Neg 3055

1000X

As-Polished

b. Scanning Electron Micrograph

Neg 3460

1000X

As-Polished

c. Scanning Electron Micrograph

FIGURE 3.25. Micrographs of the Same Area in Specimen MEE-63-4, As-Polished (a. Microprobe; b. SEM) and After Etching (c. SEM). The As-Polished Specimen Was Vapor Deposited with Carbon. The Etched Sample Was Gold Coated

characterizations of microstructure and provide the bases for establishing the limitations for making such quantitative characterizations. It has been demonstrated that the continuous magnification ranges required for statistical analysis of pore size and distribution can be readily achieved by sequential examinations using optical metallography, electron microprobe, and scanning electron microscope techniques. Similar examinations of process samples, using replica methods when necessary, will provide additional data for both the preirradiation porosity characterization and fuel swelling and analytical modeling programs, and will provide a means for comparing unirradiated and irradiated fuel materials.

An idealization of one portion of the swelling model used in the EXPAND computer code has been made for subsequent quantitative statistical evaluation. This idealization assumed that the intragranular bubbles are nucleated on dislocations with a uniform spacial distribution and that they obtain a steady-state size distribution during the early stages of irradiation. Attainment of this steady-state size distribution should be possible through adaption of the techniques used by Baroody⁽⁷⁾ or Gruber.⁽⁸⁾ Bubbles leave their nucleation site when they reach the critical size and breakaway from the dislocations. This critical size is a function of the temperature and the temperature gradient. Since the size distribution is assumed to be stationary and bubbles break away at a critical size, the rate of bubble break-away for migration to the grain boundaries depends upon the gas generation rate. The bubbles that migrate to the grain boundary are assumed to be immobile once they reach the grain boundary. No coalescence of bubbles during the migration is assumed, but coalescence of the bubbles as they reach the grain boundary occurs if the centers of the bubble are within a fixed region of influence.

The uniform spatial distribution of pinning sites, fixed size for bubble break-away, and stationary size distribution of intragranular bubbles results in a flux of bubbles migrating to the grain boundary. The arrival of the bubbles at the grain boundary can be described by a Poisson process. The total number of bubbles arriving per unit area of grain boundary is directly proportional to the total gas generated for the projected volume of the grain and is also proportional to the direction cosine between the temperature gradient vector and the normal to the grain boundary surface. For the proposed model of grain boundary bubble growth by only coalescence, all the bubbles are visualized as striking the grain boundary. The probabilities of no coalescence, of one or more bubbles coalescing, of two or more bubbles coalescing, etc., are determined by considering the probability distribution of nearest neighbors, next nearest neighbors,⁽⁴⁾ etc. Let:

- m bubbles arrive per unit area.
- R_1 be the radius of interaction for two bubble centers.
- R_2 be the radius of interaction for three bubble centers if two bubbles have interacted.
- N be the number of bubbles interacting.
- $P \{ \cdot \}$ be the probability of interactions associated with any randomly selected bubble.

For the assumed interaction model, the resultant probabilities of the selected bubble interacting with additional bubbles are:

$$\begin{aligned}
 P \{N = 0\} &= \exp (-\pi m R_1^2), \\
 P \{N \geq 1\} &= 1 - \exp (-\pi m R_1^2), \\
 P \{N \geq 2\} &= 1 - \exp (-\pi m R_1^2) - (\pi m R_1^2) \exp (-\pi m R_2^2).
 \end{aligned}$$

These probabilities of interaction can be determined for any number of interactions. Once the number of interactions is

determined, then the size distribution can be determined by using the equilibrium conditions between surface tension, internal gas pressure, external mean stress, and conservation of fission gas.

When the bubbles that have migrated to the grain boundary are observed on a two-dimensional section, they may be categorized as being intragranular because the planar section may be inclined so as not to have the grain boundary passing through the intersection. A correction factor must be applied to account for the bubbles classified as intragranular which are really intergranular. If R voids are on grain boundaries and A voids are not on grain boundaries, the corrected total number estimates are $R(1 + 1/p)$ and $A(1 - 1/p)$, respectively. Here, p is the probability that an intergranular void appears on a grain boundary when the void is sectioned by a randomly positioned and oriented plane.

The value of p was calculated assuming intergranular pores are lenticular. Two alternative formulas for p were derived as the method of numerical calculation of p depends upon whether the pores are near spherical or not. Specifically,

$$p = \frac{a}{2} \sum_{j=0}^{\infty} B\left(\frac{3}{2}, \frac{j+1}{2}\right) b^j. \quad (1)$$

$$p = \frac{a}{b^2} \left[\frac{\pi}{2} + b - \sqrt{1 - b^2} \tan^{-1} \left(\sqrt{\frac{1 - b^2}{b}} \right) \right]. \quad (2)$$

where

$$a = \frac{2 \sin \theta_1 \sin \theta_2}{\sin \theta_1 + \sin \theta_2},$$

$$b = \frac{\sin (\theta_1 + \theta_2)}{\sin \theta_1 + \sin \theta_2}$$

and $B(x,y)$ is the complete beta function. Here, θ_1 and θ_2 are the cap angles which define the shape of the lenticular pore. Formula (2) is best for near disk-like voids and (1) for near spherical voids.

In-Reactor Swelling Measurements

F. E. Panisko

In-reactor studies are in progress which isolate effects of fuel density, temperature, temperature gradient, fission rate and burnup on fission gas swelling of UO_2 -25 wt% PuO_2 . Irradiated specimens will be characterized through the use of quantitative electron microscopy in terms of analytical swelling models which are also being developed under this task. The experiment consists of simultaneously irradiating multiple (~ 20) fuel wafers with various thicknesses and fission atom densities to achieve different temperature gradients, burnups and fission rates. Fuel pressure and ambient temperature are controlled by externally heating a pressurized NaK annulus which surrounds the fuel capsule* and by varying insulation thickness adjacent to individual wafers.

Fuel densities in initial experiments are 94% and >99% of theoretical, the former being obtained from stocks of standard EBR-II experimental fuel and the latter produced by hot-pressing. Porosity fraction is a variable because pores and voids are expected to provide internal accommodation for much of the swelling.

Fuel temperature will be above 700 °C, a reasonable fuel surface temperature for FTR, and below ~ 2000 °C. Above the upper limit, all fission gas is assumed to be released. Fuel temperature influences fission gas swelling because fuel creep and pore or bubble migration are thermally activated processes.

* For a description of the controlled temperature-pressure irradiation capsule, see BNWL-SA-583 or USAEC-CONF-660511.

At higher temperatures, pores migrate more rapidly up the temperature gradient toward the fuel center.

Temperature gradient will be varied between ~ 0 and 8000 °C/cm. The lower value will be used as a basic reference; the higher value represents a maximum expected for oxide fuels in fast reactors. The combination of higher temperature and temperature gradient provides the necessary energy and force for bubble migration by a surface diffusion mechanism.

The fission rate will vary from 1.0 to 4.3×10^{14} fissions per cm^3 -sec. These rates are approximately the minimum and maximum achievable with available fuel and test facilities. The fission rate is expected to be most important in determining the dynamic concentration of bubbles in transit up the thermal gradient. The higher the fission rate the greater the anticipated saturation value of steady-state swelling.

Burnups for a constant time in the reactor will be varied from 20,000 to 80,000 MWd/MTM to provide a range representative of expected FTR conditions.

Fuel temperature is expected to be one of the most influential variables. Meaningful measurements require accurate knowledge of fuel temperature. Uncertainties in such parameters as neutron spectrum, cross-section, and interface heat transport are reflected in unacceptable imprecisions in calculated temperature profiles. To correct this, a prototype capsule will be irradiated during April 1970 to provide both in-pile calorimetric data and postirradiation, ceramographic evidence of fuel operating temperatures. This test will also provide an overall appraisal of procedures for capsule fabrication and postirradiation examination. Twenty-seven, 94%-dense, UO_2 -25 wt% PuO_2 fuel wafers will be included in the first test. The fuel diameter is 0.2 in. and lengths are 0.01, 0.10 and 0.20 in. The fuel contains 0.71 (normal), 30, and

93.15% enriched uranium and 11% ^{240}Pu in the plutonium fraction. Fifteen of the fuel wafers have flat, polished surfaces which mate with flat, polished surfaces on adjacent molybdenum heat transfer plugs. This special surface preparation is expected to minimize the temperature drop across the fuel-molybdenum interfaces. To provide a comparison with standard surfaces, the remaining specimens are not polished. Table 3.9 gives dimensions, locations, enrichments, and surface conditions for each specimen included in the first capsule. Figure 3.27 shows a molybdenum specimen holder. The holder is 0.605 in. by 9 in. long with 18-0.210 in. diameter, threaded holes. Complete, three-dimensional thermal characterization will be provided by five Chromel-Alumel thermocouples positioned inside the specimen holder and four placed outside the stainless steel cladding in the NaK.

TABLE 3.9. Fuel Arrangement for Initial UO_2 -25 wt% PuO_2 Swelling Capsule

Capsule Position	Surface Preparation	Fuel Wafer Location, Composition and Length		
		Top (Left) Uranium Enrichment-Length (in)	Center Uranium Enrichment-Length (in)	Bottom (Right) Uranium Enrichment-Length (in)
1	None	93%-0.013	93%-0.013	93%-0.013
2	Polished	93%-0.013	93%-0.013	93%-0.013
3	None	N-0.013	N-0.013	N-0.013
4	Polished	N-0.013	N-0.013	N-0.013
5	None	93%-0.013	93%-0.013	93%-0.013
6	Polished	93%-0.013	93%-0.013	93%-0.013
7	Polished		N-0.200	
8	Polished		N-0.013	
9	Polished		30%-0.013	
10	None		30%-0.013	
11	Polished		93%-0.013	
12	None		93%-0.013	
13	None		93%-0.013	
14	---	No fuel	---	
15	Polished		N-0.100	
16	Polished		30%-0.100	
17	None		30%-0.100	
18	Polished		93%-0.100

Neg 700979-2

FIGURE 3.26. Molybdenum Specimen Holder for UO_2 -25 wt% PuO_2 Irradiation Swelling Experiment. Also Shown Are Thermocouples and Stainless Steel Clad.

MIGRATION BEHAVIOR IN MIXED OXIDES

C. A. Hinman

Oxide nuclear fuel materials have a relatively low thermal conductivity in the temperature range where they operate, resulting in very steep thermal gradients when run at high specific power levels. A thermal gradient results in potential field gradients which can be considered as driving forces for mass transport. There are two main interacting components of this force which are being investigated theoretically and experimentally. These are the electronic component, characterized by the electronic thermoelectric power (electronic heat of transport) and the ionic component, characterized by the ionic heat of transport.

Flux Equation

As a first approximation, the diffusion controlled mass transport in a thermal gradient can be described by a relationship which states that the mass flux is directly proportional to the sum of the forces which are imposed upon the diffusing species. The constant of proportionality is called the diffusion coefficient. This is stated mathematically by:

$$J = -D \Sigma(\text{Forces})$$

Diffusion Coefficient

In a system such as UO_2 or $(\text{U,Pu})\text{O}_2$ the diffusion coefficients highly depend upon composition. This can be understood by examining the form of the random-walk diffusion coefficient:⁽¹⁰⁾

$$D = \frac{1}{6} \Gamma \alpha^2$$

where Γ is the atomistic jump frequency and α is the actual jump distance. The jump distance depends upon the mechanism of diffusion and the crystal structure. The jump frequency

not only depends upon the mechanism of diffusion but more directly upon the defect structure of the oxide and is a probabilistic function of the ability of an ion to move from one position through an energy col to another. Since an ion cannot diffuse in a perfect lattice, defects must be present for mass transport. The probability of position change of the diffusing specie is directly related to the defect density. The defect structure is also directly related to the degree of nonstoichiometry and thus both the diffusion mechanism and the compositional dependency are highly dependent upon the degree and type of deviations from stoichiometry.

Experimental data for the diffusion of oxygen in UO_{2+x} are available⁽¹¹⁾ and the concentration dependencies have been and are being investigated.⁽¹²⁾ The treatment by Thorn and Winslow^(12a) and experimental data by Auskern and Belle^(12b) for hyperstoichiometric UO_2 was used in these mass transport calculations since they included concentration and temperature dependencies in the equation. The diffusion coefficient for oxygen in UO_2 is given by:

$$D(C,T) = 2.016 \times 10^{-2} e^{-\frac{1.52}{kT}} \times \left\{ (O/M - 2) + \left[(O/M - 2)^2 + 495 e^{-\frac{1.62}{kT}} \right]^{1/2} \right\}.$$

This equation is good only for $O/M \geq 2.0$ and cannot validly be applied to the hypostoichiometric case. (The coefficient becomes negative for some values of T and $O/M < 2$). There are no experimental diffusion coefficients in UO_{2-x} . It was assumed that for negative deviations from stoichiometry the value for the diffusion coefficient again increases because of an increase in the concentration of defects. The quantitative nature of this increase is unknown so that for the purpose of the calculation a mirror image of the compositional dependency of the equation for UO_{2+x} was used for UO_{2-x} .

Forces

The simple assumption that the flux is directly proportional to the magnitude of the summation of the forces is not altogether correct. The fundamentals of nonequilibrium thermodynamics⁽¹³⁾ state that there exist cross-coupling terms reflecting interactions between forces. However, for these purposes the cross-coupling terms will be neglected.

At the present time, three forces are being considered in the case of mass transport in oxide fuel in a thermal gradient. The first force is the chemical force or the classical concentration gradient as found in Fick's First Law for isothermal diffusion in a concentration gradient.

Chemical Force = ∇C .

In the case of thermal diffusion, when steady-state is achieved ($J = 0$) it is the ∇C_{ss} (steady-state) force which exactly balances the sum of any other forces present.

The general form for the force term is given by:⁽¹⁰⁾

$$\text{Generalized Force} = \frac{C}{kT} \nabla E$$

where C is the concentration, T is the absolute temperature, and ∇E is the potential energy gradient.

For the thermal gradient situation, the gradient of the temperature, ∇T , must be related to the forces generated by it. These ∇T forces at the present time are being divided into two separate components, the one resulting from electronic processes and the other resulting from ionic processes.

The ionic portion is characterized by an ionic heat of transport parameter (Q_i^*) and the mathematical form for this force is given by:

$$\text{Ionic Thermal Force} = \frac{Q_i^* C}{kT^2} \nabla T$$

where ∇T is the thermal gradient.

The electronic portion of the thermal force which is exerted on the ions can be described in terms of the electronic thermoelectric force which builds an electrical potential gradient across the fuel. This force can be described by:

$$\text{Electronic Thermal Force} = \frac{qC}{kT} \nabla \phi$$

where q is the charge on the ion and $\nabla \phi$ is the electrical potential gradient. The term $\nabla \phi$ can be given in terms of the thermoelectric emf, Θ_e , as:

$$\nabla \phi = \Theta_e \nabla T.$$

Using these equations the flux equation can be rewritten as:

$$J = -D \left[\nabla C + \frac{C}{kT} \left(\frac{Q^*}{T} + q\Theta \right) \nabla T \right].$$

Fryxell and Aitken⁽¹⁴⁾ have published a value of ~ 1 eV (~ 23 kcal/mole) for the heat of transport for O^{2-} in UO_{2-x} . Their experimental technique imposed a possible gas phase transfer and an electrical shorting of the electronic thermoelectric emf so that the value obtained may or may not represent the true ionic heat of transport for O^{2-} in UO_{2-x} . The data indicated little, if any, dependency of Q^* on concentration.

Bates and Dau⁽¹⁵⁾ measured the thermoelectric power of "nearly stoichiometric" UO_2 including both single and polycrystal specimens. They found a p-n switch in the sign of the thermoelectric power at about 1200 °C. The data obtained for the polycrystalline specimen were curve-fitted to the equation:

$$\theta = 2.021 e^{-0.00479T} - 0.00255. \quad (\text{volts}/^{\circ}\text{K})$$

Since these measurements were made in a constant (but undetermined) oxygen partial pressure and small thermal gradients, it might be assumed that these values are from electronic processes alone and that a negligible concentration gradient was formed.

All of the above parameters are inserted into the calculation in a cylindrical system where the initial O/M is set at 2.00 and a parabolic temperature gradient exists along the radial direction. The central temperature was taken as 2000 °C and the outer surface temperature taken as 1200 °C. The value for the thermal gradient increases linearly from zero at the center and reaches its maximum at the surface of 1600 °C/cm.

The signs for both the ionic and electronic thermal forces indicate that for the temperature range used, the oxygen ion flux would be directed from the hot center toward the cooler skin. However, in examining the magnitude of the two forces and comparing them with reasonable values for the resulting ∇C , it is found that the ionic thermal force is at least one order of magnitude larger than a reasonable ∇C and that the electronic thermal force is at least two orders of magnitude larger than a reasonable concentration gradient.

The reasons for these discrepancies are presently under investigation. The primary contributing factor is the limited knowledge of how the vital parameters depend upon both stoichiometry and temperature.

The interpretation of the presently available literature data is also under review. The derivation of the ionic heat of transport, Q_i^* , depends upon the experimental measurement of the steady-state O/M gradient which developed in UO_{2-x} which was maintained in a temperature gradient.⁽¹⁴⁾ In the

steady-state, the ionic flux disappears and a further assumption is made that the metal container for the experiment shorted out the electronic forces so that the third term in the flux equation is zero. Under these conditions, the relationship is given by:

$$\nabla C = -\frac{Q_i^* C}{kT^2} \nabla T.$$

Manipulation of this differential equation gives:

$$\frac{d \ln C}{d(1/T)} = \frac{Q_i^*}{k}$$

which implies that the slope of the $\ln C$ versus $1/T$ curve should be constant (under the above assumptions and, further, that Q_i^* is concentration and temperature independent) and yields a value proportional to the ionic heat of transport. However, the value of 1 eV obtained by Fryxell and Aitken was determined from the slope of the $\ln x$ (in UO_{2-x}) versus $1/T$ plot which gives a completely different result than the equation above. A replotting of Fryxell and Aitken's data in the form of $\ln(O/M)$ versus $1/T$ gives curved lines and much smaller slopes. The implications of this derivation are being investigated.

The thermoelectric power term is also being reviewed, as it is the dominant calculated force. It may be that there is a significant ionic thermoelectric emf contribution to the measured potentials. An experimental technique is being devised to investigate the nature of the electrical potentials developed in oxide fuel when subjected to both small and large thermal gradients.

REFERENCES

1. T. A. Lauritzen et al. Experimental Studies of Plutonium and Fission Product Redistribution in Fast Reactor Fuels, GEAP-4466. General Electric Company, APO, San Jose, California.
2. A. S. Bain. Microscopic, Autoradiographic, and Fuel/Sheath Heat Transfer Studies on UO_2 Elements, AECL-2588. Atomic Energy of Canada, Ltd., Chalk River, Ontario, June 1966.
3. Robert N. Duncan. "CVTR Fuel Capsule Irradiations," CVNA-153. Carolinas-Virginia Nuclear Power Associates, Inc., Charlotte, N.C., August 1962.
4. W. E. Baily et al. "Effect of Diametral Gap Size on the In-Pile Performance of Fast Ceramic Reactor Mixed-Oxide Fuel," Trans. Am. Nucl. Soc., vol. 9, no. 1, p. 40, June 1966.
5. C. N. Craig et al. Heat Transfer Coefficients Between Fuel and Cladding in Oxide Fuel Rods, GEAP-5748. General Electric Company, APO, San Jose, California, January 1969.
6. W. E. Baily et al. "Thermal Conductivity of Uranium-Plutonium Oxide Fuels," Nuclear Metallurgy Symposium, Camelback Inn, Phoenix, Arizona, October 1967.
7. E. M. Baroody. "Calculations on the Collosional Coalescence of Gas Bubbles in Solids," J. Appl. Phys., vol. 38, pp. 4893-4903. 1967.
8. E. E. Gruber. "Calculated Size Distributions for Gas Bubbles Migration and Coalescence in Solids," J. Appl. Phys., vol. 38, pp. 243-250. 1967.
9. H. R. Thompson. "Distribution of Distance to Nth Neighbor in a Population of Randomly Distributed Individuals," Ecology, vol. 37, pp. 391-394. 1956.
10. P. G. Shewmon. Diffusion in Solids. McGraw-Hill, New York, 1963.
- 11a. J. Belle. "Oxygen and Uranium Diffusion in Uranium Dioxide (A Review)," J. Nucl. Mater., vol. 30, pp. 3-15. 1969.
- 11b. J. F. Marin and P. Contamin, "Uranium and Oxygen Self-Diffusion in UO_2 ," J. Nucl. Mater., vol. 30, pp. 16-25. 1969.

- 12a. R. J. Thorn and G. H. Winslow. "Correlation of Thermodynamic Properties and Atomic Transport in the Uranium Dioxide Phase," Thermodynamics, vol. 2, p. 213. IAEA, Vienna, 1966.
- 12b. A. B. Auskern and J. Belle. J. Nucl. Mater., vol. 3, p. 267. 1961.
- 12c. P. Contamin and J. F. Marin. Rapport Semestreil No. 1, Periode du 1er Janvier au 30 Juin 1969. Dept. de Metal. de Grenoble, France.
13. S. R. de Groot and P. Mazur. Non-Equilibrium Thermodynamics. N. Holland Publishing Company, Amsterdam, 1963, or R. E. Howard and A. B. Lidiard. Rep. Prog. Phys., vol. 27, p. 161. 1964.
14. R. E. Fryxell and E. A. Aitken. "High Temperature Studies of Urania in a Thermal Gradient," J. Nucl. Mater., vol. 30, p. 50. 1969.
15. J. L. Bates and G. J. Dau. Unpublished Data. Battelle-Northwest, Richland, Washington, 1968.

4.0 IRRADIATION DAMAGE TO REACTOR METALS

IRRADIATION BEHAVIOR OF STRUCTURAL MATERIALS

R. P. Marshall

Irradiation Facilities Operation

A. L. Ward

The purpose of this phase of the program has been to provide for the timely accomplishment of irradiation, testing, and subsequent processing of the data obtained from a variety of reactor structural materials. Irradiation experiments in the ETR G-7 pressurized hot water loop have been discontinued and the in-pile pressure tube has been removed.

The experiments discharged from the G-7 loop for Cycles 103 and 104 are summarized in Table 4.1 along with estimated neutron fluxes and fluences. Table 4.2 gives the operating history of the ETR and the loop for Cycles 103 and 104.

In-Reactor Measurements

E. R. Gilbert and N. E. Harding

Irradiation-induced creep in Types 304 and 316 SS has been measured in a thermal reactor (flux-tailored and enhanced test facility in a Hanford K Reactor) at total neutron fluences up to 5×10^{21} n/cm² ($E > 10^{-10}$ MeV). These measurements extend the temperature range over which irradiation-induced creep has been observed up to 370 °C, which is within the operating temperature range for LMFBR and FFTF components. Irradiation-induced creep is pertinent to the design and performance of stainless-steel components subjected to large amounts of intense radiation in fast-reactor cores. In some components, irradiation-induced creep is expected to be beneficial by permitting stress relaxation in cases where thermal stresses and distortions caused by irradiation-induced swelling might otherwise seriously limit reactor performance.

TABLE 4.1. Summary of Experiments Discharged
at Conclusion of ETR Cycles 103
and 104

Experiment Designation	No. of Specimens	Specimen Type	Estimated Flux, $n/cm^2/sec$ (a)	Estimated Fluence, n/cm^2 (a)
GEH-20-264	2	A-542, DCB	4.5×10^{13}	2.2×10^{21}
GEH-20-265	2	A-542, DCB	4.5×10^{13}	2.2×10^{21}
GEH-20-333	72	S/S, Tensile	9.6×10^{13}	1.6×10^{21}
GEH-20-334	72	S/S, Tensile	7.2×10^{13}	1.2×10^{21}
GEH-20-326	72	S/S, Tensile	9.0×10^{13}	2.9×10^{21}
GEH-20-327	72	S/S, Tensile	1.1×10^{14}	3.4×10^{21}
GEH-20-289	18	S/S, Tensile	2.7×10^{13}	8.6×10^{20}
GEH-20-180	18	Ni-Base, Tensile	3.0×10^{13}	3.1×10^{21}
GEH-20-315	18	S/S, Tensile	1.1×10^{14}	3.0×10^{20}
GEH-20-354	9	S/S, Tensile	1.1×10^{14}	2.2×10^{21}
GEH-20-316	18	S/S, Tensile	1.1×10^{14}	3.0×10^{20}
GEH-20-312	20	S/S, Tensile	1.1×10^{14}	1.9×10^{21}
GEH-20-353	9	S/S, Tensile	6.0×10^{13}	1.2×10^{21}

NOTE: Total specimens discharged, ETR G-7 loop Cycles 103, 104 = 402.
Total specimens discharged, ETR G-7 loop to date = 4500.

a. Neutrons with energies greater than 1 MeV.

The data are shown in Figure 4.1 as irradiation-induced tensile strain per unit tensile stress as a function of total neutron fluence. The temperatures and stresses are sufficiently low that after initial loading, strains saturate within a few hours and further deformation is not observed in the absence of neutron irradiation. The creep rate induced by neutron irradiation can be represented in terms of a linear stress dependence, thus allowing the normalized strain parameter to be used. Four different capsules were used to generate these data as indicated in Figure 4.1. Strain transients which occurred during the tests, such as upon changing stress or temperature in Capsule IV-36, were subtracted out to allow plotting the continuous strain increase with neutron fluence.

TABLE 4.2. Summary of Operating History for ETR and G-7
Hot Water Loop - ETR Cycles 103 and 104

<u>Operating History</u>		<u>ETR Cycle Number</u>	
1.	<u>Reactor History</u>	<u>Cycle 103</u>	<u>Cycle 104</u>
	Start up ^(a)	7-10-69	9-7-69
	End of Cycle	8-28-69	11-7-69
	Megawatt Days	5875	8664
	Effective Days at 175 MW	33.57	49.51
	Number of Scrams ^(b)	7	10
	Number of Shutdowns	2	3
2.	<u>G-7 Hot Water Loop History</u>		
	Maximum Temperature	544	545
	Effective Days Above 200 °F	35.7	52.4
	Effective Days at Operating Temp.	32.2	47.2
	Operating Efficiency	94.1%	95.4
	Temperature Efficiency ^(d)	90.3%	90.2

a. Includes flux runs at low power.

b. Power drops below half of full power (175 MW) with immediate recovery.

c. Effective days of loop operation above 200 °F (95 °C) with respect to effective days of reactor operation at 175 MW.

d. Effective days of loop operation between 500 and 550 °F (260 and 290 °C) with respect to days of operation above 200 °F (95 °C).

Neg 700712-1

FIGURE 4.1. Irradiation-Induced Tensile-Strain per Unit Tensile Stress Versus Total Neutron Fluence for Austenitic Stainless Steels in a K Reactor Flux Tailored and Enhanced Facility

The creep strain in Figure 4.1 appears to follow a transient and a steady-state behavior. The 20% cold-worked Type 316 SS underwent more transient strain than the solution-treated Type 304 SS. This observation is in qualitative agreement with UK data which showed more transient irradiation-induced creep in 20% cold-worked than in solution-treated Type 316 SS for measurements made on helical springs at 100 °C in the DMTR.⁽¹⁾ The irradiation-induced creep strain can be represented by the McVetty Equation⁽²⁾

$$\epsilon = A \frac{\sigma}{E} (1 - e^{-\frac{\phi t}{B}}) + C \sigma \phi t \quad (1)$$

where ϵ is the irradiation-induced strain, A is the transient strain amplitude which is on the order of 0.1, σ is the applied stress, E is the elastic modulus, ϕ is neutron flux, t is time, B is a decay constant, and C is a rate constant which is on the order of 10^{-30} cm²/psi. Fluence in place of the time parameter in the original McVetty equation has been used by Hesketh⁽³⁾ to describe the irradiation-induced creep strain in the austenitic stainless steel alloy EN58B in the Herald Reactor at 43 °C.

Irradiation-induced creep experiments in the form of stress-relaxation tests have resulted in more stress relaxation in tension⁽⁴⁾ than in torsion,⁽⁵⁾ as shown in Figure 4.2. Likewise, comparison of the tensile irradiation-induced creep data in Figure 4.1 with data obtained on helical springs⁽⁶⁾ in the same temperature range show more creep per unit fluence per unit stress for the tensile test. In making the comparison, the differences in the state of stress were accounted for with the conventional equations for effective strain and effective stress.⁽⁷⁾ A relationship between strain rate and stress for an isotropic material was developed by Gilbert and Straalsund⁽⁸⁾ which could account for the observed differences between irradiation-induced creep in torsion and

in tension. Whereas the conventional equations for effective strain and effective stress are based on conservative volume or a Poisson's ratio of $1/2$, the derived relationship can be applied to the case where volume increases occur during creep and Poisson's ratio relating the plastic Young's modulus and the plastic modulus of rigidity can vary from $1/2$ for conservative volume to -1 for isotropic swelling. For the irradiation-induced creep data on austenitic stainless steels, the analysis indicated that large ratios of β to α as noted in Figure 4.2 reflect stress-induced volume changes during creep and about $5/6$ of the measured creep strain in the uniaxial tensile test is due to stress-induced swelling. Less irradiation-induced creep strain is observed in the pure shear torsion test due to the absence of a hydrostatic-pressure component which could increase swelling.

FIGURE 4.2. Fractional Stress Relaxation Versus Fluence as Measured in Tensile⁽⁴⁾ and Torsional⁽⁵⁾ Stress Relaxation Experiments and Compiled from Tensile Creep Experiments

A comparison of available in-reactor creep data is presented in Figure 4.3 for solution-treated austenitic stainless steels. Since these data were obtained in tests involving different states of strain and stress, the data were converted to effective strain and effective stress with the relationship described above which was not limited to conservative-volume creep.

The transient creep term in Equation (1) has been attributed to dislocation climb.⁽⁹⁾ Under irradiation, dislocations climb by absorbing more interstitials than vacancies. The application of stress leads to small departures from their equilibrium positions in directions which produce strain. The steady-state component has been associated with the collapse of vacancy cascades to dislocation loops,⁽³⁾ with stress providing a preference among the available planes upon which the loops may collapse. Since cascade production and loop formation depend primarily upon displacement cross sections and elastic properties of the alloys, austenitic stainless steels possessing similar displacement cross sections and elastic properties should display similar steady-state behavior.

The data shown in Figure 4.3 for DFR and K Reactor at comparable temperatures are in agreement; however, lower temperature data from Herald Reactor and DMTR show more creep. Thus it may be concluded that irradiation-induced creep decreases slightly with increasing temperature. This trend is opposite to that usually displayed by thermally-activated creep processes. Further analysis of these data which attempts to separate the temperature and the neutron-energy dependence is presented elsewhere.⁽¹⁰⁾

Neg 700712-2

FIGURE 4.3. Effective Strain per Unit Effective Stress Versus Neutron Fluence for Solution-Treated Austenitic Stainless Steels

Fracture Analysis

C. L. Hellerich

The purpose of this program is to gain an understanding of the deformation and fracture of the candidate materials for advanced LMFBR applications. Selected specimens from the tensile and uniaxial creep-rupture studies are being examined to determine the effects of fast neutron irradiation and environment on fracture behavior and to correlate any transitions in this behavior with the relevant mechanical property changes.

Work is currently in progress to determine the effects of aging temperature, test temperature, and irradiation conditions on the creep-rupture fracture of Type 316 SS. In order to isolate the role of aging temperature, specimens aged at 800, 900 and 1400 °F and creep-rupture tested at 1000 °F under a stress of 50,000 psi were examined in the scanning electron microscope (SEM). Test results for these specimens have been reported previously,⁽¹¹⁾ but for convenience are shown again in Table 4.3. Examination of the fractographs in Figures 4.4, 4.5 and 4.6 shows that a transition from an intergranular fracture to a transgranular, plastically dimpled fracture occurred as the aging temperature increased. Quantitative evaluation, Table 4.3, of the mixed mode behavior showed very little difference in the fracture of the 800 and 900 °F aged material; however, at an aging temperature of 1400 °F, the fracture was entirely plastic.

Close examination of the grain boundary facets of the 800 and 900 °F aged materials, Figures 4.4b and 4.5b, showed that they were void of any recognizable plastic fracture mechanisms, and the intergranular portion of the failure must be classed as brittle, intergranular fracture. High magnification metallography, Figure 4.7, of polished and cathodically etched longitudinal sections of the material

TABLE 4.3. Uniaxial Creep-Rupture Data on Type 316 Stainless Steel

Specimen No.	Aging Temp., °F for 3045 hr	Tested at		Creep Rate, in./in./hr	Rupture Time, hr	Total Elongation, %	Intergranular Fracture (a) %
		Temp., °F	Stress, ksi				
9317	800	1000	50		631.4	22.1	65-70
9157	900	1000	50	0.000014	450.8	22.1	60-65
9282	1400	1000	50	0.000433	377.7	42.22	0
9163	Sol. Treated	1000	40	0.000009	2795	13.9	65-70
9134	Sol. Treated	1100	32	~0.0001	1113.2	19.5	ND (b)
9319	800	1200	25	0.000685	193.4	39.8	5-10

a. Remainder is transgranular plastic dimpling.

b. ND - Not determined.

a

300X

b

1000X

Neg 701100-4

FIGURE 4.4.

Mixed Mode Fracture in Type 316 SS Solution-Treated 1950 °F/1 hr, Then Aged at 800 °F/3045 hr. Creep-Rupture Tested at 1000 °F, 50,000 psi, 22% Total Elongation. Fracture is Primarily Brittle Intergranular (70%). Remainder is Plastic Dimpling (30%)

a

300X

b

3000X

Neg 701100-3

FIGURE 4.5.

Mixed Mode Fracture in Type 316 SS Solution-Treated 1950 °F/1 hr, Then Aged at 900 °F/3045 hr. Creep-Rupture Tested at 1000 °F, 50,000 psi, 20% Total Elongation. Fracture is Primarily Brittle Intergranular (65%). Remainder is Plastic Dimpling (35%)

a**300X** **b****3000X**

Neg 701100-2

FIGURE 4.6.

Transgranular Plastic Dimpling in Type 316 SS Solution-Treated
 1950 °F/1 hr, Then Aged at 1400 °F/3045 hr. Creep-Rupture Tested
 at 1000 °F, 50,000 psi, 42% Total Elongation. Region of Plastic GB
 Separation Can Be Seen at Arrows in **a** but Fracture Is Predominately
 Well-Defined Transgranular Plastic Dimpling as Shown in **b**

a

100X

b

3000X

c

3000X

Neg 701100-8

FIGURE 4.7. Metallographic Longitudinal Section of Type 316 SS Solution-Treated at 1950 °F/1 hr and Aged at 900 °F/3045 hr. Creep-Rupture Tested at 1000 °F, 50,000 psi, 22% Total Elongation. Note Numerous Grain Boundary Microcracks Below Fracture Surface in a. Cathodic Etching Revealed Presence of a Continuous Platelet Form of Precipitate at the Grain Boundary Microcracks. b and c Show Platelets at Cracks Indicated by Arrows in a.

aged at 900 °F revealed the presence of uniform platelets of carbide precipitates at the grain boundaries. These were best revealed at microcracks below the actual fracture surface. It appears that the presence of this platelet form of precipitate at the grain boundaries leads to the formation of brittle intergranular microcracks during creep-rupture testing. Ultimate failure occurs by the link up of these microcracks by transgranular, plastic dimpling, as shown in Figures 4.4 and 4.5. In contrast to this behavior, the material aged at 1400 °F showed extensive carbide precipitation and agglomeration both at the grain boundaries and throughout the matrix, Figure 4.8. The formation of voids (dimples) around the precipitates is apparent in Figure 4.8b and demonstrates the dimpling process that leads to the plastically dimpled fracture in Figure 4.6. Also, grain elongation and voiding at grain boundary precipitates, indicative of a high ductility fracture, can be seen.

The observed transition in fracture behavior correlates well with an abrupt change in ductility (total elongation). It may seem a misnomer to call a fracture that has seen 22% elongation a brittle, intergranular fracture or to call a change in ductility from 22% to 42% abrupt; however, if one subtracts out the transient strain data, it can be seen that a significant portion of the total deformation occurred as time independent, plastic deformation during specimen load up. For example, a solution treated control tested at 50,000 psi and 1000 °F failed with a total elongation of 18.2% but 17.3% of this occurred as time independent, transient strain.⁽¹¹⁾ Thus, the specimen strained only ~1% during creep-rupture, a rather low ductility event. This is contrasted to the 1400 °F aged material which had ~14% transient strain but failed with a total elongation of 42%. In this case, the specimen strained

a

1000X

b

3000X

Neg 701100-1

FIGURE 4.8.

Metallographic Longitudinal Section of Type 316 SS Solution-Treated 1950 °F/1 hr and Aged at 1400 °F/3045 hr. Creep-Rupture Tested at 1000 °F, 50,000 psi, 42% Total Elongation. Note Uniform Nucleation and Agglomeration of Carbides Throughout the Matrix and at Grain Boundaries a. Voiding at Both Grain Boundary and Matrix Precipitates Is Indicative of Highly Deformed Structure.

~30% during creep-rupture. This marks a sharp ductility transition (1% to 30%) and correlates well with the observed fracture behavior transition.

To establish the effect of test temperature, the fracture surfaces of specimens tested at 1000, 1100 and 1200 °F were examined. As shown in Figure 4.9, these specimens also exhibited a transition in fracture mode from intergranular to predominately transgranular fracture by a plastic dimpling mechanism. It is of interest to note the cavities in the fracture surface of the 1200 °F test, Figure 4.9. These cavities resulted from the formation of grain boundary microcracks and subsequent plastic growth during the creep-rupture of the specimen. This sequence of events is indicated by the presence of grain boundary detail at the bottom of the cavities and is consistent with the model of microcracking at carbide platelets previously suggested.

Although it is difficult to pinpoint from these initial results just when the grain boundary platelet type of precipitates form, it is probable that they are forming during the actual creep-rupture test, especially in the solution treated and the 800 and 900 °F aged materials. Being a dynamic process which passes through the platelet stage to the formation of agglomerated carbides, the precipitation process is influenced by both the test temperature and the applied stress. It is possible that as the test temperature increases, homogeneous nucleation predominates and the precipitation process becomes so dynamic that it passes through the platelet stage, overaging to form agglomerated carbides before substantial microcracking and crack propagation can occur. Final failure then occurs as in the overaged (1400 °F aged) material. This sequence of events could lead to the mixed mode behavior and cavitation observed in the 1200 °F fracture. Work is currently being conducted to determine the precipitate morphology at the grain boundary microcracks.

a

100X

b

100X

Neg 701100-6

FIGURE 4.9.

Type 316 SS Creep-Rupture Fracture. a Mixed Mode Fracture at 1000 °F, 40,000 psi, 14% Total Elongation. Fracture Is Primarily Intergranular (70%) with Plastic Dimpling (30%) Linking Together the Brittle Intergranular Microcracks. b Plastic Fracture at 1200 °F, 25,000 psi. Fracture Is Primarily Transgranular, Plastic Dimpling. Cavities in Surface (see arrows) Formed by Plastic Growth from Intergranular Microcracks.

Work has also been started to determine the effect of irradiation on creep-rupture behavior. Specimens irradiated at 900 °F to a fluence of 1×10^{22} n/cm² ($E > 0.1$ MeV) and tested in creep-rupture (Table 4.4)⁽¹²⁾ were examined. The specimen tested at 1000 °F under a stress of 45,000 psi, Figure 4.10, exhibited a mixed mode fracture. However, as can be seen in Figure 4.10a, the mixed mode fracture occurred as two separate regions: one a predominately brittle, intergranular fracture, Figure 4.10b, and the other a predominately shear (transgranular, plastically dimpled) fracture, Figure 4.10c. The significance of this well-defined division of fracture mode is in describing the chronological sequence of events that led to final failure and in revealing the necessary occurrence of certain fracture processes. Because of the low ductility nature of the brittle, intergranular fracture, this region of failure necessarily developed during the early stages of fracture. The nearly total absence of any form of plastic fracture in this region, Figure 4.10b, is indicative of the relative ease with which grain boundary microcracks were able to link up and form a continuous, propagating flaw. Therefore, the sequence of events leading to fracture began with the early generation of intergranular microcracks which then linked up to form a propagating flaw covering ~35% of the specimen cross-sectional area. This resulted in an engineering net section stress of 69,000 psi, which is in line with the ultimate strength of the material (63,300 psi) at 1000 °F.⁽¹³⁾ At this point, final failure was by plastic instability resulting in the shear or transgranular, plastically dimpled fracture in Figure 4.10c.

The 1400 °F test, Figure 4.11, fractured by a single mode, intergranular. As seen in Figure 4.11a, oxidation of the fracture surface occurred during the test and resulted in the demarcation of microcracks that developed during

testing. As in the 1000 °F test, the microcracking is intergranular. Because all of the fracture surface was intergranular, however, final instability must have occurred by a rapid intergranular fracture process. Figure 4.11b is typical of the majority of the specimen and represents the region of rapid fracture. As can be seen, the failure is intergranular, but by a plastic dimpling process at the grain boundaries, i.e., a plastically dimpled, intergranular fracture.

TABLE 4.4. Uniaxial Creep-Rupture of Irradiated Type 316 SS

Specimen No.	Irrad. Temp., °F	Test Temp., °F	Stress, psi	Minimum Creep Rate, in./in./hr	Rupture Irrad.	Time, hr Control	% Elongation	Intergranular Fracture %
14B66-1	900	1000	45,000	0.000230	107.8	1032	5.8	35
14B66-5	900	1400	10,000	0.000222	146.5	1050	5.3	100

NOTE: EBR-II Fluence for both specimens is 1×10^{22} n/cm² (E > 0.1 MeV)

Thus, irradiation of Type 316 SS at 900 °F to a total fluence of 1×10^{22} n/cm² (E > 0.1 MeV) produced two main changes in the fracture behavior during creep-rupture. First, when compared to the solution treated material, increased crack propagation characteristics were readily apparent. Secondly, the fracture mode at high temperature (1400 °F) for both the slow crack propagation and rapid unstable fracture was intergranular, whereas at 1000 °F the fracture mode during instability was transgranular. This suggests that in the irradiated material there was a transition in fracture

a

35X

b

300X

c

300X

Neg 701100-7

FIGURE 4.10.

Mixed Mode Fracture in Type 316 SS Irradiated 900 °F to a Fluence of 1×10^{22} n/cm². Creep-Rupture Tested at 1000 °F, 45,000 psi, 6% Total Elongation. Note Well-Defined Regions of Intergranular (35%) and Transgranular (65%) Fracture in a. Intergranular Fracture is Brittle as Shown in b. Transgranular Fracture, c, Is by a Plastic Dimpling Mechanism.

a

100X

b

300X

Neg 701100-5

FIGURE 4.11.

Intergranular Fracture in Type 316 SS Irradiated at 900 °F to a Fluence of 1×10^{22} n/cm². Creep-Rupture Tested at 1400 °F, 10,000 psi, 5% Total Elongation. Note Oxidation Deposits in a Mark Location of Intergranular Microcracks. b Shows Typical Region of Rapid Intergranular Fracture. Note Dimples on Grain Facets Indicating a Plastic Dimpling Process at the Grain Boundary.

mode from transgranular to intergranular as test temperature increased, whereas in the control material the transition was from intergranular to transgranular. This contrasting behavior apparently was due to the complex interrelationship between the mechanical and metallurgical states of the material and the test conditions which together act to control the microcrack initiation process as well as the crack propagation rate and mode.

Finally, the observation of microcrack initiation and subsequent propagation in the creep-rupture tests of the irradiated materials points out a potential flaw sensitivity in these materials. Because of the importance of understanding the conditions responsible for this phenomenon, efforts directed towards defining the mechanical and metallurgical states as well as the stress conditions conducive to crack initiation and propagation have been started. These efforts have the objectives of:

- Experimentally demonstrating the crack propagation phenomena.
- Quantitatively defining the phenomena.
- Metallurgically describing the material state and its relationship to microcrack initiation and propagation.

The Stability of Voids in Neutron-Irradiated Nickel

G. L. Kulcinski, B. Mastel, H. E. Kissinger and T. K. Bierlein

As described in the previous quarterly report,⁽¹⁴⁾ voids observed in nickel after irradiation at 450 °C to a fluence of 3×10^{21} n/cm² (E > 0.1 MeV) are extremely stable during high temperature annealing. In fact, very little change in either the size or the number density of voids occurs in regions greater than 15 μ from a boundary during two hour anneals at temperatures to 975 °C. The overall picture of void annealing as revealed in the present study is

one in which the outside surface of each grain becomes denuded of voids while the inside of the grain retains the character of the as-irradiated sample. This "unique" mode of void annealing must be considered in the interpretation of post-irradiation-high-temperature mechanical properties of irradiated nickel.

A particularly significant observation was made in samples which had been annealed at 1050 and 1150 °C. During these high temperature anneals, all voids had disappeared, but cavities had formed on the grain boundaries as shown in Figure 4.12. Similar cavities were not present on the grain boundaries of the unirradiated controls after annealing at 1050 or 1150 °C. Careful re-examination of the irradiated samples which were annealed at lower temperatures revealed that grain boundaries in these samples were normal and contained no cavities whatsoever. The observed cavities have several features common to bubbles. They are generally spherical instead of polyhedral. They enlarge during annealing: from 200 Å after the 1050 °C anneal to 500 Å after the 1150 °C anneal. Although an accurate count of the cavities is difficult to obtain, approximately 5×10^8 bubbles/cm² of grain boundary were present after the 1150 °C anneal. If these cavities are filled with gas to balance the surface tension forces, then the number of gas atoms required to stabilize these cavities is ~1 ppm. This is close to the 3 ppm of helium atoms which would be produced by this irradiation for an (n,α) cross-section of 1.03b.⁽¹⁵⁾

Figure 4.13 displays the average void diameters determined by four small angle X-ray scattering analysis techniques and compares them to the average diameter determined by transmission electron microscope (TEM) studies. It is apparent that the methods of Roess and Shull⁽¹⁶⁾ and that of Porod⁽¹⁷⁾ give sizes

A)

UNIRRADIATED CONTROL

1 μ

B)

SAMPLE IRRADIATED TO 3×10^{21} n/cm²
(>0.1 MeV)

Neg 700262-6

FIGURE 4.12. Typical Grain Boundaries in Nickel After 2 hr
Anneal at 1050 °C, Showing Cavities on Boundary

Neg 700262-2

FIGURE 4.13. Comparison of Average Void Diameters in Irradiated Nickel Obtained by Small Angle Scattering and Transmission Electron Microscopy (TEM) Techniques

which are in particularly good agreement with the average diameters obtained by TEM. The average of the rectangular distribution of Roess and Shull shows the best agreement with the TEM results. It is particularly gratifying that the Roess and Shull and Porod diameters also reflect the observed TEM drop in void diameter from 420 to 360 Å after the 975 °C anneal. As expected, the Guinier diameter was always 10 to 20% higher than that determined by TEM studies. The diameters obtained from the method described by Harkness⁽¹⁸⁾ were consistently 20% smaller than the observed diameters. An important point of Figure 4.13 is that the SAS results substantiate the TEM result: the average void diameter is relatively constant at ~400 Å up to the point of complete annealing (between 975 to 1050 °C).

Considerable success was also obtained in matching the void size frequency distribution determined from SAS studies to that derived from TEM studies. Figure 4.14 compares both the Maxwellian and rectangular distributions with the distribution determined by TEM techniques. The areas under all three curves were normalized to 100%. The agreement is quite acceptable considering the completely different manner in which the TEM and SAS distributions are derived. The agreement between the SAS and TEM distributions after the higher temperature anneals was also quite good.

Finally, it was found that one can use SAS to obtain semiquantitative estimates of the void density after various annealing treatments. Figure 4.15 shows that SAS techniques tend to overestimate the void density by a factor of ~2, relative to the TEM values. Nevertheless, the general shape of the annealing curve is so similar to that determined by TEM that its shape is considered accurate.

Neg 700262-1

FIGURE 4.14. Comparison of Void Size Distribution Determined by Transmission Electron Microscopy with Maxwellian and Rectangular Distributions Fitted to X-Ray Scattering Results. Sample: As-Irradiated High-Purity Nickel, $3 \times 10^{21} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$) at 450°C .

Neg 700262-4

FIGURE 4.15. Densities of Voids in Irradiated Nickel as Determined by Small Angle Scattering (SAS) and Transmission Electron Microscopy (TEM) Techniques

It is felt that the present work shows that, at least for nickel, the void size distribution and the void number density can be determined by SAS techniques. While SAS techniques will never replace good TEM results, SAS measurements offer certain advantages. First of all, the time required to characterize a sample is considerably less than that required to properly thin a highly radioactive nickel sample, take proper pictures of the magnetic material, and perform the tedious task of measuring the size of several hundred voids in order to obtain an average size. An advantage of the SAS technique is that it averages over many voids ($\sim 10^{11}$) as compared to the several hundred customarily treated by TEM studies.

One of the disadvantages of SAS is that other unknown components of damage might contribute to the scattering. However, only a few transmission photomicrographs are required to settle this question. In the case of nickel, there were no other visible components of damage which could scatter X-rays and significantly alter the results. Other areas in which SAS is obviously lacking are the determination of the morphology of the voids and the detection of inhomogeneous void distributions such as grain boundary denuding. Therefore, it is felt that an in-depth study of voids in nickel after various thermal treatments can be accomplished most efficiently by combining SAS and TEM studies, the former for void size and density determinations and the latter for microstructural details and as a check on the density measurements.

It is worth noting at this point that the volume change, $0.64 \pm 0.14\%$, measured in the as-irradiated nickel by TEM techniques agrees quite well with the volume change from immersion density measurements made by Holmes,⁽¹⁹⁾ who reported a 0.7% volume change for Ni-270 irradiated in the same capsule as the foils used in this study.

There are two rather unexpected results in the annealing behavior of the voids in the nickel samples studied: the relatively high temperature (~ 1000 °C) required to remove all voids and the constancy of the average void size during the annealing treatments. Because both of these results were substantiated by results from three separate techniques, microhardness measurements, TEM, SAS, there is little doubt about the validity of these observations. However, the results are contrary to results reported by Brimhall and Mastel⁽²⁰⁾ who found that all the voids in nickel irradiated to a fluence of 1×10^{20} n/cm² ($E > 0.1$ MeV) disappeared at 800 °C and that the average size of voids increased with increasing annealing temperature.

Past studies of the annealing of voids in copper and aluminum^(21,22,23) suggest that for an isolated void with nearby vacancy sinks, the shrinkage rate can be described as

$$-dr/dt = \frac{D_s}{r} [\exp(\frac{2\gamma\Omega}{rkT}) - 1] \quad (2)$$

where r is the radius, D_s is the self diffusion coefficient (including structure factor), Ω is the atomic volume, γ is the surface energy, T is the absolute temperature, k is Boltzmann's constant, and t is the time.

Equation (2) assumes that whenever a vacancy is removed from one void it is absorbed by a vacancy sink and that the average void size should decrease with increasing temperature. An implicit assumption in Equation (2) is that the spacing between the voids and sinks is much smaller than the average spacing between the voids themselves and that the emitted vacancy does not significantly change the vacancy concentration (C_v) from the equilibrium value (C_v^e). However, when the spacing between the voids is much less than between the voids and sinks,

it is reasonable to expect that some voids will capture the vacancies emitted by other voids. Such a situation can exist in heavily neutron-irradiated metals and is thought to be the reason why previous authors have found that larger voids grow at the expense of smaller voids.^(20,24) This latter situation would result in an increase in average void size with increasing temperature. However, the constant average void size with increasing annealing temperature found in this study does not fit either of the above situations.

A clue to the unusual behavior of the voids may be found in a recent analysis by Bullough and Perrin⁽²⁵⁾ who treated the case of partially gas-filled cavities. The word cavities is used to distinguish between voids which contain only vacancies, and bubbles which are in mechanical and thermodynamic equilibrium with external restraints. These investigators found that the shrinkage rate of a cavity is given by

$$-\frac{dr}{dt} = \frac{1}{4} \frac{D_s}{a} \left[\exp \frac{E_I(\ell)}{kT} \right] \left[\exp \left(\frac{F_m}{kT} \right) - \frac{C_v}{C_v^e} \right] \quad (3)$$

where "a" is the interatomic spacing in the direction of the jump, $E_I(\ell)$ is the spacial attractive interaction energy between a vacancy and a void at a distance ℓ , usually taken as $3a/2$, and F_m is the mechanical force tending to shrink the cavity which is equal to $P + \frac{2}{r} - \frac{3nkT}{4\pi r^3}$, where P is the external pressure and n is the number of gas atoms in the cavity. Bullough and Perrin⁽²⁵⁾ considered the case of postirradiation annealing where the vacancy concentration depends on the rate at which vacancies can diffuse away from the cavity. For $P = 0$ they found

$$-\frac{dr}{dt} = \frac{D_s}{2a} \frac{\left[\exp \left(\frac{2\gamma\Omega}{rkT} - \frac{3n\Omega}{4\pi r^3} \right) - 1 \right]}{\left[1 + \frac{r}{2a} \exp \frac{-E_I(\frac{3a}{2})}{kT} \right]} \quad (4)$$

Comparing Equation (4) with Equation (3), one finds that two forces tend to slow down the shrinkage rate: (1) the pressure of the gas atoms in the cavity, and (2) the attraction between the vacancy and the cavity. Furthermore, one can see that for small r and large n the term $\exp[\frac{2\gamma}{rkT} - \frac{3n}{4\pi r^3}] \Omega$ can be less than 1, and it is possible for small cavities to grow and large cavities to shrink. Such behavior would result in a relatively constant average size and would tend to increase the thermal stability of the cavities. These observations are entirely consistent with the data presented in Figure 4.16, which illustrates the constancy of average void size during annealing up to 900 °C, along with the fact that annealing causes the maximum diameter to decrease and the minimum to increase. It is therefore reasonable to hypothesize that the gas atoms stabilize the cavities in nickel which has been irradiated heavily with neutrons.

Rather conclusive evidence for the presence of gas in the irradiated samples is offered by the microstructure shown in Figure 4.11. Since the amount of gas measured experimentally is consistent with that which could be produced by the (n,α) reaction in nickel, it is very likely that the gas is helium. We therefore conclude that the "voids" present in the irradiated nickel of this study are not true voids in the sense that they contain only vacancies, but that in fact the voids are partially gas-filled.

This conclusion would also explain the difference in annealing behavior between the nickel used in this study and that used by Brimhall and Mastel.⁽²⁰⁾ Our samples contain ~ 30 times more neutronically produced helium which would tend to increase the temperature stability of the cavities.

Neg 700262-3

FIGURE 4.16. Effect of Annealing Temperature on the Void Diameters Observed with Transmission Electron Microscopy Techniques

We should point out that the present work does not necessarily support the hypothesis that voids are nucleated by gas atoms. It is not possible at this time to determine whether the gas atoms entered the void after it formed or if they indeed nucleated the void.

Grain Boundary Stabilization

G. L. Guthrie and T. K. Bierlein

The purpose of this program is to study the recrystallization and grain growth characteristics of metals containing small concentrations of inert gases. It is hoped that grain boundary pinning can be achieved by small concentrations of inert gases at the boundaries. If the grain size is small initially and remains small due to boundary pinning, the ready proximity of boundary regions may provide convenient sinks for vacancies, thereby depressing the vacancy concentration during irradiation and resulting in reduced swelling.

Work has continued on the previously reported helium-ion-bombarded powder compacted by the Dynapack process. Some of the ion-bombarded Dynapacked samples have been included in a capsule which is to be irradiated in the EBR-II reactor. Meanwhile, out-of-reactor annealing studies of helium-injected Dynapacked nickel powder samples have continued, and helium gas content has been determined for several classes of specimens, including:

- (1) Dynapacked specimens exhibiting grain size stabilization under annealing.
- (2) Dynapacked specimens exhibiting a lack of grain size stabilization when annealed.
- (3) The injected but not-yet-compacted powders used to make the Dynapacked samples of (1) and (2).

From these studies, it appears that approximately 8×10^{-4} atm-cm³ of retained helium per gram of compacted powder results in a

stabilized grain size when 4 to 7 μ powder is used as a starting material. Such compacts resisted grain growth when annealed at temperatures up to 850 °C. Finished compacts of this helium density have resulted from Dynapacking of powders at 700 °C where the starting powders contained about 2.2×10^{-3} atm-cm³ of the helium per gram of nickel powder. Noticeably lower starting concentrations (approximate factors of 2) have resulted in incomplete or even negligible stabilization at the elevated temperatures of the anneals.

The gas quantities which appear necessary to achieve stabilization are low enough to be encouraging in regard to retention of reasonable mechanical properties. The atom fraction of injected gas is of the order of 0.3 ppm on a volume basis and is of the order of one atom in 15 at the grain boundaries.

Studies of retained gas in the ion-bombarded powder as a function of injection parameters have led to the conclusion that helium pressures of less than 7.5×10^{-2} Torr during ion-bombardment result in unreasonably long bombardment times to reach satisfactory injection quantities. Helium pressures in the range from 125 μ to 375 μ all appear satisfactory. Present evidence indicates a noncritical optimization at about 250 μ .

Total helium injection appears to be approximately proportional to injection current density for a fixed bombardment time. Increased injection is also noted at increased time, but the relationship does not appear to be directly proportional to the first power of the bombardment time. This may be due in part to recoil ejection of helium already imbedded earlier in the run.

With the present apparatus, only a small amount of the powder is exposed to the ion stream at any given time, and some of the current goes directly to the stirring mechanism. Under these circumstances, exposures of 0.4 mA for 1 hr at

250 μ have been adequate to achieve gas quantities typical of stabilization in powder loads of the order of 25 g total weight. However, it is anticipated that this exposure could be significantly decreased in a redesigned apparatus as the coulomb charge associated with the known required gas implantation is considerably less than the ampere-second quantity indicated by the bombardment-current signal.

In the helium gas release studies, release rate has been plotted as a function of time under conditions of linearly increasing temperature. This allows a qualitative measure of the binding energy of the imbedded gas or the depth of penetration of the gas. It is found that decreasing the gas pressure during bombardment and increasing the ampere setting of the bombarding apparatus both result in a delayed or higher temperature gas release, indicating deeper penetration of the helium in the imbedding process. Presumably, both correlations result from an increase in the potential difference between the powder surface and the point in space where the injected ion suffered its last inelastic gas collision prior to colliding with the metal particle.

IRRADIATION EFFECTS ON THE FRACTURE OF HEAVY SECTION PRESSURE VESSEL STEELS

C. W. Hunter and J. A. Williams

Specimen Irradiations

Specimen irradiations, including one in.-thick compact tension (1T CT) and tensile specimens, were conducted during Cycles 101 through 104 in the M-3 loop facility of the ETR reactor. The M-3 facility is capable of producing specimens irradiated to 1 to 2×10^{19} n/cm² ($E > 1$ MeV) at approximately 540 °F in one reactor cycle. The irradiation capsule was designed so that removal and replacement and positioning of specimens could be accomplished during reactor outages. The

capsule is also adaptable for use in irradiating other types of specimens. The total irradiations completed to date include 12 CT specimens of base material at approximately 1.5 to 2.5×10^{19} n/cm² (E > 1 MeV), 12 CT specimens of weld and HAZ to a fluence of approximately 1.5 to 2.5×10^{19} (E > 1 MeV), and 4 CT specimens of base material at 7 to 8×10^{19} (E > 1 MeV). A total of 64 tensile specimens have been irradiated to levels from 1.5 to 8×10^{19} n/cm² (E > 1 MeV).

At the completion of Cycle 104, the G-7 loop facility was removed from the ETR. This was of major significance to operation of the M-3 facility, since, through the use of heat exchangers, the in-pile G-7 hot water loop supplied the necessary heat input for maintaining temperature in the M-3 facility. Prior to removal of the G-7 tube, plans were initiated to install an electrical heating system in the M-3 loop facility for maintenance of test temperature. With the electrical heating system temperatures other than the current operating temperatures (500 to 510 °F) will be obtainable. To date, all designs have been completed, components ordered and installation is scheduled for Cycle 107 of the ETR during April.

The temperature distribution along the M-3 capsule for Cycle 101 has been previously described.⁽²⁶⁾ In Cycle 102 and beyond, the inlet water temperature was increased 5 to 10 °F. Considering this change as well as the axial temperature distribution along the capsule, the tensile specimen temperature range has been 500 to 510 °F and CT specimen range has been 530 to 545 °F, with the majority of the specimens in the upper portion of the respective ranges.

The M-3 capsule cycle flux monitors for Cycles 101 through 104 have been analyzed and are plotted in Figure 4.17. The flux monitors are iron, titanium and aluminum-cobalt. Additional values will be obtained and the fluence values refined as the analysis of flux monitors located in CT specimen holes is

Neg 700453-6

FIGURE 4.17. Fast Flux Distribution Over the Length of the ETR, M-3 Loop, HSST Specimen Irradiation Assembly for Reactor Cycles 101 Through 104

completed. The cycle flux monitors occupied the same position during Cycles 101 through 103, near the front of the capsule with respect to reactor center. During Cycle 104 the monitor was exchanged with a tensile specimen tube near the rear of the capsule, approximately 1-1/2 in. radial distance from the position of the previous cycles. Comparisons of flux data obtained by this method do not yield an interpretation of the radial flux gradient as was initially expected. Peak flux of Cycles 101 through 103 shown in Figure 4.17 varied a maximum of about minus 20% from the largest value. Based on other reactor data it was estimated that a decrease of as much as 30% may be expected between the monitor position of Cycle 104 and Cycles 101 through 103, which would have placed the flux values during Cycle 104 at least as low as the lowest values obtained during the previous cycles. The flux values obtained in the 104 position were, however, higher than those previously obtained.

The experimental gamma heat survey of candidate positions for irradiating a large specimen capsule in the ETR has been completed. The survey was prompted by the uncertainty of gamma heat data, a factor of major importance in the capsule design. For the farthest position from the core available the gamma heat rate would be 0.9 W/g at the front surface and 0.3 W/g at the back surface of a 4-in. thick specimen. Based upon earlier heat transfer calculations⁽²⁶⁾ this gamma heat rate would result in a temperature differential of approximately 100 °F in the specimen. In order to maintain 550 °F at the specimen center with this high gamma heat rate, a gas gap of only 0.002 in. would be required. These results and evaluations demonstrated that a lower value of gamma heat, such as that in the ATR, would be preferable. In the ATR, reflector positions are available with gamma heat rates of 0.3 to 0.1 W/g front to rear respectively through a 4-in. specimen as shown in Figure 4.18. Under these conditions the temperature differential through the specimen will be about 30 °F.

Neg 700453-3

FIGURE 4.18. Gamma Heat Distribution for Positions to Be Occupied by the ATR-4T Irradiation Facility. Curves Numbered 3, 4, 5, 6, 7 Are at the Front Plane and 8, 9, 10, 11, 12 Are at the Back Plane of the Fracture Specimens Shown in Figure 4.19.

Estimates of the flux in the candidate positions of the ATR have been made and flux measurements are in progress. The highest estimate of fast flux is 2×10^{12} n/cm²/sec ($E > 1$ MeV) based on theoretical attenuation from a known position. An estimate based on the ratio of fast flux to gamma heat also yields 2×10^{12} n/cm²/sec ($E > 1$ MeV) at the center of the 4T capsule position. Based on these flux values, two reactor cycles with rotation of specimens between cycles would yield a specimen exposure of 1.2×10^{19} n/cm² ($E > 1$ MeV). Ten cycles with rotation after five cycles will yield a fluence of 6×10^{19} n/cm² ($E > 1$ MeV). The proposed ATR-4T irradiation capsule is shown in Figure 4.19. With this arrangement of two capsules with two specimens each the following irradiations could be produced:

- 2 base plate, WR orientation, 6×10^{19} n/cm² ($E > 1$ MeV).
- 4 base plate, WR orientation, 1.2×10^{19} n/cm² ($E > 1$ MeV).
- 4 base plate, RW orientation, 1.2×10^{19} n/cm² ($E > 1$ MeV).
- 2 weld metal, 1.2×10^{19} n/cm² ($E > 1$ MeV).

Irradiations 2, 3 and 4 would run concurrently with irradiation 1. The lowest estimate of fast flux, obtained from the ratio of fast to thermal flux, is approximately 9×10^{11} n/cm²/sec ($E > 1$ MeV). Irradiations at this level would require twice as many cycles to attain the similar goal exposures discussed above.

The ATR-4T irradiation facility in Figure 4.19 will replace ten reflector position filler pieces. The capsule support structure will be approximately 9.5-in. $\times$ 22.5-in. $\times$ 57-in. long. Two capsules will be irradiated concurrently with each capsule containing two 4T fracture specimens, and the specimen grip holes and notches will be filled with tensile specimens, charpy specimens, flux monitors, and thermal monitors. The capsules

FIGURE 4.19. Schematic of ATR-4T Irradiation Facility

Neg 700453-2

will be instrumented with thermocouples. Temperature control will be obtained by regulation of gas mixture supplied to the capsule.

Base Material Studies: Annealing and Fluence Effects

A cursory examination was conducted of the recovery of irradiation damage of ASTM A533 B tensile properties. The objective of this study was to determine the temperature range in which significant recovery occurred and the extent of recovery which could be expected. As previously reported herein, the irradiation temperature of tensile specimens in the M-3 loop is between 500 and 510 °F while maximum temperatures at the center of the 1T CT specimens was between 530 and 545 °F. It was therefore desirable to determine if tensile properties at the lower irradiation temperature were similar to those which would be obtained at the higher temperature.

Tensile specimens irradiated to a higher fluence, $\sim 4.3 \times 10^{19}$ n/cm² (E > 1 MeV), were also tested and compared to the specimens which were irradiated at approximately 2×10^{19} n/cm² (E > 1 MeV). These will be supplemented with future tests to determine the effect of fluences to 8×10^{19} n/cm² (E > 1 MeV) on tensile and fracture properties.

The results of these studies are presented in Table 4.5 and Figures 4.20, 4.21 and 4.22. Tensile specimens irradiated to 1.7 to 2.4×10^{19} n/cm² (E > 1 MeV) at a temperature between 500 and 510 °F were annealed at 540, 600, 650 and 725 °F for 130 hr. The tensile specimens annealed at 725 °F had been previously annealed at 650 °F for 130 hr. The specimens were tested in a range from room temperature to 500 °F. As shown in Figure 4.20, approximately 70% of the change in room temperature yield strength due to irradiation was recovered after annealing at 650 °F. Increasing the annealing temperature to 725 °F did not yield any significant

TABLE 4.5. Tensile Properties of Irradiated A533, Grade B Class I Steel from HSST Plate 02, Longitudinal Specimens

Specimen Identification	Test Temperature,		Plate (a) Position	Fluence, $\times 10^{19}$ n/cm ² (E > 1 MeV)	Yield Strength, 0.2% Offset $\times 10^3$ psi	Ultimate Strength, $\times 10^3$ psi	Annealing (b) Temperature, °F
	°F	°C					
02GA 156	90	32	5/19	2.26	93.9	110.6	540
02GA 157	500	260	5/19	2.40	82.3	104.4	540
02GA 232	90	32	9/19	1.8	80.2	98.7	600
02GA 233	250	121	9/19	1.9	76.0	94.1	600
02GA 234	500	260	9/19	1.97	70.7	94.9	600
02GA 238	83	28	9/19	1.8	73.8	92.8	650
02GA 239	250	121	9/19	1.9	69.4	88.2	650
02GA 240	500	260	9/19	1.97	66.2	90.7	650
02GA 186	84	29	7/19	2.26	75.7	97.1	725 ^(c)
02GA 187	500	260	7/19	2.40	67.5	92.4	725 ^(c)
02GA 193	-100	-73	7/19	4.31	118.1	130.2	---
02GA 164	89	31	6/19	4.41	103.4	118.1	---
02GA 168	250	121	6/19	4.31	96.4	112.7	---
02GA 191	500	260	7/19	4.41	86.3	107.2	---

a. Fraction of thickness at specimen center line.

b. Annealing time, 130 hr.

c. Re-annealed after annealing 130 hr at 650 °F.

Neg 700453-1

FIGURE 4.20. Recovery of Room Temperature Yield Strength During Postirradiation Annealing for 130 hr. ASTM A533-B Base Material from HSST Program Plate 02.

Neg 700453-7

FIGURE 4.21. Yield Strength of ASTM A533-B HSST Program Plate 02 at Two Fluence Levels and After Postirradiation Anneal

Neg 700453-8

FIGURE 4.22. Ultimate Tensile Strength of ASTM A533-B HSST Program Plate 02 at Two Fluence Levels and After Postirradiation Anneal

additional recovery over the 650 °F anneal. No significant recovery of irradiated properties was obtained below 550 °F. The results of Figure 4.20 are typical of both the yield strength and the ultimate strength over the temperature to 500 °F, as demonstrated by Figures 4.21 and 4.22.

If indeed the change in yield strength is a measure of the change in fracture properties, then one could expect significant recovery of fracture properties at annealing temperatures as low as 600 °F. Similarly, it may be expected that appreciably less degradation in toughness will occur during irradiation at 600 °F. The fact that as much residual damage exists after annealing at 725 °F as at 650 °F suggests that much higher temperatures would be required to recover all the irradiation damage, if such an event is possible at all in the range of practical interest. Studies at NRL⁽²⁷⁾ reveal that Charpy-V notch ductility of 6-in., A302-B steel was observed to exhibit approximately 70% recovery after annealing at 800 °F for 168 hr. Varying degrees of recovery of notch ductility for a number of materials and conditions are reported elsewhere.^(28,29)

The lack of any observed recovery of tensile properties at 540 °F lend confidence to the results of the 500 to 510 °F tensile irradiations being closely representative of tensile properties which might be obtained when irradiating at 540 °F. If any significant difference were to be expected at the 540 °F irradiation temperature, then one would also expect a significant change as a result of postirradiation heat treatment at the 540 °F temperature.

The tensile specimens irradiated to a fluence of 4.3 to 4.4×10^{19} n/cm² (E > 1 MeV) were tested from -100 °F to 500 °F. Test results showed an approximately 6 to 7% increase in room temperature ultimate and yield strength over those properties observed after irradiation to 1.7 to 2.4×10^{19} n/cm² (E > 1 MeV).

Fatigue Crack Preparation in CT Specimens

In linear elastic fracture toughness determination, the critical stress intensity (K_{IC}) is that for fracture initiation from a sharp fatigue crack. Hence, the preparation of a fatigue crack in the fracture specimens is a necessary preliminary to actual toughness determinations. Consideration of the loading conditions during such fatigue crack preparation provides an opportunity to extract some information on fatigue crack behavior. To be sure, these loading conditions were not intended to provide pure propagation data, such as the crack growth rate per cycle (da/dN) versus cyclic stress intensity (ΔK) relationship developed by Paris.⁽³⁰⁾ Nevertheless, this information does offer some examples of and insight into the practical situations of crack initiation, stalling, and re-initiation, (incubation) as well as propagation.

The specimens were IT CT specimens. The materials were the same 12-in. thick, A533-B HSST program plate and subarc weldment used throughout this investigation. The base material specimens were RW orientation from quarter through center positions. The fracture planes of the weldment specimens were parallel to the weld and located at positions A, B, and C throughout the weldment. As previously described,⁽²⁶⁾ A is the center of the weld metal, B is the weld dilution region (on the weld side of the fusion line), and C is the austenitic grain growth region in the HAZ (on the base metal side of the fusion line). All load cycling was from zero to tension to provide the intended ΔK and was performed at 1800 cycles/min in air at room temperature.

The fatigue cracks were initiated from a machined notch of 0.002 to 0.005 in. radius and subsequently grown a total of 0.200 in. in several steps at different levels of ΔK . Three different patterns of stress intensity were employed; the various steps in each of these three patterns are listed

in Table 4.6. The crack lengths for each step in Table 4.6 are the distance from the point of load application to the crack tip in the specimens. All three patterns were used in the fatiguing of the base material specimens. Only Pattern III was employed with the weldment specimens. With Patterns I and II, the ΔK was substantially reduced in Step 2 after crack initiation in Step 1. Typically, this reduction greatly reduced the propagation rate and in some instances stalled it. In the specimens of stalled propagation, the higher ΔK of Step 3, Pattern II was employed to re-initiate propagation. After re-initiation, Steps 4 and 5 of Pattern II were similar to Steps 2 and 3 of Pattern I. Therefore, Patterns I and II were the same except that Pattern II incorporated a step to re-initiate the stalled cracks which sometimes occurred during Step 2. Stalling was not prevalent in Pattern III because the ΔK decrease from Step 1 to 2 was not so large.

TABLE 4.6. Basic Stress Intensity and Propagation Patterns Employed in the Fatigue Crack Preparation of 1T CT Specimens of A533-B Plate and Weld

Steps	Pattern I	Pattern II	Pattern III
1 (from machined notch)	26 ksi $\sqrt{\text{in.}}$ 0.78 to 0.80 in.	26 ksi $\sqrt{\text{in.}}$ 0.78 to 0.80 in.	26 ksi $\sqrt{\text{in.}}$ 0.78 to 0.80 in.
2	17.5 to 20 0.80 to 0.89 in.	17.5 crack propagation stalled at 0.80 in.	21 to 24 0.80 to 0.90 in.
3	16.5 to 19 0.89 to 0.98 in.	21 to 22 crack propagation restarted to 0.84 in.	17 to 19 0.90 to 0.97 in.
4		18 to 20 0.84 to 0.89 in.	13 0.97 to 0.99 in.
5		16.5 to 19 0.89 to 0.98 in.	

The corresponding values of effective crack growth rate and ΔK for each step are plotted in Figure 4.23 for the base material specimens and in Figure 4.24 for the weldment specimens. It must be emphasized that these values of crack extension rate per fatigue cycle include periods when the crack was initiating, stalled, and re-initiating as well as pure propagation. The typical relationship between ΔK and pure crack propagation is indicated in Figures 4.23 and 4.24.

The extent by which the data in Figures 4.23 and 4.24 fall below the band for typical propagation is a measure of the amount of hindrance provided by initiation, etc. The crack initiation in Step 1 from a machined notch is hindered a reasonable amount. The large ΔK decrease between Steps 1 and 2 (26 to 17.5 ksi $\sqrt{\text{in.}}$) in Patterns I and II always hindered extension. The ΔK decrease from Step 1 to 2 in Pattern III (26 to 21 ksi $\sqrt{\text{in.}}$) effected far less hindrance. Steps 3 in Patterns I and III and Step 5 in Pattern II were not appreciably hindered. With the base material specimens, Step 4 of Pattern III was not hindered, even though it involved a ΔK decrease from 19 to 13 ksi $\sqrt{\text{in.}}$. Apparently, both a ΔK decrease and a fairly high prior value of ΔK are requisites for re-initiation hindrance.

The behavior of the weldment specimens during Step 4 (13 ksi $\sqrt{\text{in.}}$) is quite varied. The specimens near the fusion line (areas B and C) exhibited cracking rates approaching typical propagation behavior. However, the cracking in the center of the weld metal (area A) was markedly hindered. A possible explanation for the hindered cracking in the weld metal may be the presence of rounded microinclusions which blunt the fine crack tip.

The direction which a crack wanders in the fusion region is also of potential significance. Table 4.7 lists the positions of the original machined notch and final crack tip in specimens near the fusion line (areas B and C). The distance,

Neg 700453-4

FIGURE 4.23. Effective Fatigue Crack Extension Behavior in 1T CT Specimens Subjected to Different Patterns of Cyclic Stress Intensity. ASTM A533-B Base Material from HSST Program Plate 02.

Neg 700453-5

FIGURE 4.24. Effective Fatigue Crack Extension Behavior in 1T CT Specimens in Weld Metal and near the Fusion Line. ASTM A533-B Submerged Arc Weldment from HSST Program Weldment 51A.

in mils, into either area B or C for both sides of the cracks are indicated. Based on these results, neither B or C areas offer a preferential path for fatigue cracks.

TABLE 4.7. Position of the Machined Notch and Fatigue Crack on the Two Sides of the CT Specimens with Respect to the Fusion Line

Specimen No.	Distance from Fusion Line, mils			
	Machined Notch		Fatigue Crack Root	
	B Weld-Dilution Region	C Region of γ Grain Growth	B Weld-Dilution Region	C Region of γ Grain Growth
51A 5021	5,10		5,9	
51A 5022	1	20		3,30
51A 5023	10,20		11,42	
51A 5024	1	10	0	18
51A 5025	1	1	4,15	
51A 5040	15,40		0,50	

FUEL SUBASSEMBLY MATERIALS

T. T. Claudson

EBR-II Irradiations

P. K. McDaniel

The EBR-II irradiations program is to provide coordination and direction for the timely accomplishments of fast reactor irradiations on structural materials.

Structural material irradiation experiments are in various stages of design, construction and irradiation in EBR-II. Several changes have been made in scheduled EBR-II experiments. The schedule for the FFTF prototypal materials subassembly and control rod swelling materials experiment (Pins BNW B41, B42 and B43) originally scheduled to be inserted in EBR-II by February 1, 1970, has been changed. Two pins of the original 3-pin experiment (Pins BNW B41 and B42) have been combined with BNW Subassembly 5 (metallurgical variables swelling experiment). Pin BNW B43 which is to contain the tantalum control rod alloys for swelling studies experiment is still pending final AEC approval.

Pin BNW 24, the third in a series of five swelling experiments directed at defining effects of cold work, fluence and temperature on Types 304 and 316 SS, Inconel 600 and Nickel 200 in Subassembly X021 will be removed at the end of run 40 after accumulating about 3×10^{22} n/cm² (E > 0.1 MeV). An identical pin, BNW 25, will replace BNW 24 for an exposure of about 1×10^{22} n/cm² (E > 0.1 MeV).

An updated summary of experiments in EBR-II is given in Table 4.8. The accumulative exposure values in the past have been total exposure to the center of the subassembly. These values are now given as accumulative exposure based on fluence for neutrons having energies greater than 0.1 MeV. The new fluence estimates are based on unfolded spectra from the run 31F dosimetry experiment.

Damage Analysis

J. L. Straalsund, J. F. Bates and H. R. Brager

The objectives of this effort are to establish the radiation-induced swelling characteristics of FFTF alloys, relate fast-reactor induced substructural changes in microstructure to corresponding changes in mechanical properties and to provide a basis for extrapolation and interpolation of data using both microscopic modeling and empirical approaches.

Recent experiments concerning the effects of fast-reactor irradiation on stainless steels have demonstrated that a considerable volume change occurs during neutron bombardment.⁽³¹⁾ Unlike volume changes in fuel materials, swelling in structural alloys and pure metals is due to the condensation of irradiation-produced excess vacancies in the form of three dimensional cavities. United Kingdom investigators have found as much as 6.8% volume change in Type 316 SS irradiated to 7.8×10^{22} n/cm² total fluence. Volume changes of this magnitude, if not taken into account, are sufficient to cause distortion in reactor components.

TABLE 4.8. Summary of Experiments in EBR-II

Subassembly Designation	Reactor Position	Date Charged	Estimated Discharge Date	Type of Specimens	Material	Accumulative Exposure, n/cm^2	Goal Exposure, n/cm^2
X018B	4E2	10-2-69	Oct.	70 Density	304, 316 SS	5.17×10^{21}	3.0×10^{22}
X020	6B5	1-13-67	March	70 28 Tensile 8 Uniaxial Creep	304 SS 316 SS	3.3×10^{22}	5.6×10^{22}
X021B(c)	2D1	2-27-67	Dec.	70 501 Tensile or Uniaxial Creep 65 Biaxial Creep	304 SS, 316 SS, 348 SS, 321 SS (d) Ni Base Alloys	5.47×10^{22}	1×10^{23}
Pin 24					Pin 24	2.68×10^{22}	3×10^{22}
X021	4B3	11-21-67	July	70 5 Tensile 3 Uniaxial Creep	304 SS	3.8×10^{22}	5×10^{22}
X041	7C4	7-24-68	June	70 713 Tensile or Uniaxial Creep 74 Biaxial Creep	304 SS, 316 SS, 321 SS, 348 SS	2.1×10^{22}	3×10^{22}
X057	2B1	2-24-69	March	71 577 Tensile or Uniaxial Creep 164 Biaxial Creep 58 Density	304 SS, 316 SS, (d) 321 SS, 348 SS, Ni Base Alloys	2.68×10^{22}	6×10^{22}

a. Accumulative to end of run 39C.

b. Fluences greater than 0.1 MeV.

c. Original X021 subassembly with two single pin interchanges. Pin 24 in now.

d. Variety of other alloys.

Maximum target fluences for FFTF are as high as 10^{24} n/cm². Since such a high fluence cannot be attained in a reasonable time using currently available reactors, design data for high exposures will be based on extrapolation using physically realistic swelling models. The approach taken in this study is to develop swelling data at fluence levels that can be obtained in reasonable times and to use this data to develop the required swelling models.

Density measurements have been completed on a pin (A4) which was used to contain tensile specimens for irradiation in Row VII (Subassembly X010 position 7F3) in the EBR-II. During irradiation the inside of the pin was exposed to helium and the outside to coolant sodium. The pin is made of mill annealed Type 304 SS. The results of the measurements are shown in Figure 4.25. In the figure, the density and estimated exposure are plotted as a function of distance from the midplane. The error bands represent the spread of 5 independent measurements for each points.

Two aspects of the results are different than had been expected:

- The maximum swelling was significantly less than would be obtained from the swelling equation⁽³²⁾ for solution-treated stainless steel. While the irradiation temperature is not very well known, it is certain that the temperature at the midplane was no less than the inlet temperature, 700 °F. Using 700 °F and a fluence of 3×10^{22} n/cm² (E > 0.1 MeV) in the swelling equation gives a value of 0.5% compared to the observed value of about 0.3%.
- The gradient in swelling along the axial length appears to be much less pronounced than one would expect. For example, the temperature may be expected to increase somewhat as one proceeds from the bottom of the pin to the midplane, but

Neg 0695652

FIGURE 4.25. Irradiation Dose and Swelling Profile
for Pin A4, Irradiated in Row VII in EBR-II

one may estimate a minimum expected change in swelling by assuming no change in temperature. Doing this and using the estimated fluences in Figure 4.25 with the fluence exponent of 1.7 in the swelling equation, one may calculate a value of 2.6 for the minimum expected ratio of swelling at the midplane to swelling 8 in. below the midplane. As may be seen from Figure 4.25, however, very little difference exists in the amount of swelling observed at the two positions.

The above features may possibly be due to a flux rate effect or perhaps a spectrum effect. It is noteworthy that this is the first set of data that we have obtained from Row VII in the EBR-II. For positions this far from the core center, the flux is much lower, and the energy spectrum is considerably different. However, the irradiation conditions and material characterization need to be defined more completely before any meaningful interpretations may be made.

Empirical equations are presently being developed to describe the effects of neutron exposure and irradiation temperature on void sizes and concentration in Types 304 and 316 SS. The purpose of this work is to develop a set of three self-consistent equations describing the void diameter, void concentration, and void volume, or swelling, over as wide a range of experimental conditions as possible. A consistent set of equations accounting for all of the observed trends will provide more confidence in extrapolating to the higher exposures expected in the FTR. The best fits obtained to date for the void concentration data developed at PNL are:

$$\rho = (3.31 \times 10^{-5}) \left(\frac{\phi t}{10^{22}} \right)^{0.90} (\rho^{7030}/T - 1.0). \quad (1)$$

$$\rho = (1.21 \times 10^{-5}) \left(\frac{\phi t}{10^{22}} \right)^{-0.45 + 0.0019T} (\rho^{7740}/T - 1.0) \quad (2)$$

where

$$\phi t = n/\text{cm}^2 \quad (E > 0.1 \text{ MeV})$$

$$\rho = \text{voids}/\text{cm}^3 \times 10^{-15}$$

$$T = \text{temperature, } ^\circ\text{K}.$$

These equations are based on transmission microscopy results from 15 specimens that were irradiated over a temperature range from 370 to 620 $^\circ\text{C}$ and fluence from 0.4 to $5.9 \times 10^{22} \text{ n}/\text{cm}^2$ ($E > 0.1 \text{ MeV}$).

Because of the limited amount of data presently available and the rather large degree of uncertainty associated with the irradiation conditions, it is difficult to determine which equation provides the better fit with the data. The essential difference between the two equations is that Equation (1) indicates a power relationship between fluence and void concentration with a constant exponent of 0.9, whereas Equation (2) indicates the same relation except for a temperature dependent exponent which varies from about 0.76 at 370 $^\circ\text{C}$ to 1.26 at 620 $^\circ\text{C}$. Both equations appear to predict "high" values at high irradiation temperatures indicating that the form of temperature dependence chosen for the intercept ($\rho A/T-1$) should be changed. Several equations have been developed which provide good fits to the void size data. Two examples are:

$$d = 417 + 0.87T \tag{3}$$

$$d = 4150 \left(\frac{\phi t}{10^{22}} \right)^{0.082} \rho^{-2202/T} \tag{4}$$

While different in form, these equations predict very similar values over the range of conditions for which we have data. Equation (3) actually fits the data over the whole range of conditions slightly better than Equation (4) which was included here to emphasize the insensitivity of void size to neutron dose. The estimated standard deviation on the fluence

exponent in Equation (4) was 0.042, half as large as the exponent itself. This indicates that statistically, the fluence dependency of the void size data is almost insignificant.

This effect is shown in Figures 4.26 and 4.27. Figure 4.26 illustrates the structure observed in Type 304 SS after a dose of 7×10^{21} n/cm² ($E > 0.1$ MeV) at 565 °C, whereas Figure 4.27 illustrates the structure after 3×10^{22} n/cm² ($E > 0.1$ MeV) at 595 °C. Despite the large differences in fluence, approximately the same void size was observed. It is noteworthy, however, that for the low fluence, the voids are predominately located on precipitates. Extraction replicas have identified only the complex carbide $M_{23}C_6$. At present, this phenomenon has been observed only at high irradiation temperatures.

The low correlation between void size and fluence is an important feature which must be accounted for in a theoretical description of void formation and growth. It appears that the voids may be considered to nucleate and grow, in small regions independent of all other regions, to a size range which is limited only by irradiation temperature. The interstitial loops which are also formed during irradiation may play a major role in this limitation. The loops increase both in size and spacing with irradiation temperature as shown in Figures 4.28 and 4.29. Consequently, if one assumes that close proximity of a loop prevents growth of a void, the increased void size with increased irradiation temperature may be explained by the increasing loop spacing.

An important corollary to the above argument is that stainless steels with high stacking fault densities produced by thermomechanical treatment or aging should have improved swelling resistance. Austenitic stainless steels with these structures have been developed for this purpose at BNW. These materials were developed under Task A of the IRDM Program, but irradiation experiments designed to determine their swelling

Neg 8513-A

69,420X

FIGURE 4.26. Absorption Contrast Condition Showing Voids and Rod-Shaped Precipitates in Type 304 SS Irradiated at 1050 °F to 0.7×10^{22} n/cm² (E > 0.1 MeV)

Neg 8516-C

69,420X

FIGURE 4.27. Large Faulted Loops in Same Material as Shown in Figure 4.25, Except Oriented for Diffraction Contrast, $g = [T11], [w11]$ Zone Axis

Neg 8712-B2

78,000X

FIGURE 4.28. Small Irregular Faulted Loops in Type 304 SS
Irradiated at 700 °F to 0.4×10^{22} n/cm²
($E > 0.1$ MeV). The Foil Is Oriented to the
Diffraction Contrast for $g = [002]$, $[110]$
Zone Axis

Neg 8301-B

69,420X

FIGURE 4.29. Voids in Type 304 SS Irradiated at 1100 °F
to 3.0×10^{22} n/cm² (E > 0.1 MeV)

behavior have been initiated in a cooperative effort between the Cladding Evaluation Section and the Metallurgy Section of the Chemistry and Metallurgy Division.

One may use Equations (1) through (4) to calculate the void volume based on transmission electron microscopy. If this is done, and Equation (2) is used for the void concentration, a temperature-dependent fluence exponent for void volume is obtained, suggesting similar behavior should be observed in the bulk density change data. Close examination of the data indicates that such a trend may be present.

Mechanical Testing of Fuel Pin Cladding

R. L. Fish

The purpose of this program is to provide a basis for evaluating the combined effects of fuel cladding interaction, fluence, and operating temperature on the postirradiation burst and stress-rupture properties of fuel pin cladding.

Burst properties of fuel cladding sections from PNL-1 series fuel pin irradiations at 900 °F have been given in previous quarterly reports. The most important finding in these previously reported studies was a very low strength and ductility region above the reactor midplane. The low strength is thought to be due to grain boundary attack during testing of sensitized cladding. The nature of the corrodant has not been identified.

Additional fueled cladding specimens prepared in a dry, inert atmosphere, but stored for about 10 months in hot cell air after puncture of cladding for fission gas analysis, have revealed intergranular cracks during leak testing. These specimens were from the upper fueled regions of PNL 1-16 and 1-18 pins.

The origin of the stresses thought to be necessary to cause this cracking is not known, but the crack orientation suggests the pins were bent during storage. The intergranular failure in PNL 1-16 and 1-18 occurred at ambient temperatures which demonstrates the boundary weakness effect is not confined to elevated temperatures.

The cause of the boundary weakness is very important to fast reactor technology. If the weakness is due to storage environment, then pin handling procedures must be improved in order to preserve data. Alternately, if the effect is inherent in fuel pin performance, then the cladding will not be capable of supporting the loads which appear safe from unfueled irradiation experiments.

In order to clarify the mechanism involved with the boundary weakness, a number of definitive experiments have been conducted and additional experiments are planned.

The electron microprobe has been used to examine grain boundaries in failed areas to determine the extent of average penetration by fission products.

Figure 4.30 illustrates the typical cladding integrity found at the fissures in the fueled specimens from above the reactor midplane and also shows the results of the electron microprobe examination performed on the same specimen from PNL 1-18. The scan across the grain boundary indicated in the micrograph revealed the presence of cesium. There was no fuel associated with this cesium as is the case along the inside diameter of the cladding. Cesium was found in one other grain boundary area in the cladding and was again unassociated with fuel. No definite conclusion can be drawn from these observations, however, because of the possibility of cesium penetration or deposition along the grain boundaries subsequent to specimen failure.

PNL 1-18
MICROPROBE RESULTS

CLADDING MICROSTRUCTURE AT FAILURE

Neg 700160-6

FIGURE 4.30. Electron Microprobe Results from Above Fuel Midplane in PNL 1-18

Initial results were obtained from a ring test, on a specimen cut from a biaxial test specimen which failed by grain boundary fracture prematurely in a biaxial test. In the ring test, a 1/4-in. long section of cladding is pulled in a tensile machine in such a way as to create tangential stress in the cladding wall. Fuel and fission products are removed prior to testing at 900 °F. The metallography on these ring test ruptures is shown in Figure 4.31. The good ductility and strength exhibited by this cladding with the fuel and fission products removed contrasts greatly with the very low ductility ($<1.0\% \Delta D/D$), intergranular failures observed at the same test temperature in fueled cladding from the same region of the fuel pin. This indicates that the premature failures are due to corrosive attack from within the fuel pin and are not a result of external attack or inherent cladding grain boundary weakness. Two burst tests (one fueled and one with fuel removed) are planned to verify the effects observed in these ring tests. Carbon extraction replicas are also being obtained from the ring test samples for further characterization of their microstructure.

Fuel pins PNL 1-9, 1-12, 1-13, and 1-14 were helium leak tested for possible fissures. These pins have not been punctured nor otherwise intentionally compromised. This testing did not reveal any fissures or leaks. The outside surface of the cladding on these same fuel pins was replicated with cellulose acetate plastic. This replication did not reveal any intergranular cracking or other cladding perforations. The only significant characteristic revealed by the replicas was a pronounced delineation of the grain boundaries in the upper fueled region of PNL 1-14 (Figure 4.32). This is believed to be heavy carbide precipitation and is not unexpected in view of the calculated temperatures (~ 900 °F) of this cladding during irradiation.

(a) Irradiation Conditions: $\phi t \approx 3.7 \times 10^{21} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$)
 $T_{\text{ave}} \approx 945 \text{ }^\circ\text{F}$

(b) Irradiation Conditions: $\phi t \approx 2.8 \times 10^{21} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$)
 $T_{\text{ave}} \approx 940 \text{ }^\circ\text{F}$

FIGURE 4.31. Microstructure of Ring Test Samples (Test Temp = 900 °F)

FIGURE 4.32. Precipitate Delineation of Cladding Grain Boundaries in Upper Fueled Region of PNL 1-14

FAST REACTOR SUPPORTING STUDIES

Acceptance Testing and Clad Characterization

M. M. Paxton

The clad characterization program is being conducted to ensure that high quality stainless steel cladding is obtained and used throughout the FFTF fuels development and associated irradiation testing program. Under this program, selected mechanical properties are being determined and analyzed to evaluate fabrication variables and assist in vendor qualification.

The characterization of PNL tube lots E, F, G and H are essentially complete, except for a few long-term, stress-rupture tests. The characterization of PNL lot N-1, Carpenter mill release numbers 83628 and 83629, and PNL lot N-2, Carpenter

mill release number 83626, 83627, and 83631 has been initiated and is progressing satisfactorily. The current status of all PNL lots being characterized is contained in Tables 4.9 through 4.13.

A check analysis has been completed by Lukens Steel Company on PNL lots N-1 and N-2, Tables 4.14 and 4.15. This analysis is in agreement with that supplied by the vendor.

The volume of data being generated on each lot of tubing and its distribution to the individuals that use the data has shown the need for a data handbook for each lot of tubing. Such a handbook containing all available data on a particular lot of tubing is presently being assembled. The first distribution is expected about April 15, 1970.

Tensile Studies of Types 304 and 316 Stainless Steels

A. L. Ward

The objective of this program is to provide mechanical property and metallurgical stability data for candidate LMFBR vessel and core-structural materials in order to define adequate lifetime performance under anticipated service conditions.

The effect of fast reactor irradiation on the ductility of solution-annealed AISI Types 304 and 316 SS has been investigated. Ductility losses caused by low fluence irradiation (less than about 10^{22} n/cm²) have been extensively studied in many metals. Usually the losses are described in terms of two temperature domains divided approximately by one-half the absolute melting temperature ($T_m/2 \approx 560$ °C). At irradiation and test temperatures below $T_m/2$, irradiation-produced point defect clusters increase the flow stress and lead to the early onset of plastic instability. Above $T_m/2$, displacement damage anneals rapidly and does not lead to irradiation hardening. However, at such temperatures, helium generated from various (n,α) reactions readily migrates to grain boundaries. Gas bubbles formed by agglomeration of helium atoms

TABLE 4.9. Mechanical Properties PNL Tube Lot E, Mill-Annealed Type 304 SS,
0.250-in. OD x 0.218-in. ID

Test Temp., °F	Burst Test			Tensile Test				Stress Rupture			
	Press., psi	Max Hoop Stress	% $\Delta D/D$	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Uniform Elong., %	Total Elong., %	Max Hoop Stress	Rupture Time, hr	% $\Delta D/D$	$\dot{\epsilon}$ ($\Delta D/D$), %/hr
R.T.	10,700	78,540	-	42,090	97,940	73.9	86.3				
R.T.	10,700	78,540	-	46,550	93,110	53.6	63.9				
R.T.	10,800	79,270	-	51,300	93,110	40.6	61.7				
800				21,580	74,120	40.5	43.7				
800				23,840	68,360	36.7	42.1				
800				24,180	66,670	36.3	40.7				
1000	7,390	54,240	-	18,950	68,220	40.1	43.9	50,000	38.0	12.6	0.3315
1000								49,000	40.8	10.8	0.2647
1000	7,430	54,540	-	20,900	62,710	34.5	40.0	47,000	74.9	10.1	0.1348
1000	7,410	54,390	-	22,370	62,260	35.2	41.1	46,000	177.3	7.8	0.0439
1000								42,000	409.0	7.2	0.0176
1000								38,200	1,720.1	-	-
1200	6,260	45,950	-	16,780	48,550	26.7	33.9	35,300	9.8	9.3	0.9489
1200	6,230	45,730	-	20,190	54,350	30.1	35.3	29,000	44.5	10.9	0.2449
1200	6,280	46,095	-	21,530	54,750	31.6	36.8	28,050	80.0	-	-
1200								27,150	125.0	5.7	0.0456
1200								24,300	268.2	4.1	0.0152
1200								22,100	821.0	-	-
1200								22,100	853.0	-	-
1200								19,150	4,173.0	7.5	0.0017
1400	5,000	36,700	-	15,710	28,980	15.6	40.1				
1400	5,020	36,850	-	17,740	39,660	26.3	45.9				
1400	5,060	37,140	-	16,950	43,670	28.2	38.9				
1600	3,510	25,760	-	10,310	12,970	7.5	36.6				
1600	3,530	25,910	-	15,820	26,660	19.8	59.0				
1600	3,550	26,060	-	14,750	31,470	22.2	60.7				
1800	2,340	17,175	-								
1800	2,460	18,060	-								
1800	2,600	19,080	-								

**TABLE 4.10. Mechanical Properties PNL Tube Lot F, Mill-Annealed Type 304 SS,
0.250-in. OD x 0.218-in. ID**

Test Temp., °F	Burst Test			Tensile Test				Stress Rupture			
	Press., psi	Max Hoop Stress	$\dot{\epsilon}$ min ⁻¹	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Uniform Elong., %	Total Elong., %	Max Hoop Stress	Rupture Time, hr	$\dot{\epsilon}$ ($\Delta D/D$), %/hr	$\dot{\epsilon}$ ($\Delta D/D$), %/hr
R.T.	9,950	73,030	0.01	46,810	101,440	68.6	83.6				
R.T.	9,950	73,030	1.0	50,510	93,820	50.6	62.4				
R.T.	9,900	72,670	10.0	54,010	94,800	43.5	52.4				
800			0.01	23,480	67,600	33.8	38.0				
800			1.0	30,760	66,070	32.0	36.3				
800			10.0	25,420	64,740	34.2	39.0				
1000											
1000	7,020	51,530	0.01	23,700	64,310	34.3	40.8	49,000	16.8	17.5	1.0416
1000	7,070	51,890	1.0	21,020	57,910	30.0	35.2	48,000	26.4	15.5	0.5871
1000	7,075	51,930	10.0	24,580	58,650	32.3	38.7	47,000	31.0	14.7	0.4741
1000								46,000	50.3	12.8	0.2544
1000								44,000	119.3	10.6	0.0888
1000								42,000	231.3	9.	0.0389
1000								39,000	1,105.1	7.7	0.0069
1000								37,500	1,683.5	-----	-----
1200	5,790	42,500	0.01	20,110	44,210	24.5	47.5	27,800	17.8	11.3	0.6348
1200	5,820	42,720	1.0	19,960	51,190	26.9	31.4	26,900	41.8	-----	-----
1200	5,830	42,790	10.0	20,450	50,396	28.9	34.6	24,300	112.5	-----	-----
1200								21,650	281.9	7.7	0.0273
1200								18,950	1,388.6	6.1	0.0043
1200								16,800	4,729.0	9.2	0.0019
1200								15,250	6,690.7	7.3	0.0010
1400	4,460	32,740	0.01	13,930	22,970	16.5	59.6				
1400	4,450	32,660	1.0	17,290	36,190	23.4	51.2				
1400	4,400	32,300	10.0	19,710	42,200	26.4	39.8				
1600	3,170	23,270	0.01	8,420	10,540	14.8	43.3				
1600	3,200	23,490	1.0	14,690	22,090	18.8	65.8				
1600	3,090	22,680	10.0	14,520	29,040	22.2	66.3				
1800	2,040	14,970									
1800	2,000	14,680									
1800	2,100	15,410									

TABLE 4.11. Mechanical Properties PNL Lot G, Mill-Annealed Type 316 SS,
0.250-in. OD x 0.218-in. ID

Test Temp., °F	Burst Test		Tensile Test				Stress Rupture			
	Press., psi	Max Hoop Stress	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Uniform Elong., %	Total Elong., %	Max Hoop Stress	Rupture Time, hr	% $\Delta D/D$	$\dot{\epsilon}$ ($\Delta D/D$), %/hr
R.T.	10,950	80,370	42,230	92,740	65.7	79.8				
R.T.	11,100	81,470	46,470	93,110	42.4	59.6				
R.T.	10,700	78,540	51,530	96,000	38.6	56.9				
800			27,350	80,280	65.7	79.8				
800			27,460	71,130	33.6	37.1				
1000			27,230	70,740	34.2	37.4				
1000	8,110	59,530	24,040	75,200	34.0	38.3				
1000	8,300	60,920	25,310	66,890	30.6	33.3				
1000	8,150	59,820	24,750	66,670	32.6	36.8				
1200	6,960	51,090	21,100	53,470	19.5	44.4				
1200	7,220	52,990	21,240	59,100	27.6	33.0				
1200	7,210	52,920	22,150	60,680	28.3	32.9				
1200							58,000	217.4	16.0	0.0735
1200							56,000	504.0	11.9	0.0236
1200							55,000	712.5	10.6	0.0148
1200							37,800	8.3	15.7	1.8915
1200							34,700	20.9	16.1	0.7703
1200							32,300	42.4	13.7	0.3231
1200							30,170	84.2	17.7	0.2102
1200							30,310	94.9	14.1	0.1485
1200							28,190	172.1	11.7	0.0679
1200							26,940	327.8	11.7	0.0356
1200							23,860	818.4	9.7	0.0118
1200							21,360	1,926.1	14.0	0.0072
1200							20,000	1,606.9	---	---
1200							19,890	2,913.0	10.3	0.0035
1200							19,000	4,256.6	---	---
1400	5,610	41,180	16,980	28,670	12.0	59.0				
1400	5,620	41,250	20,230	43,110	22.9	48.0				
1400	5,640	41,400	19,660	49,490	26.6	39.3				
1600	4,050	29,730	11,440	14,040	9.2	46.7				
1600	4,160	30,530	15,650	27,520	17.8	65.8				
1600	4,060	29,800	18,250	35,530	19.6	60.7				
1800	2,780	20,405								
1800	2,740	20,110								
1800	2,980	21,870								

**TABLE 4.12. Selected Mechanical Properties PNL Tube Lot H, Mill-Annealed
Type 316 SS, 0.250-in. OD x 0.218-in. ID**

Test Temp., °F	Burst Test			Tensile Test			Stress Rupture			
	Press., psi	Max Hoop Stress	% $\Delta D/D$	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Uniform Elong., %	Total Elong., %	Max Hoop Stress	Rupture Time, hr	$\dot{\epsilon}$ ($\Delta D/D$), %/hr
R.T.	11,000	80,740	-	44,400	94,780	57.2	73.0			
R.T.	10,650	78,170	-	48,360	89,830	45.2	57.2			
R.T.	10,900	80,010	-	51,300	94,010	40.6	55.8			
800				26,130	78,450	38.1	41.4			
800				29,490	72,160	33.1	36.7			
800				27,120	69,036	31.8	39.7			
1000	7,770	57,030	-	23,870	75,370	35.9	39.6	56,000	272.1	11.2
1000	8,260	60,630	-	23,160	66,440	30.0	32.9	57,000	358.8	14.0
1000	8,380	61,510	-	24,740	65,650	32.0	36.5	55,000	444.7	41.9
1200	7,120	52,260	-	21,270	51,550	22.7	39.4	34,720	10.5	10.8
1200	6,940	50,940	-	23,280	59,320	29.3	35.3	31,430	33.5	13.6
1200	7,140	52,410	-	21,020	59,890	24.0	33.4	30,170	60.5	13.7
1200								20,440	222.9	12.8
1200								23,860	665.1	9.3
1200								21,730	1,581.9	13.6
1200								20,410	1,762.0	11.3
1400	5,440	39,930	-	17,630	28,870	12.9	58.2			
1400	5,340	39,200	-	20,110	41,240	21.2	47.0			
1400	5,450	40,000	-	18,870	48,530	26.0	39.1			
1600	3,880	28,480	-	10,310	14,430	7.7	46.5			
1600	3,820	28,040	-	19,600	28,580	15.8	64.2			
1600	3,950	28,990	-	16,380	34,070	19.5	61.1			
1800	2,680	18,940	-							
1800	2,630	19,300	-							
1800	2,640	19,380	-							

TABLE 4.13. Selected Mechanical Properties on PNL Tube Lot N-1, MFG's
Lot 83628 and 83629, 20% CW Type 316 SS, 0.280-in. OD
× 0.200-in. ID

Test Temp., °F	Burst Test			Tensile Test			Stress Rupture				
	Press., psi	Max Hoop Stress	%ΔD/D	ε̇ min ⁻¹	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Uniform Elong., %	Total Elong., %	Max Hoop Stress	Rupture Time, hr	ε̇ (ΔD/D), %/hr
R.T.	18,560	134,560	0.8	0.01	90,990	119,210	14.2	21.1			
R.T.	18,220	132,090	0.7								
R.T.	18,640	135,140	0.9								
800	14,980	108,600	0.8	0.01	75,540	98,850	7.5	9.1			
800	14,460	104,840	0.7								
800	15,280	110,780	1.1								
1000	13,770	99,830	0.6	0.01	70,860	91,810	9.5	11.9	90,600	113.1	0.0132
1000	13,580	98,460	1.0						72,500	544.2	0.0022
1000	13,850	100,410	1.2								
1200	11,400	82,650	1.6	0.01	54,060	64,090	3.2	19.7	61,630	1.2	0.6667
1200	10,920	79,170	1.6						43,500	32.8	0.1737
1200									36,250	193.6	0.0340
1200									30,000	1,042.3	0.0051
1400	8,460	61,340	2.5	0.01	33,170	37,460	1.5	29.0			
1400	8,420	61,050	3.6								
1400	8,260	59,890	2.7								
1600	5,560	40,310	8.1	0.01	16,370	19,440	2.0	39.7			

TABLE 4.14. Chemical Analysis PNL Lot N-1, by Lukens Steel Company

Tube No.	N	Al	C	Mn	P	S	Si	Ni	Cr	Mo	Cb	Ti	Cu	B
28-073	0.006	0.015	0.059	1.48	0.007	0.013	0.48	13.30	16.79	2.52	0.04	0.002	0.07	0.0012
28-087	0.006	0.008	0.059	1.48	0.006	0.011	0.49	13.68	16.84	2.51	0.05	0.002	0.07	0.0017
29-055	0.006	0.007	0.055	1.47	0.008	0.014	0.49	13.30	16.95	2.54	0.04	0.002	0.07	0.0017
29-184	0.006	0.003	0.055	1.42	0.006	0.012	0.49	12.92	16.93	2.41	0.05	0.003	0.07	0.0012
29-180	0.006	0.008	0.055	1.48	0.007	0.012	0.49	13.44	16.60	2.47	0.05	0.002	0.07	0.0019

TABLE 4.15. Chemical Analysis PNL Lot N-2, by Lukens Steel Company

Tube No.	N	Al	C	Mn	P	S	Si	Ni	Cr	Mo	Cb	Ti	Cu	B
26-162	0.005	0.010	0.056	1.47	0.008	0.011	0.49	13.50	16.75	2.45	0.06	0.002	0.07	0.0019
27-113	0.006	0.008	0.054	1.47	0.005	0.011	0.49	13.36	16.79	2.50	0.05	0.002	0.07	0.0025
27-158	0.006	0.005	0.053	1.52	0.007	0.012	0.50	13.44	16.82	2.63	0.05	0.002	0.07 ⁵	0.0032
31-089	0.006	0.005	0.053	1.56	0.007	0.010	0.48	13.44	16.77	2.65	0.05	0.002	0.07 ⁵	0.0040
31-100	0.006	0.009	0.056	1.48	0.005	0.010	0.48	13.36	16.90	2.52	0.04	0.002	0.07	0.0025

result in weakened boundaries and subsequent premature failure. At high fluences (10^{22} n/cm² and above) these concepts must be modified. In this fluence region displacement damage in the form of dislocation loops and voids accumulates to levels which can cause appreciable strength increases at irradiation temperatures as high as $0.7 T_m$. Thus, at high fluences, ductility loss may be controlled by the combined effects of helium embrittlement and displacement damage.

In order to clarify the roles of helium and displacement hardening, test results from irradiated specimens were compared with those from identical specimens annealed for 1 hr at 1100 °C, a temperature sufficient to removal all displacement damage. The very low solubility of helium in metals causes the helium embrittlement process to remain operative after annealing, so that the role helium plays in ductility loss is reflected in the tests conducted after annealing. Once the effects of helium are known, the relative roles of helium and displacement can be deduced.

Type 316 SS specimens were taken from a group irradiated in an experimental subassembly in the Experimental Breeder Reactor-II (EBR-II) to a peak fluence of 2.7×10^{22} n/cm² ($E > 0.1$ MeV) at temperatures ranging from 390 to 620 °C. Type 304 SS specimens were obtained from the EBR-II, 5C3 control rod thimble irradiated to a peak fluence of 3.3×10^{22} n/cm² ($E > 0.1$ MeV) at temperatures between 370 and 470 °C. These specimens contain helium concentrations ranging from 2 to 16 ppm (atomic).

Postirradiation annealing produces nearly complete restoration of preirradiation tensile properties in specimens tested below $T_m/2$ (Table 4.16). In Type 304 SS specimens tested at 370 and 480 °C (0.38 and $0.45 T_m$), the strength and flow properties returned to those typical of the unirradiated material, which indicates that the postirradiation

TABLE 4.16. Effect of Postirradiation Annealing at 1100 °C on the Tensile Properties of AISI Types 304 and 316 SS

Condition	Irrad. Temp., °F	Test Temp., °F	Fluence, $n/cm^2(a)$	304 Stainless Steel			Uniform Elong., %	Total Elong., %	Reduction of Area, %
				Yield Stress, ksi	Ultimate Stress, ksi				
As-Irradiated	400	370	3.3×10^{22}	107.5	108.3		0.7	1.9	41.7
Annealed	400	370	3.3×10^{22}	19.5	67.0		40.9	42.9	73.2
Un-Irradiated	---	370	---	19.2	62.3		35.0	50.0	62.5
As-Irradiated	400	480	3.3×10^{22}	95.8	96.0		0.8	1.7	29.9
Annealed	400	480	3.3×10^{22}	16.8	61.2		42.1	43.4	72.6
Un-Irradiated	---	480	---	18.0	59.0		36.0	41.5	69.0
As-Irradiated	400	600	3.3×10^{22}	56.3	56.3		0.4	0.5	7.9
Annealed	400	600	3.3×10^{22}	16.1	38.6		13.8	15.1	43.7
Un-Irradiated	---	600	---	16.5	46.3		27.5	33.2	46.0
As-Irradiated	400	700	3.3×10^{22}	30.8	30.8		0.2	0.3	9.4
Annealed	400	700	3.3×10^{22}	15.5	22.5		5.8	6.8	19.7
Un-Irradiated	---	700	---	14.5	25.0		14.0	26.0	33.5
As-Irradiated	460	480	0.38×10^{22}	49.2	74.6		22.9	27.1	61.4
Annealed	460	480	---	16.3	61.3		34.4	36.1	53.3
Un-Irradiated	---	480	---	18.0	59.0		36.0	41.5	69.0
As-Irradiated	460	600	0.38×10^{22}	40.0	49.0		8.9	10.4	39.5
Annealed	460	600	0.38×10^{22}	12.7	35.5		18.4	19.3	42.1
Un-Irradiated	---	600	---	16.5	46.3		27.5	33.2	46.0
As-Irradiated	460	700	0.38×10^{22}	26.7	30.1		5.0	7.9	16.4
Annealed	460	700	0.38×10^{22}	12.3	22.4		9.5	11.1	34.1
Un-Irradiated	---	700	---	14.5	25.0		14.0	26.0	33.5
				316 Stainless Steel					
As-Irradiated	580	650	2.7×10^{22}	42.6	46.9		2.0	2.8	11.4
Annealed	580	650	2.7×10^{22}	14.1	33.4		9.6	10.4	19.5
Un-Irradiated	---	650	---	16.8	49.0		28.0	42.0	47.5
As-Irradiated	515	650	1.6×10^{22}	50.6	52.5		1.3	2.7	8.1
Annealed	515	650	1.6×10^{22}	14.4	33.5		9.7	10.6	22.8
As-Irradiated	480	650	1.1×10^{22}	50.2	51.5		0.8	2.5	13.8
Annealed	480	650	1.1×10^{22}	14.1	34.2		10.5	11.4	21.1
As-Irradiated	440	650	0.9×10^{22}	42.4	45.2		2.6	4.4	21.1
Annealed	440	650	0.9×10^{22}	13.9	35.0		11.7	12.6	18.7

a. Neutrons with energies greater than 0.1 MeV.

anneal eliminated all displacement damage. At these temperatures, the helium embrittlement process is not active, and no residual ductility losses are expected once displacement damage is removed. At test temperatures greater than $T_m/2$, the postannealing data presented here show a marked departure from low fluence results. At low fluences, ductility is not significantly improved after annealing because of the insolubility of the helium. In contrast, substantial increases in ductility are observed in the high fluence case (Table 4.16). However, a significant nonrecoverable ductility loss remains. The ductility improvement is attributed to the removal of dislocation loops and voids as a result of the annealing, while the nonrecoverable loss is attributed to the classical helium embrittlement process.

The magnitude of the nonrecoverable ductility loss is consistent with results obtained by Kramer et al.^(33,34) from studies in which helium concentrations from 0.1 to 30 ppm were introduced by cyclotron injection of alpha particles. Indirect evidence previously published by Holmes⁽³⁵⁾ suggested that the helium embrittlement mechanism, acting independently of displacement damage, controls high-fluence, elevated-temperature ductility. The present findings show that in the range from 593 to 704 °C, both processes contribute to the total ductility loss and suggest that helium embrittlement dominates only at sufficiently high temperatures to eliminate the displacement damage.

Uniaxial Creep-Rupture Studies of Types 304 and 316 Stainless Steel

A. J. Lovell

The objectives of this program are to evaluate the effects of fast neutron irradiation and environment on the postirradiation uniaxial creep-rupture properties of candidate vessel and

core structural materials for LMFBR and to establish the usable materials limits to assure their safe and reliable performance.

The effects of fast neutron irradiation to 1.2×10^{22} n/cm² (total) at ~900 °F (~480 °C) on the postirradiation creep-rupture properties of Types 304,⁽¹²⁾ 316,⁽¹²⁾ and 348⁽³⁶⁾ SS have been reported. The comparison of these three sets of results at a test temperature of 1000 °F (538 °C) are shown in Figures 4.33 and 4.34. In the unirradiated condition, the three alloys have substantially different rupture life and minimum creep rates. After irradiation, the rupture life of all alloys is reduced, with the Types 348 and 316 SS materials exhibiting larger reductions than the Type 304 material. Minimum creep rates of the Types 348 and 316 alloys are increased as a result of irradiation. The end result is that these alloys, which were quite different prior to irradiation, now exhibit similar creep-rupture properties after irradiation.

Weldment Studies

A. L. Ward and A. J. Lovell

Postirradiation tensile test results are presented in Tables 4.17 and 4.18 for base metal (AISI Type 304 SS) and all-weld specimens representing the submerged-arc, TIG, MIG, and stick-electrode weld processes. The specimens included in this study were irradiated in Pins No. 35, 36, and 37 of Sub-assembly X067 in Row 4 of the EBR-II. Listed fluences are based on a peak of 8.75×10^{21} n/cm² (total) and a fluence versus axial distance profile established by dosimeter activation measurements in another Row 4 irradiation experiment. The irradiation temperature for specimens represented in Table 4.17 was 800 °F and as indicated for those in Table 4.18. Reported temperatures are based on heat transfer calculations. Figures 4.35 and 4.36 show yield strength and total elongation, respectively, as functions of neutron fluence ($E > 0.1$ MeV) for

FIGURE 4.33. Postirradiation Rupture Properties of Stainless Steels
 Irradiated to 1×10^{22} n/cm² (total) at ~ 900 °F (482 °C),
 Tested at 1000 °F (538 °C)

Neg 701101-1

FIGURE 4.34. Postirradiation Creep Properties of Stainless Steels Irradiated to 1×10^{22} n/cm² (total) at ~ 900 °F (~ 482 °C), Tested at 1000 °F (538 °C)

TABLE 4.17. Tensile Properties of Weldment Materials Irradiated in the EBR-II at 800 °F

Specimen	Type	Test Temp., °F	ϕt ($10^{21} n/cm^2$)	Proportion of Elastic Limit, psi	Yield, psi	Ultimate, psi	Uniform Elong., %	Total Elong., %	Reduction of Area, %
35-T-B22A	Base	600	4.4	36,000	49,400	72,000	26.6	34.2	68.0
35-T-B23A	Base	700	4.4	40,600	51,200	74,300	23.6	30.1	59.4
36-T-B40A	Base	800	4.4	33,800	49,600	69,800	21.8	30.5	57.6
36-T-B43A	Base	900	4.4	38,100	50,000	67,500	25.4	33.2	52.8
35-T-W10	TIG	600	3.8	52,800	69,600	77,900	6.2	14.4	60.8
35-T-W11	TIG	700	3.8	53,500	70,900	78,000	3.8	10.5	56.9
36-T-W48	TIG	800	3.8	48,000	66,900	75,500	4.8	11.0	67.5
36-T-W49	TIG	900	3.8	52,800	67,200	70,800	2.9	9.7	74.0
35-SAI-W10	Sub-Arc	600	3.8	41,200	56,000	73,200	15.1	17.0	43.9
36-SAI-W18	Sub-Arc	700	3.8	39,800	55,000	75,400	14.7	15.5	37.4
35-SAI-W11	Sub-Arc	800	3.8	40,600	56,400	78,300	18.1	19.5	35.8
36-SAI-W19	Sub-Arc	900	3.8	39,800	54,800	66,300	9.6	10.7	39.0
35-SEI-W19	St Elect	600	3.8	39,800	62,600	80,300	15.0	19.0	41.5
35-SEI-W24	St Elect	700	3.8	40,600	59,100	77,600	16.6	21.2	57.7
36-SEI-W39	St Elect	800	3.8	43,800	59,300	77,600	14.5	18.1	46.3
36-SEI-W40	St Elect	900	3.8	38,200	58,200	74,800	14.2	20.4	52.0
35-M1-W16	MIG	600	3.8	49,600	62,600	76,100	11.0	16.6	51.2
35-M1-W17	MIG	700	3.8	52,800	65,000	70,200	4.6	9.1	52.0
36-M1-W24	MIG	800	3.8	43,100	59,900	65,600	9.3	13.6	55.3
36-M1-W25	MIG	900	3.8	44,700	59,100	77,600	7.4	13.6	57.7

TABLE 4.18. Tensile Properties of Weldment Materials Irradiated in the EBR-II

Specimen	Type	Test Temp., °F	ϕt ($10^{21} n/cm^2$) Total	ϕt ($10^{21} n/cm^2$) >0.1	Proportion of Elastic Limit, psi	Yield, psi	Ultimate, psi	Uniform Elong., %	Total Elong., %	Reduction of Area, %
35-T-B21A(a)	Base	600	1.3	0.61	13,800	21,700	65,000	45.0	52.6	84.6
36-T-B37A(b)	Base	700	1.3	0.61	13,700	23,700	64,500	45.2	52.7	69.1
35-T-B18A(b)	Base	800	1.3	0.61	10,000	18,500	64,600	44.6	51.9	63.4
36-T-B35A(b)	Base	900	1.3	0.61	12,000	20,100	61,000	37.0	44.0	65.6
35-T-W9(a)	TIG	600	1.1	0.54	45,500	58,300	71,800	9.6	16.5	64.8
36-T-W47(a)	TIG	700	1.1	0.54	35,800	56,800	71,300	10.7	16.5	75.6
35-T-W8(b)	TIG	800	1.1	0.54	36,600	51,300	67,800	12.8	19.2	74.8
36-T-W46(b)	TIG	900	1.1	0.54	39,000	53,900	68,500	11.8	17.3	64.2
35-SA1-W9(a)	Sub-Arc	600	1.5	0.75	25,600	43,100	73,000	19.9	23.0	40.5
36-SA1-W17(a)	Sub-Arc	700	1.1	0.54	28,500	41,500	70,500	18.7	19.1	35.0
35-SA1-W8(b)	Sub-Arc	800	1.1	0.54	26,800	38,100	66,100	24.2	25.2	39.0
36-SA1-W15(b)	Sub-Arc	900	1.1	0.54	28,100	39,100	67,600	22.1	23.8	35.0
35-SE1-W17(a)	St Elect	600	1.1	0.54	43,000	49,800	71,800	18.5	23.7	61.8
35-M1-W15(a)	MIG	600	1.1	0.54	27,600	42,600	66,700	21.6	27.0	60.2

a. Irradiation Temperature 700 °F

b. Irradiation Temperature 800 °F

Neg 700190-1

FIGURE 4.35. Yield Strength of Fast Reactor Irradiated AISI Type 304 SS as a Function of Neutron Fluence ($E > 0.1$ MeV)

Neg 700190-2

FIGURE 4.36. Tensile Ductility of Fast Reactor Irradiated AISI Type 304 SS as a Function of Neutron Fluence (E > 0.1 MeV)

Type 304 SS specimens from several EBR-II irradiation experiments (open symbols). These plots represent data selected on the basis of:

- Test temperatures in the range from 700 to 1000 °F
- Fluences from 6×10^{20} to 3.7×10^{22} n/cm² ($E > 0.1$ MeV)
- Irradiation temperatures from 700 to 900 °F.

Results from the weldment study (closed symbols) represent test temperatures of 600, 700, 800, and 900 °F.

The weld-deposited materials show smaller irradiation-induced yield strength increases and smaller ductility losses than the base metal. Note, however, that the preirradiation yield values were higher and the ductilities lower in the weld material. At fluences of 5.4 and 6.1×10^{20} n/cm² ($E > 0.1$ MeV), properties are essentially unaffected by irradiation, while at 2.6×10^{21} n/cm² yield stress increments of about 20,000 psi and significant reductions in total elongation are observed for all welds except the stick-electrode. Corresponding base metal specimens exhibited ~30,000 psi yield stress increases and 2 to 15% reductions in total elongations (Figures 4.35 and 4.36).

Tensile test results are reported from a study to determine whether a larger diameter specimen would improve the reproducibility of test results and also be more representative of the bulk weld material being studied. Table 4.19 lists the results from the larger specimens (0.250-in. gage diameter, 1.125-in. gage length) tested in the range from 600 to 900 °F. The data is in general agreement with previously reported results for the miniature (0.125-in. gage diameter, 1.25-in. gage length) specimen used in the weldment irradiation experiment. However, no consistent improvement in the magnitude of data scatter is observed upon comparison of the two sets of results. Therefore,

TABLE 4.19. Tensile Properties of 0.250 in. Gage-Diameter Weldment Specimens

Specimen	Type	Test Temp., °F	Proportion of Elastic Limit, psi	Yield, psi	Ultimate, psi	Uniform Elong., %	Total Elong., %	Reduction of Area, %
TX-1	TIG	R.T.	59,100	66,700	83,800	23.5	35.3	77.4
TX-2	TIG	600	36,700	57,000	67,600	11.4	21.2	70.5
TX-3	TIG	700	26,500	38,700	70,800	21.4	28.1	52.8
TX-4	TIG	800	30,600	52,600	63,500	11.0	19.5	61.1
TX-5	TIG	900	29,500	48,400	61,200	11.2	17.8	63.5
APX-1	Sub-Arc	R.T.	30,600	55,500	90,200	37.5	41.9	45.8
APX-2	Sub-Arc	600	31,700	40,400	63,500	21.4	23.3	64.2
APX-3	Sub-Arc	700	20,800	35,800	64,400	25.9	27.2	38.2
APX-4	Sub-Arc	800	26,200	42,300	67,900	19.9	22.4	37.8
APX-5	Sub-Arc	900	25,200	32,900	68,400	20.7	23.9	32.8
D2X-1	St Elect	R.T.	38,900	61,600	93,300	40.7	42.9	38.1
D2X-2	St Elect	600	29,900	45,200	66,700	18.6	22.8	51.7
D2X-3	St Elect	700	31,400	43,700	62,400	16.6	23.3	54.8
D2X-4	St Elect	800	27,600	42,200	65,400	18.9	24.4	46.8
D2X-5	St Elect	900	30,700	42,900	64,100	19.1	22.7	48.5
F2X-1	MIG	R.T.	27,300	48,400	76,100	32.8	54.0	68.3
F2X-2	MIG	600	21,100	36,900	60,500	22.2	28.9	59.9
F2X-3	MIG	700	26,600	53,300	67,000	13.6	19.9	57.8
F2X-4	MIG	800	27,700	41,600	59,400	20.5	26.2	52.5
F2X-5	MIG	900	28,500	41,800	65,600	17.1	23.9	60.1
F3X-1	MIG	R.T.	37,800	55,200	84,900	37.4	43.9	58.3
F3X-2	MIG	600	34,900	48,800	67,400	15.2	19.9	46.6
F3X-3	MIG	700	26,800	41,300	62,200	16.0	22.0	47.6
F3X-4	MIG	800	26,100	38,600	57,500	15.2	20.6	53.6
F3X-5	MIG	900	25,800	39,900	55,900	13.5	19.5	50.7

present plans for future irradiation experiments involving weldment materials call for continued use of the miniature specimen.

Results from creep-rupture tests on weldment specimens irradiated to $\sim 8 \times 10^{21}$ n/cm² (total) at ~ 1100 °F are presented in Table 4.20. This irradiation had little effect on the rupture life of the submerged arc weldment. The TIG weldment material, on the other hand, showed substantial losses in rupture life after irradiation. However, irradiation specimens from the TIG weldment failed at the extreme end of the gage length. Since this failure may actually occur at the weld/base metal interface, these results should be considered preliminary until the fracture is better defined.

TABLE 4.20. Uniaxial Creep-Rupture Properties of Irradiated Stainless Steel Welds

Specimen No.	Fluence, n/cm ² (total)	Irrad. Temp., °F	Test Temp., °F	Stress, ksi	Minimum Creep Rate in./in./hr × 10 ⁵		Rupture Time, hr		Total Elongation, %	
					Irrad.	Control	Irrad.	Control	Irrad.	Control
Sub-Arc Weld, 308 SS Filler										
		1100	1100	45		2360				21.7
		1100	1100	40		380				28.4
35A-SA1-W13	8.5 × 10 ²¹	1100	1100	30	45	210	82.5	48.8		15.2
35A-SA1-W12	7.9 × 10 ²¹	1100	1100	25	5.95	21	455.7	285.0		10.6
35A-SA1-W14	8.8 × 10 ²¹	1100	1100	23	3.69	5.1	599.7	589.0		4.8
TIG Weld, 308 SS Filler										
		1100	1100	45		BOL (a)		BOL		11.5
		1100	1100	40		1220		4.4		14.8
		1100	1100	35		57		73.3		12.2
35-T-W13	7.9 × 10 ²¹	1100	1100	30	24.9	16.2	33.4	205.0		10.0
35-T-W44	7.9 × 10 ²¹	1100	1100	25	6.1	0.23	79.3	1525.8		1.8
MIG Weld, 308L SS Filler										
		1100	1100	45		BOL		BOL		15.6
		1100	1100	40		3820		3.2		25.0
		1100	1100	30		178		38.1		10.3
		1100	1100	25		15.8		270.0		10.7
		1100	1100	18		2.62		1099.0		4.8
Stick Electrode Weld, 16-8-2 Filler										
		1100	1100	45		1270		7.2		18.7
		1100	1100	40		97.8		29.9-45.3		6.2
		1100	1100	35		30.2		103.4		11.4
		1100	1100	30		0.74		728.0		2.3
		1100	1100	25		0.22		OnTest		OnTest

a. Broken on Loading.

REFERENCES

1. G. W. Lewthwaite, D. Mosedale and I. R. Ward. "Irradiation Creep in Several Metals and Alloys at 100 °C," Nature, vol. 216, p. 472. 1967.
2. P. G. McVetty. "Factors Affecting the Choice of Working Stresses for High Temperature Service," Trans. ASME, vol. 55, p. 99. 1933.
3. R. V. Hesketh. "Collapse of Vacancy Cascades to Dislocation Loops," Proc. Intern. Conf. Solid State Physics Research with Accelerators, BNL-50083, p. 389. Brookhaven National Laboratory, Upton, New York, 1967.
4. J. W. Joseph. Stress Relaxation of Torsional Stresses in Stainless Steel During Irradiation, DP-369. E.I. du Pont de Nemours and Co., Wilmington, Delaware, June 1959.
5. R. E. Schreiber. Relaxation of Torsional Stresses in Stainless Steel During Irradiation, DP-669. E.I. du Pont de Nemours and Co., Wilmington, Delaware, April 1962.
6. D. Mosedale, G. W. Lewthwaite, G. O. Leet and W. Sloss. "Irradiation Creep in the Dounreay Fast Reactor," Nature, vol. 224, p. 1301. 1969.
7. G. E. Dieter. Mechanical Metallurgy. McGraw-Hill Book Company, Inc., New York, 1961. p. 66.
8. E. R. Gilbert and J. L. Straalsund. "A Relationship for Non-Conservative-Volume Creep Under Different States of Stress," Nucl. Eng. and Design, vol. 9. 1970.
9. R. V. Hesketh. "Irradiation Creep in Non-Fissile Metals," Brittleness and Irradiation Effects, 10th Symposium on Special Metallurgy, Saclay, France. 1967. p. 185.
10. W. N. McElroy, R. E. Dahl, Jr. and E. R. Gilbert. "Damage Functions and Data Correlation," Quarterly Progress Report, October, November, December 1970, Reactor Fuels and Materials Development Programs for the Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1279, p. 5.18. Battelle-Northwest, Richland, Washington, February 1970.

11. A. J. Lovell and J. F. Bates. "Uniaxial Creep-Rupture Studies," Quarterly Progress Report, July, August, September, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1223, p. 4.98. Battelle-Northwest, Richland, Washington, November 1969.
12. A. J. Lovell. "Uniaxial Creep-Rupture Studies," Quarterly Progress Report, April, May, June, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1144, p. 12.48, Battelle-Northwest, Richland, Washington, August 1969.
13. A. L. Ward. "Tensile Properties of Fast-Reactor-Irradiated AISI Type 316 Stainless Steel," Trans. Am. Nucl. Soc., vol. 12, no. 2, p. 584. 1969.
14. G. L. Kulcinski and B. Mastel. "Annealing of Voids in Neutron Irradiated Nickel," Quarterly Progress Report, October, November, December, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1279, p. 4.7. Battelle-Northwest, Richland, Washington, February 1970.
15. H. R. Brager. Unpublished Data. Battelle-Northwest, Richland, Washington. (Personal Communication)
16. L. C. Roess and C. G. Shull. J. Appl. Phys., vol. 18, pp. 3-8. 1947.
17. A. Guinier and G. Fournet. Small Angle Scattering of X-Rays. J. Wiley & Sons, Inc., New York, 1955. pp. 78-80.
18. S. D. Harkness, R. W. Gould and J. J. Wren. Phil. Mag., vol. 19, p. 115. 1969.
19. J. J. Holmes. Trans. Am. Nucl. Soc., vol. 12, p. 117. 1969.
20. J. L. Brimhall and B. Mastel. Unpublished Data. Battelle-Northwest, Richland, Washington. (To be published)
21. T. E. Volin and R. W. Balluffi. Phys. Stat. Sol., vol. 25, p. 163. 1968.
22. K. H. Westmacott, R. E. Smallman and P. S. Dodson. Met. Sci. J., vol. 2, p. 177. 1968.

23. H. G. Bowden and R. W. Balluffi. Phil. Mag., vol. 19, p. 1001. 1969.
24. J. L. Straalsund. J. of Metals, p. 44A. March 1969.
25. R. Bullough and R. C. Perrin. Radiation Damage in Reactor Materials, Vol. II. p. 233. IAEA, Vienna, 1969.
26. C. W. Hunter and J. A. Williams. "Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels," Quarterly Progress Report, July, August, September, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1223. p. 4.14. Battelle-Northwest, Richland, Washington, November 1969.
27. L. E. Steele, C. Z. Serpan, Jr., J. R. Hawthorne, Uldis Potapovs and R. A. Gray, Jr. Irradiation Effects on Reactor Structural Materials, Quarterly Progress Report, August 1-October 31, 1968, NRL-Memo-1937. Naval Research Laboratory, Washington, D.C., November 15, 1968.
28. L. E. Steele, J. R. Hawthorne, H. E. Watson, C. Z. Serpan, Jr. and R. A. Gray, Jr. Irradiation Effects on Reactor Structural Materials, Quarterly Progress Report, August 1-October 31, 1969, NRL-Memo-2058. Naval Research Laboratory, Washington, D. C., November 15, 1968.
29. L. E. Steele, J. R. Hawthorne, C. Z. Serpan, Jr., Uldis Potapovs, H. E. Watson and R. A. Gray, Jr. Irradiation Effects on Reactor Structural Materials, Quarterly Progress Report, November 1, 1968-January 31, 1969, NRL-Memo-1962. Naval Research Laboratory, Washington, D.C., February 15, 1969.
30. H. H. Johnson and P. C. Paris. "Subcritical Flaw Growth," Engineering Fracture Mechanics, vol. 1, no. 1, pp. 3-45. 1968.
31. C. Cawthorne and E. J. Fulton. "Voids in Irradiated Stainless Steel," Nature, vol. 216, p. 575. 1967.
32. J. L. Straalsund, H. R. Brager, R. L. Fish and J. F. Bates. "Damage Analysis," Quarterly Progress Report, October, November, December, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1279, p. 4.40. Battelle-Northwest, Richland, Washington, February 1970.

33. D. Kramer, H. K. Brager, C. G. Rhodes and A. G. Pard. "Helium Embrittlement in Type 304 Stainless Steel," J. Nucl. Mater., vol. 25, pp. 121-131. 1968.
34. D. Kramer, K. R. Garr, C. G. Rhodes and A. G. Pard. "Helium Embrittlement of Type 316 Stainless Steel," J. Iron and Steel Inst., pp. 1141-1146. 1969.
35. J. J. Holmes, R. E. Robbins and J. L. Brimhall. "Effect of Fast Reactor Irradiation on the Tensile Properties of 304 Stainless Steel," J. Nucl. Mater., vol. 32, pp. 330-339. 1969.
36. A. J. Lovell. "Uniaxial Creep Rupture Studies of Type 348 Stainless Steel," Quarterly Progress Report, October, November, December, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1279. p. 4.4. Battelle-Northwest, Richland, Washington, February 1970.

5.0 FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS

DAMAGE FUNCTIONS AND DATA CORRELATION

W. N. McElroy, R. L. Simons, and L. D. Blackburn

Previous reports⁽¹⁻⁵⁾ discussed the development and study of a method for generating energy-dependent damage functions for the correlation and prediction of neutron-induced property changes.

A preliminary form for a damage function, $G(E)$, is given in Figure 5.1 for a change in yield strength (YS) to 60 ksi in 304 SS irradiated and tested at ~ 450 °C. The solid curve represents the most reasonable of a number of possible solutions for the semi-empirically derived $G(E)$.

The dashed curve represents a vacancy cluster (greater than 7) production rate model developed by Russcher^(6,7) for austenitic steel. Based on the solid curve in Figure 5.1, damage from low-energy neutrons appears to be more prevalent than indicated by Russcher's input model.

Russcher's curve, the multiple foil-derived spectra for approximately 20 measured data points,* and the corresponding fluence estimates for a change to a 60 ksi yield strength were read into the SAND-II code to derive the solid curve solution.⁽⁸⁾ Preliminary results of a solution uniqueness study suggest reasonably good definition for neutron energies above $\sim 10^{-3}$ MeV and poorer definition below this point. This was expected since there is only a single thermal reactor data point.

* Use of multiple foil-derived values rather than calculated values of spectra and flux levels for the PNL and ANL data made some significant differences (up to a factor of ~ 2) in total fluence estimates, particularly for out-of-core data points.

NEG 700671-1

FIGURE 5.1. Absolute Damage Function for a Change in Yield Strength to 60 ksi in 304 Stainless Steel Irradiated and Tested at ~450 °C.

The experimental data used to derive the damage function, Figure 5.1, are presented in Figure 5.2.* Except for the single ETR thermal reactor, data point 42, all data points are from irradiations in EBR-II.

To translate the experimental data into a meaningful form,** it was necessary to establish for each data point (each spectrum) a value of fluence to produce a change in yield strength to 60 ksi. Grouping data points into three spectral groups (see Figure 5.2) made it possible to establish a shape for a single curve that would approximate the property change versus total fluence for each data point (each spectrum); that is, a single shaped curve (solid lines in Figure 5.2) whose lateral displacement at the 60 ksi level represents the energy dependence of damage for these data points.

Analytically, this curve can be represented by the equation

$$YS(\text{ksi}) = 1856 - 89.49 \ln \beta \Phi t + 1.062 (\ln \beta \Phi t)^2, \quad (1)$$

where Φt is the total fluence ($E > 10^{-10}$ MeV in units of n/cm^2) and β is a coefficient (representing the neutron energy dependence of damage), which determines the intersection of the curve with the dashed horizontal line at the 60 ksi level. The use of Equation (1) is limited to yield strengths in the range from ~20 to 100 ksi.

* For these PNL and ANL data, the initial undocumented compilation (with references) provided by the FFTF project (dated December 16, 1968 by J. W. Helm and R. Moen of PNL) has been modified and expanded by L. D. Blackburn. New and/or modified references will be provided for all of these data in future publications.

** The data used in this study were selected after a careful review to eliminate, as far as possible, all variables except spectra as a source of the difference in property change versus fluence.

Neg 701383-9

FIGURE 5.2. Yield Strength (YS) Versus Total Fluence for Different Reactor Spectra

The derived $G(E)$, Figure 5.1, is used directly (or in a group-averaged form) with any neutron spectrum, $\phi(E)$, of interest to determine the fluence required to cause a yield strength change to 60 ksi.⁽⁴⁾ This fluence, Φt , is then used to define the coefficient β for the spectrum, $\phi(E)$, using the equation

$$\beta = 1.90 \times 10^{22} / \Phi t. \quad (2)$$

Substitution of the value of β into Equation (1) yields an equation which can be used to estimate the yield strength (within the range of 20 to 100 ksi) resulting from an exposure Φt in the specified spectrum $\phi(E)$. Such estimates are useful in reactor design, damage analysis studies, and irradiation program planning.

SAND-II SOLUTION ACCURACY STUDY FOR EBR-II SPECTRA

R. L. Simons and W. N. McElroy

The accuracy of the integral flux greater than a specified energy $\Phi(>E)$, determined by using the SAND-II code, depends on uncertainties in cross sections, measured reaction rates, and uniqueness of solution. A study of the effect of these uncertainties on the accuracy of derived energy-dependent integral fluxes for the EBR-II run 31F dosimetry test has been initiated. For a specified integral flux solution $\Phi(>E)$, propagation of errors using a computerized technique will be applied to determine the error due solely to uncertainties in cross sections and reaction rates.

This report considers the contribution of lack of solution uniqueness to the overall inaccuracy of energy-dependent integral fluxes, $\Phi(>E)$. Previous error-estimate studies suggest that the combined errors from all sources should introduce no more than about a 10 to 30% absolute uncertainty in integral flux values.^(9,10)

The SAND-II code calculates

$$\Phi^K(>E) = \int_E^{18 \text{ MeV}} \phi^K(E) dE, \quad (1)$$

where $\phi^K(E)$ is the energy dependent differential flux calculated from a set of N reaction rate integral equations

$$A_i = \int_{10^{-10} \text{ MeV}}^{18 \text{ MeV}} \phi^K(E) \sigma_i(E) dE, \quad i = 1, 2, 3, \dots, N. \quad (2)$$

A_i is the measured reaction rate of the i^{th} reaction, $\sigma_i(E)$ is its corresponding energy-dependent cross section, and K refers to the number of iterations to obtain the $\phi^K(E)$ solution.

This study is intended to show the uncertainty of SAND-II $\Phi(>E)$ solutions for EBR-II run 31F dosimetry data using the available set of 14 reactions, Table 5.1. Data were analyzed from four bounding locations, Rows 2 and 7 at midplane and 59.2 cm above midplane, of the dosimetry test to establish the limit of uniqueness uncertainty at these positions and intermediate locations. The uniqueness limits were determined by obtaining solutions with an extreme input and comparing the results with either a known or a best solution.

TABLE 5.1. Foil Reactions Used for Determining Integral Fluxes

$^{235}\text{U}(n, f)$	$^{24}\text{Na}(n, \gamma)$
$^{237}\text{Np}(n, f)$	$^{54}\text{Fe}(n, p)$
$^{238}\text{U}(n, f)$	$^{58}\text{Ni}(n, p)$
$^{58}\text{Fe}(n, \gamma)$	$^{46}\text{Ti}(n, p)$
$^{59}\text{Co}(n, \gamma)$	$^{47}\text{Ti}(n, p)$
$^{197}\text{Au}(n, \gamma)$	$^{48}\text{Ti}(n, p)$
$^{63}\text{Cu}(n, \gamma)$	$^{27}\text{Al}(n, \alpha)$

The extreme input was required to be a physically reasonable approximation to the correct spectral shape. This approximation for each position was determined by inputting to the SAND-II code an initial fission spectrum form above 3 MeV, a constant form from 3 MeV down to 10^{-4} MeV, and a form proportional to energy below 10^{-4} MeV and the calculated or measured reaction rates for each location. The iterative solution to this initial input was expected to be approximately correct in magnitude at the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ and $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ major resonances, and for threshold monitors above ~ 1 MeV. Thus, an extreme (but physically reasonable) input could be defined from the initial solution by extrapolating an E form below the gold resonance energy, using straight line log-log interpolations between the gold and cobalt resonance energies and between the cobalt resonance energy and 1 MeV, and the initial solution shape above 1 MeV. This spectrum should be a reasonable approximation of the real spectrum in each case but still could be considered as an extreme input form because it is defined entirely by the experimental data; that is, it has no features derived from reactor physics calculations.

Two approaches were taken in determining the uniqueness bounds. First, a completely analytical case was studied in which there were no errors in reaction rates or cross sections and the true form of the spectrum was known. This was done by calculating reaction rates for four known 2DB-calculated⁽¹¹⁾ spectra (for the bounding locations) using the current SAND-II cross sections. These calculated reaction rates and the SAND-II cross sections were then used with the extreme input for each of the four locations to obtain integral flux solutions. The integral flux solutions were compared with the 2DB-calculated integral fluxes by plotting the ratio of solution $\Phi(>E)/2\text{DB calculated } \Phi(>E)$ versus energy. A ratio of one indicates that the solution $\Phi(>E)$ and 2DB-calculated

$\Phi(>E)$ are in agreement, Figure 5.3. The second approach used measured reaction rates from the ^{31}F dosimetry test with their associated experimental errors (estimated to be $\sim 7\%$ of one standard deviation for each of the 14 reactions) and the extreme input to obtain solutions at the four bounding locations. These results are compared to Figure 5.4 with the best solutions by plotting the ratio of the extreme input solution $\Phi(>E)/\text{best input solution } \Phi(>E)$ versus energy. The best solutions were obtained using the same four sets of measured reaction rates, but with the best available input spectral forms (assumed to be the four known calculated 2DB spectra).

Figure 5.3 shows that the maximum differences in $\Phi(>E)$ along the reactor midplane ($z = 0$) and the top bounding position ($z = 59.2$ cm) are $\sim 5\%$ and $\sim 25\%$, respectively. These absolute differences are converted to statistical uncertainties for subsequent error propagation with reaction rate and cross section errors under the conservative assumption that the differences are limits of error at the 2σ (95% confidence) level. Consequently, at a 1σ (67% confidence) level, the estimated solution uniqueness uncertainties are 2.5% and 12.5% at midplane and at the top of the rows, respectively. Since there were no errors in the reaction rates or cross sections used in this analytical case, these uncertainties result from the lack of solution uniqueness associated with the use of 14 foil reactions rather than an infinite set.

For the second case studied, Figure 5.4 shows that the maximum differences in $\Phi(>E)$ along midplane and at the top locations are $\sim 8\%$ and $\sim 50\%$, respectively. At the 1σ confidence level, these uncertainties are $\sim 4\%$ at midplane and $\sim 25\%$ at the top locations.* The results in Figure 5.4 show the effect of uniqueness uncertainty due to errors in measured activities and

* In Figure 5.4, the results for the $r = 5.2$ cm, $z = 59.2$ cm position seem to be excessive relative to the $r = 33.6$, $z = 59.2$ cm results. These results are being rechecked.

FIGURE 5.3. Integral Flux (>E) for Extreme Input/True Integral Flux

FIGURE 5.4. SAND-II Integral Flux (>E) for Extreme Input/SAND-II
Integral Flux (>E) for 2DB Integral Flux Input

cross sections as well as the finite size of the foil set used. These results show that the uncertainties due to lack of solution uniqueness can be increased by a factor of ~ 2 (in the cases studied) by errors in reaction rates and cross sections if extreme, but still physically reasonable, input forms are used.

For both cases, the ratios in Figures 5.3 and 5.4 indicate that the greatest uniqueness uncertainty exists in the energy range from ~ 0.05 to 1 MeV. This is a result of the lack of proper sensitivity of any of the present monitors to neutrons in this region of the spectrum.*

In conclusion, the contribution to the limit of inaccuracy of $\Phi(>E)$ from lack of solution uniqueness for EBR-II run 31F using 14 activation monitors is estimated to be no more than 4 to 25%, 1σ , in the absence of the use of any knowledge about the real form of the spectrum. If available reactor physics results were used, however, the errors should be much less. The lower number would be typical for core and bordering regions. In the blanket, the uncertainty would progressively increase to about 25% at a distance of ~ 59 cm from the core.

FAST REACTOR MATERIALS DOSIMETRY CENTER

J. A. Ulseth and J. L. Jackson

The data file of the Fast Reactor Materials Dosimetry Center has been enlarged and new data handling and reporting methods have been incorporated into the center.

Since last reported,⁽¹²⁾ the data file has been enlarged by an inclusion of unfolded neutron spectra for 13 axial locations (0 to 76 cm above midplane) for each of 6 radial positions (0 to 33 cm) from core center out into the inner

* Recent studies suggest that the addition of the $^{238}\text{Pu}(n,f)$ reaction will improve foil coverage in the 0.1 to 1 MeV range.

blanket region -- bringing the total of unfolded spectra to over 100. These spectra were unfolded (using the SAND-II code)⁽¹³⁾ from 14 measured monitor reaction rates from the run 31F dosimetry experiment.⁽¹⁴⁾ The 14 reaction rates at each reactor position are in the form of a 10×18 matrix.⁽¹⁵⁾

The spectral data are stored in the form of printed output that includes absolute differential and integral flux for 620 energy points ranging from 10^{-10} to 18 MeV, spectral-averaged cross sections for 35 different monitor reactions, and the energy ranges over which the 35 monitors respond in that particular spectrum. The differential flux is punched on IBM cards and is plotted for 620 energy groups.

As requests to the dosimetry center for flux and/or fluence analyses for EBR-II irradiations increase, the data handling and analysis of data become more complicated and time consuming. To facilitate usage, automated procedures have been written for rapid retrieval of information from the expanding data file, and axial and radial flux and fluence distributions for the EBR-II core and blanket can now be rapidly obtained from it. For example, if the distribution of flux is desired for a particular radius, the integral flux above or below any specified energy (up to 20 different energies) are printed in a table suitable for publication. Any or all of the columns in the table can be automatically plotted to give an axial distribution of the integral flux above or below the selected specified energy. Similarly, fluence can be estimated if the megawatt seconds of irradiation are input to the program, and radial flux or fluence distribution in the form of tables or plots can be provided. Two examples of the type of output that can now be obtained from the data file are shown in Figures 5.5 and 5.6. Figure 5.5 is a typical table showing axial distribution of integral flux above various lower energy limits. Figure 5.6

EBR-II RUN 31F - RADIUS = 5.2CM			INTEGRAL FLUX ABOVE SPECIFIED ENERGIES					
CM FROM REACTOR MIDPLANE	AVERAGE ENERGY OF SPECTRUM (MEV)	TOTAL FLUX (N/CM**2-SEC.)	NORMALIZED INTEGRAL FLUX ABOVE (>) OR BELOW (<) SPECIFIED ENERGIES (ENERGIES IN MEV)					
			(NORM. FACTOR)	>1.0-02	>1.0-01	>4.0-01	>1.0+00	>3.0+00
.0	.8174	2.060+15	9.979-01	8.791-01	5.287-01	2.515+01	4.770-02	5.184+03
5.0	.8095	1.992+15	9.977-01	8.757-01	5.232-01	2.477+01	4.738-02	5.114+03
12.3	.7510	1.714+15	9.952-01	8.388-01	4.786-01	2.260+01	4.428-02	4.100+03
15.2	.6930	1.550+15	9.892-01	8.155-01	4.661-01	1.885+01	4.012-02	3.463+03
18.3	.6224	1.336+15	9.775-01	7.825-01	4.268-01	1.733+01	3.076-02	2.735+03
21.4	.5524	1.196+15	9.655-01	7.437-01	3.836-01	1.496+01	2.453-02	2.130+03
25.0	.4717	1.066+15	9.508-01	6.990-01	3.306-01	1.186+01	1.835-02	1.564+03
31.0	.3888	8.130+14	9.144-01	6.246-01	2.835-01	8.917-02	1.235-02	1.028+03
39.4	.2908	5.953+14	8.707-01	5.420-01	2.095-01	5.595-02	6.615-03	5.531-04
50.0	.2143	3.883+14	8.166-01	4.529-01	1.486-01	3.370-02	3.178-03	2.814-04
59.2	.1619	2.671+14	7.767-01	3.785-01	1.008-01	2.123-02	1.550-03	1.571-04
69.2	.1232	1.788+14	7.432-01	3.151-01	6.540-02	1.315-02	7.172-04	7.700-05
76.0	.1081	1.302+14	7.350-01	2.936-01	5.055-02	9.075+03	4.479-04	4.789-05

Neg 701383-7

FIGURE 5.5. Example of Tabular Computer Data Output

EBR II RUN 31F - RADIUS = 5.2CM
 INTEGRAL FLUX ABOVE SPECIFIED ENERGIES

Neg 701383-5

FIGURE 5.6. Example of Plotted Computer Data Output

is a plot of the flux greater than specified energies as a function of axial distance above midplane.

Any or all of the 14 saturated reaction rates can be plotted (as a function of r or z) from the 31F dosimetry test reaction rate matrix. Thus with this aid, an experimenter can determine activity levels for monitors to be included in future EBR-II irradiations. Flux levels at any core or blanket position can also be estimated.

The 31F dosimetry test was conducted during the last quarter of 1968. The general configuration of the core and the inner blanket has not changed to this date, therefore the 31F saturated reaction rates and flux levels continue to be valid.

COMPUTER SIMULATION OF RADIATION DAMAGE

D. G. Doran

The computer simulation of displacement cascade overlap in α -iron has continued. The purpose is to determine how the presence of defect clusters in a lattice influences the defect production in displacement cascades that are subsequently produced.

The results of the simulation of the production and short-term annealing of nine 20 keV cascades were reported previously;⁽¹⁶⁾ these runs will be called Pass 1. The procedure adopted here was to select the final defect configuration from the high temperature annealing simulation of one of these spikes (No. 2051) and modify it for use as the initial damage state. The modification consisted of removing the mobile defects and rearranging the clusters into more stable configurations. Specifically, 13 single vacancies (designated V_1) were removed and the V_5 and V_{21} clusters were transformed into compact polyhedra; 17 single and 4 di-interstitials were removed, and the two I_3 , the I_4 ,

and the I_5 clusters were transformed into compact platelets in {110} planes. Note that the initial numbers of vacancies and interstitials are not equal.

This cluster configuration was then used as the initial damage state in the CASCADE-CLUSTER code for the simulation of nine new displacement spikes (Pass 2). The take-off points and initial directions of the primary knock-on atoms (PKA) into a perfect crystal for Pass 2 were the same as for the previously reported Pass 1.

The resulting cascades were treated with a simulated short-term anneal with the HAP-4 (Hybrid Anneal Program) code. This code, written for a BNW hybrid computer, includes all the operations of the ANNEAL code used previously, plus a MODE 4 in which simultaneous interstitial and vacancy migration are permitted.* Each spike was run in MODE 4 until interactions were infrequent, generally 5000 interstitial time steps. The spike was then run in MODE 3 in which only vacancy migration is permitted for 1000 vacancy time steps.**

The CASCADE-CLUSTER runs (the 0 °K configurations) were only weakly affected by the lack of crystal perfection. For 6 of the 9 spikes, the collision histories of the primary atom were identical in Passes 1 and 2, and the collision histories of higher order knock-ons were altered only slightly. The average number of defects per spike was just the average number after Pass 1 plus the number present in the initial damage state.

* One potential source of error in operating HAP should be noted. A random noise generator is the ultimate source of the random numbers used in determining the order of defect selection, whether a jump shall occur, and the jump direction. This instrument was found to require frequent adjustment to compensate for long term drift. If this is not done, an observable trend for defects to migrate in certain preferred directions can result. The drift during a single run is considered negligible.

** An interstitial (vacancy) time step is defined as one-tenth the mean jump time of an uncorrelated single interstitial (vacancy).

This result is coincidental, however, because a weak trend toward lower production of defects in 8 of 9 spikes was exactly counterbalanced by a large increase in one spike (not 2051).

During the simulation of a short-term high temperature anneal, the evolution of the defect configuration from Pass 2 followed closely that of Pass 1. The most notable difference was the generally lower fraction of mobile defects throughout the simulation. The apparent annihilation of interstitials and vacancies dropped from 79% for both to 77% and 73% respectively. These differences, however, are almost wholly due to the initial damage state present for Pass 2. The number of annihilation events actually increased 5%.

The annealed configurations for several of the spikes are shown in Figure 5.7 (the initial damage state is shown for comparison). The cluster size distributions for Passes 1 and 2 are compared in Figures 5.8 and 5.9.

Several observations can be made concerning the differences in the distributions and the relation of these differences to the initial damage state. The initial large cluster, $A V_{21}$ (the largest cluster observed after Pass 1), grew in 5 spikes, shrank in 3, and remained dormant in 1. Final sizes of this cluster range from V_{15} to V_{33} , the latter occurring in No. 2051. The smaller vacancy cluster initially present, a V_5 , survived in 6 of 9 spikes. Although 2 vacancy clusters were present initially, the Pass 2 runs average 1.3 more clusters ($V_{\geq 4}$) than in Pass 1. The incidence of large vacancy clusters correlates roughly with the proximity of the PKA take-off direction to that of the PKA of spike No. 2051. It was noted above that the fraction of mobile defects was lower in Pass 2 than in Pass 1. Figure 5.8 makes it evident that this relative decrease of

B. NO. 2051, 2ND PASS
T.O. DIRECTION $\sim [10,3,5]$

D. NO. 2021, 2ND PASS
T.O. DIRECTION $\sim [3,2,6]$

A. INITIAL DAMAGE CONFIGURATION
(MODIFIED 1ST PASS OF NO. 2051)

C. NO. 2071, 2ND PASS
T.O. DIRECTION $\sim [9,3,1]$

Neg 701383-2

FIGURE 5.7. CRT Displays of Simulated 20 keV Displacement Cascades Produced in Previously Damaged Lattice and Subsequently Annealed. (Vacancies and interstitials indicated by 0 and X symbols respectively. Take-off point at origin in direction of arrow. Edge length of box is 50 half-lattice units.)

Neg 701383-3

FIGURE 5.8. Comparisons of Vacancy Cluster Distributions Produced in Simulated Displacement Cascades

Neg 701383-4

FIGURE 5.9. Comparisons of Interstitial Cluster Distributions Produced in Simulated Displacement Cascades

small clusters extends generally to vacancy clusters of size 15 or less. The fraction of vacancies in larger clusters increased significantly.

Pass 2 of spike No. 2051 produced the largest interstitial cluster (I_{12}) yet recorded in this work. It resulted through growth of the I_5 which was present initially. This I_5 survived in 7 of the 9 spikes but grew only in No. 2051. The other large interstitial clusters resulted from the growth of smaller clusters present in the initial damage state. In 6 of the 9 spikes, the interstitial cluster size distribution extends to larger sizes in Pass 2 than in Pass 1. The average number of interstitial clusters ($I_{\geq 3}$) per spike in Pass 2 is only 1.6 more than in Pass 1, although there were 4 clusters present initially. This fact, together with the $\sim 20\%$ decrease in the fraction of mobile interstitials, emphasizes the increase in average cluster size.

As discussed in the previous quarterly report,⁽³⁾ the HAP-4 code allows parameter variations to be made with relative ease. One of the Pass 2 runs was performed with the interstitial-vacancy recombination criteria replaced by a simple distance criterion. Previous work had shown that a recombination distance of $\leq \sqrt{19}$ hlu (half lattice units), extending through 7th neighbors and including 88 sites, was too small. For the present run, the distance was increased to $\leq \sqrt{20}$ hlu, extending through 8th neighbors and including 112 sites. The annealing behavior appears to be indistinguishable from the standard runs in which the 62-site, bow-tie shaped region shown in Figure 5.10 was used.

Neg 0695124-1

FIGURE 5.10. Recombination Regions for Frenkel Pairs

COMPUTER SIMULATION OF IRRADIATION PROCESSES

G. E. Russcher

The development of the SLIDES code continues through the debugging process. Several different test problems are being used to detect weak logic procedures and improper computational techniques.

As the code has been refined, many logic tests and diagnostic information sources have been added. These additions overloaded the available computer memory storage. Consequently, the code has been modified to use the Memory Allocation Processor (MAP), which now operates the SLIDES code in three segments. The main program and necessary common-data sub-routines are in continuous operation, but most of the sub-routines are segregated by function into:

- Lattice structure description and data preparation.
- Atomic interaction calculations.
- Edit, catalog, and output of the disarranged-lattice description.

COMPUTER TABULATION OF DOUNREAY FAST REACTOR FLUX AND REACTION RATE DATA

R. E. Dahl, Jr., R. D. Bourquin, and N. J. Graves

A great deal of experience in fast reactor fuels and structural materials has been gained through irradiations performed in the Dounreay Fast Reactor (DFR). A comprehensive report⁽¹⁷⁾ has been issued which gives the results [$\phi(E)$ for 16 energy groups in r, z matrix which is 28×23] of a multi-group calculation for fluxes in the DFR core and reflector regions. These data have been incorporated in a computer program, and neutron flux and spectrum can be recalled for any point of the matrix. Thus, a highly detailed, flux profile for any subassembly or pin in the reactor can be obtained very

quickly. This is very useful in analyzing stainless steel swelling data and fuel element dilatation and in studying fuel element modeling.

In addition to the flux data presented in the reference document, other sets of previously presented detailed data were incorporated into the library program. These include lineal heat generation rates for various fuel elements commonly irradiated in the DFR, fission rates for a number of fissile isotopes, activation rates for common flux monitors, and a tabulation of expected damage rates in stainless steel according to the Thompson-Wright model. Access to the data may be obtained by request to the dosimetry center.

COMPUTER SIMULATION STUDIES OF DISLOCATION CORE STRUCTURE

R. E. Dahl, Jr. and R. D. Bourquin

To quantify models of swelling and irradiation-induced creep in stainless steels, an experimenter requires a knowledge of atomistic scale parameters which define damage production and diffusion parameters in the disordered structure. Data on the strength of interstitial and vacancy sinks, which include grain boundaries, dislocation loops and voids, comprise part of the most needed information. Diffusion coefficients and activation energies for vacancy and interstitial migration to loops and voids, and data which would define a mechanism for void growth are also essential. Dislocation loops play an extremely important role in determining swelling and plastic deformation of metals. Therefore, an initial and extremely necessary step in providing the required data was to develop the facility to simulate dislocation core structure. The GRAINS⁽¹⁸⁾ program has been modified and tested so that edge dislocations, partial or complete, can be constructed and relaxed to equilibrium configurations.

So far the simulation has been limited to fcc gamma iron. The principal slip system [(111) plane, {112} direction] for fcc metals has been simulated by constructing cells in which the frontal planes are 112 planes and the dislocation lines are in a 112 direction. The dislocations are set up initially using the method of Doyama and Cotterill,⁽¹⁹⁾ and are then moved to positions dictated by either isotropic or anisotropic elasticity theory using the equations given by Gehlen, Rosenfield, and Hahn.⁽²⁰⁾ The atoms are then relaxed (moved) to equilibrium positions according to classical mechanics. The dislocation structure is maintained throughout the computational cell by a technique whereby the atoms which have three degrees of freedom (those within the interior of the computational cell) move and then are frozen, while atoms in crystallographic equivalent planes on the front and rear surfaces of the computational cell are moved to corresponding positions. The process is repeated until the potential energy of the computational cell is at a minimum. Figures 5.11 and 5.12 show the core structure before and after relaxation. Figure 5.11 is a (112) plane in which the atoms are located in positions dictated by isotropic elasticity theory according to their distance and direction from the dislocation. The atoms in Figure 5.12 have been relaxed, and this is a basic structure upon which the initial computer simulation studies for swelling and creep will be conducted.

In these figures, the hexagons are atoms which were completely moveable during the computations, and the squares represent atoms that were initially moved to their positions according to elasticity theory but which remain immobile during the relaxation process. The exact position of each atom falls in the center of its symbol, or at the end of the vertical centerline of the symbol.

The results obtained to date and illustrated in these figures corroborate the results of Cotterill and Doyama⁽¹⁹⁾ and

Neg 700240-13

FIGURE 5.11. Computer-Drawn Plot of Computational Cell Core Structure Before Relaxation

Neg 700240-17

FIGURE 5.12. Computer-Drawn Plot of Computational Cell Core Structure After Relaxation

of Gehlen et al.⁽²⁰⁾ in that the core region of a dislocation actually has a fairly high degree of order. This is also shown in Figure 5.13 which is a computer drawn plot of the plane just below the slip plane for the edge dislocation. (The three types of symbols show the ABC stacking of the fcc (111) planes.) This also illustrates that the atoms in the system relax to a fairly well ordered structure within close proximity to the dislocation core. The results shown may be of interest since atomic positions are located quite exactly through the plotting routine. The next stages in this investigation will be to determine changes in volume and pressure caused by the addition of interstitial atoms to jogs. This will have a direct relationship to swelling studies since the absorption of interstitials onto loops is a very probable mechanism causing dilatation.

FLUENCE DETERMINATIONS USING TIME-VARYING FLUX

R. D. Bourquin

Historically, flux and fluence have been calculated assuming a constant reactor power level for an irradiation. Since no reactor operates continuously, the values calculated for flux and fluence would be more precise if the effects of reactor cycling were considered. Thus, especially for a short half-life monitor, the flux calculated by assuming a constant power level could be significantly in error if the power level fluctuates.

For example, consider a hypothetical irradiation using monitors whose daughter nuclide half-life is (1) 10 days (typical of fission monitors) and (2) 300 days (typical of threshold monitors). In addition, assume:

- Spectral-averaged cross section of 1.
- Activity of 1.0 dps/nucleus.
- One hundred and fifty-day irradiation having a power history as shown in Figure 5.14.
- No burnout of daughter nuclide.

Neg 701383-8

A. Before Relaxation

Neg 701383-6

B. After Relaxation

FIGURE 5.13. Computer-Drawn Plot of Plane Just Below Slip Plane for Edge Dislocation

Neg 701383-1
FIGURE 5.14. Actual Power Level History of a Typical Irradiation Facility

The resulting ratios would be as shown below.

	<u>Short (10 day) Half-Life Monitor</u>	<u>Long (300 day) Half-Life Monitor</u>
$\frac{\phi(\text{constant power})}{\phi(\text{actual})}$	1.480	0.977
$\frac{\phi(\text{TIMH})}{\phi(\text{actual})}$	1.000	1.000

The first line gives the result of the constant power assumption and the second line the result corrected for reactor power history using the TIMH code. For a long half-life monitor then, either method is well within the error limits of activation counting data, but as seen for the short half-life monitor, accounting for actual power level fluctuations is necessary to avoid excessively large errors in flux and fluence calculations.

A computer program is now available that uses reactor power-history data for the determination of flux and fluence by foil activation analysis. This program, TIMH,⁽²¹⁾ has successfully calculated flux and fluence using activation data from EBR-II irradiations. Additional information may be obtained from the Dosimetry Center.

REFERENCES

1. A. L. Bement, G. J. Dau, E. A. Evans, and R. E. Nightingale. Quarterly Progress Report, April, May, June 1969, Reactor Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1144. Battelle-Northwest, Richland, Washington, August 1969.
2. Quarterly Progress Report, July, August, September, 1969, Reactor Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-1223. Battelle-Northwest, Richland, Washington, November 1969.

3. Quarterly Progress Report, October, November, December, 1969 Reactor Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, edited by W. F. Sheely, BNWL-1279. Battelle-Northwest, Richland, Washington, February 1970.
4. W. N. McElroy, R. E. Dahl, Jr., and C. Z. Serpan, Jr. "Damage Functions and Data Correlation," Nucl. Appl., vol. 7, p. 561. 1969.
5. C. Z. Serpan, Jr. and W. N. McElroy. Damage Function Analysis of Neutron-Energy and Spectrum Effects Upon the Radiation Embrittlement of Steels, NRL-6925. Presented at IAEA Symposium, Vienna, Austria, June 1969.
6. G. E. Russcher. Unpublished Data. Battelle-Northwest, Richland, Washington, 1969. (Personal Communication)
7. G. E. Russcher. Calculated Damage Functions for Determining Irradiation Effectiveness, BNWL-1093. Battelle-Northwest, Richland, Washington, 1969.
8. W. N. McElroy et al. A Computer-Automated Iterative Method for Neutron Flux Spectra Determination by Foil Activation, AFWL-TR-67-41, Vol I-IV. Air Force Weapons Laboratory, Kirtland AFB, New Mexico, July 1967.
9. W. N. McElroy et al. "Measurement of Neutron Flux Spectra by Multiple Foil Activation Iterative Method and Comparison with Reactor Physics Calculations and Spectrometer Measurements," Nucl. Sci. Eng., vol. 36, pp. 15-27. 1969.
10. W. N. McElroy et al. "Implications of Recent Fission-Averaged Cross-Section Measurements," Nucl. Sci. Eng., vol. 36, pp. 110-113. 1969.
11. W. W. Little, Jr. and R. W. Hardie. 2DB, A Two-Dimensional Diffusion-Burnup Code for Fast Reactor Analysis, BNWL-640. Battelle-Northwest, Richland, Washington, 1968.
12. See pp. 5.5-5.7 in Reference 3 above.
13. See p. 15 in Reference 9 above.
14. L. S. Kellogg. Unpublished Data. Battelle-Northwest, Richland, Washington, 1969. (Personal Communication)
15. See pp. 5.4-5.5 in Reference 3 above.

16. A. L. Bement, G. J. Dau, E. A. Evans, and R. E. Nightingale. Quarterly Progress Report, January, February, March 1969, Reactor Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1058. Battelle-Northwest, Richland, Washington, May 1969.
17. H. Atkinson. "Reaction Rate Tables for the Dounreay Fast Reactor: Run 59/1," PRG Memorandum 4042 (D). United Kingdom Atomic Energy Authority, Risley, Lancs, England.
18. R. E. Dahl, Jr., J. R. Beeler, Jr., and R. D. Bourquin. GRAINS -- A Quasi-Dynamic Code for Computer Simulation Experiments of Point Line Defects in Metals, BNWL-1248. Battelle-Northwest, Richland, Washington, January 1970.
19. R. M. J. Cotterill and M. Doyama. "Energy and Atomic Configuration of Complete and Dissociated Dislocations. I. Edge Dislocation in an fcc Metal," Phys. Rev., vol. 145, no. 2, p. 465. May 1966.
20. P. C. Gehlen, A. R. Rosenfield, and G. T. Hahn. "Structure of the $\langle 100 \rangle$ Edge Dislocation in Iron," J. Appl. Phys., vol. 39, no. 11, pp. 5246-5254. October 1968.
21. R. D. Bourquin. Unpublished Data. Battelle-Northwest, Richland, Washington. (TIMH -- A Computer Program for the Determination of Fluence with a Time-Varying Flux, to be published.)

6.0 FAST REACTOR ABSORBER MATERIALS

G. L. Tingey

RADIATION EFFECTS ON BORON CARBIDE AND TANTALUM

A. L. Pitner and D. E. Baker

Irradiation in a Thermal Flux

The irradiation of boron carbide at 500 to 1200 °F in Hanford thermal reactors has been completed. The boron carbide samples were in five forms: 60 and 80% TD powders, and 65, 80, and 99% TD pellets. Pellet sample size was nominally 0.25 in. diameter $\times$ 1 in. long. The irradiations were conducted in two subassembly groups. Subassemblies 1, 2, and 3 operated at temperatures of 550 to 1200 °F, while subassemblies 4, 5 and 6 ran at 500 to 750 °F. Subassemblies 1, 2, and 4 have thus far been disassembled and examined for helium gas release and B^{10} burnup. Mass-spectroscopic analyses of isotopic ratios indicate that the average exposure for Subassemblies 1, 2, and 4 are 7×10^{20} captures/cm³ ($\sim 4\%$ B^{10} burnup), 21×10^{20} captures/cm³ ($\sim 12\%$ B^{10} burnup), and 15×10^{20} captures/cm³ ($\sim 8\%$ B^{10} burnup), respectively. Because of the high thermal cross section of B^{10} , however, nearly all of the reaction occurs on the surface of the samples, while the inner cores remain relatively unreacted. Consequently, burnup in affected areas is always very nearly 100%, and the gas release fractions are not expected to vary significantly with total irradiation exposure.

Gas release values for the three subassemblies are shown in Figure 6.1. Each data point is accompanied by a number indicating which subassembly the sample resided in. In general, the results from Subassembly 1 show higher gas release fractions than do the data from Subassemblies 2 and 4. Since Subassembly 1 had the lowest exposure, the total amount of gas generated was less than in the other two subassemblies, and the accuracy of

the gas pressure measurements would be correspondingly lower. Consequently, these values are probably the least reliable and were ignored in drawing the curves. The 99% TD pellets release the lowest fraction of gas. The 65 and 80% TD pellets do not appear to vary significantly in their gas release behavior, but both release a higher gas fraction than the 99% TD pellets. The percent of gas released from boron carbide appears to increase rapidly when the irradiation temperature exceeds 1000 °F. There is considerable scatter in the gas-release values for the powders, and no attempt was made to draw a curve through them. The gas-release fractions in the powders are, however, generally higher than in the pellets.

Postirradiation examination will continue on remaining Subassemblies 3, 5, and 6. The estimated exposure for each of these is 25×10^{20} captures/cm³ (~14% B¹⁰ burnup). Swelling measurements and postirradiation annealing/gas release studies are also planned for most of the samples.

Apparatus for Annealing Studies of Boron Carbide

An apparatus was constructed to measure helium gas release from boron carbide during postirradiation annealing. The apparatus consists of a high vacuum system, gas burette, manometer, and Toepler pump. The sample is heated by either a 10 kW induction generator or a resistance tube furnace. Boron carbide samples were heated to 2100 °C and the evolved helium was transferred with a Toepler pump to a calibrated volume for gas pressure measurement. Initial operation of the apparatus indicates that it will perform as designed and will be useful in studies of the kinetics of helium release from irradiated boron carbide.

Fast Reactor Irradiation

When the EBR-II tantalum capsule TA-4 was disassembled and the inner capsule cladding rupture observed, it was determined

that an identical capsule, TA-5 with the same irradiation history was probably similarly ruptured. Analysis of a gas sample taken from the annulus between inner and outer capsules confirmed the rupture. The TA-5 capsule was subsequently disassembled and the rupture observed in a position corresponding to that of one of the sets of melt wires.

Although TA-4 contained only melt wires for temperature monitoring, TA-5 contained a silicon carbide temperature monitor located at the hottest position in the capsule in addition to the melt wires. Postirradiation annealing of the SiC pellet indicated a maximum irradiation temperature of 2100 °F, a temperature considerably higher than 1650 °F which was estimated from heat transfer calculations.

The length and density of each of the samples in TA-5 have been measured, and the results are shown in Table 6.1. Previous measurements from TA-4 are also included for comparison. The results from the two capsules agree reasonably well and show an unusually high temperature dependence. Samples irradiated at the lower temperatures exhibited the greatest swelling, even though their exposure were lower than the higher-temperature samples.

Metallography was performed on the tantalum holder that contained the silicon carbide pellet in TA-5 to determine if any interaction occurred during the irradiation. No definite indication of interaction was observed. A out-of-reactor test at 1000 °C also continues to support the compatibility of tantalum with silicon carbide. After 1000 hr at temperature, weight changes in the silicon carbide were within the accuracy of the weighing.

The tantalum replacement capsules, BTA-6 and BTA-8, have been loaded into the X042 subassembly in EBR-II and are now under irradiation. Based on the temperature measurement from

the SiC pellet in TA-5, tantalum samples in BTA-6 and BTA-7 are expected to reach maximum temperatures of 2300 °F. Silicon carbide monitors have been included throughout the length of these capsules and should yield more accurate temperature profiles along the capsules. After BTA-6 has received an exposure of $\sim 10^{22}$ nvt, it will be replaced with an identical capsule (BTA-8) and be disassembled. Upon completion of the irradiation, BTA-7 and BTA-8 will have received exposures of ~ 3.4 and 2.4×10^{22} nvt, respectively.

TABLE 6.1. Swelling of Tantalum Irradiated in EBR-II

Sample	Est. Exposure (E > 0.1 MeV)	Temp., °F	TA-4		TA-5	
			ΔL , %	$\Delta \rho$, %	ΔL , %	$\Delta \rho$, %
1	3.0×10^{21}	1325	+0.13	-0.69	+0.18	-0.88
2	4.6×10^{21}	1600	+0.13	-0.68	+0.13	-0.72
3	6.7×10^{21}	1875	+0.11	-0.85 ^(a)	+0.07 ^(a)	-0.47
4	8.2×10^{21}	2025	-	-0.58	+0.06	-0.53
5	8.6×10^{21}	2100	+0.01	-0.42	+0.02	-0.29
6	8.2×10^{21}	2025	+0.05	-0.45	+0.03	-0.25 ^(a)
7	6.7×10^{21}	1875	+0.06	-0.62	+0.10 ^(a)	-0.61
8	4.6×10^{21}	1600	+0.09	-0.63	+0.11	-0.82 ^(a)
9	3.0×10^{21}	1325	+0.13	-0.72	+0.09 ^(a)	-0.58

(a) *Plating from nearby melt wire apparently caused measurement to be in error.*

FIGURE 6.1. Helium Gas Release of Boron Carbide Irradiated in a Thermal Flux

7.0 NUCLEAR GRAPHITE

G. L. Tingey

DIMENSIONAL CHANGES OF GRAPHITES AT HIGH NEUTRON FLUENCES AND HIGH TEMPERATURES

A. L. Pitner and W. J. Gray

Conventional nuclear graphites as well as several experimental graphites are being irradiated to very high neutron fluences at high temperatures primarily to characterize the dimensional changes that take place under these conditions. Data obtained so far have disclosed two noteworthy items:

- Post-turnaround expansion of at least some graphites is less rapid at temperatures above 1200 °C than at temperatures of 900 to 1200 °C.
- Graphites manufactured by POCO Graphite Inc., exhibit smaller dimensional changes and less rapid expansion at high fluences than do conventional graphites.

Figure 7.1 shows data at three temperatures for NC8 graphite, a conventional needle-coke grade. The 650 to 750 °C data have been previously published and are included only for comparison. At 1200 to 1300 °C the post-turnaround expansion rate in the perpendicular direction is very much less than at 950 to 1050 °C. Our data indicate that the marked temperature dependence occurs around 1200 °C, but in the range from 900 to near 1200 °C, the curves approximate that shown for 950 to 1050 °C. Suddenly, however, at temperatures above 1200 °C, the post-turnaround expansion rate is markedly reduced.

The data in the parallel direction at high temperatures is badly scattered. Sample numbers have been included beside some of the data points so that the length-change trends can be more readily seen. Initial contraction rates are greater at 1200 to 1300 °C than at 950 to 1050 °C. Post-turnaround behavior has not yet been well established. However, the

FIGURE 7.1. Irradiation-Induced Length Changes of NC8 Graphite

mechanisms that operate to produce the slow post-turnaround expansion rate at 1200 to 1300 °C in the perpendicular direction should also apply to the parallel direction.

The high-temperature data shown for NC8 graphite in Figure 7.1 is typical of at least three grades as follows:

- CSF, a non-needle coke graphite.
- RC5, a gilsonite coke graphite.
- RP4, a composite of both needle and non-needle cokes.

The most likely explanation for the slow post-turnaround expansion rate at high temperatures seems to be related to irradiation-induced creep. At all temperatures, rapid expansion of the crystallites in the c-axis direction is partially accommodated at low fluences by Mrozowski cracks that form during cool-down from graphitization temperatures. This, together with contraction of the crystallites in the a-axis direction, results in net contraction for the polycrystalline graphite. The Mrozowski cracks, however, eventually become filled. Further expansion of the crystallites in the c-axis direction and contraction in the a-axis direction can then only be accommodated by formation of new cracks or by localized plastic deformation. At temperatures below ~1200 °C, the irradiation creep constant is apparently not high enough to prevent crack formation. Therefore, the graphite expands rapidly. Above 1200 °C, enough plastic deformation is possible to accommodate the crystallite strains so that no cracks are formed and only a very slow net expansion is observed.

The seven grades of POCO graphite that are being irradiated in this program are generally described as fine-grained, isotropic, high-strength, high CTE materials. They vary in density from 1.5 to 1.9 g/cm³, and have experienced graphitization temperatures of 2300 to 2800 °C. All seven grades show generally similar irradiation behavior that is typified in Figure 7.2, which presents the results for AXF-8Q1. This

FIGURE 7.2. Irradiation Induced Length Changes of POCO Graphite AXF-8Q1

graphite was graphitized at 2800 °C and has a density of 1.8 g/cm³. The dotted curves in the figure show the comparative behavior of a conventional, needle-coke graphite, NC8 at 1000 °C. At irradiation temperatures around 1100 °C, the POCO graphite shows expansion from the onset of irradiation. This expansion, however, saturates after an exposure of $\sim 5 \times 10^{21}$ n/cm², and the material does not begin expanding significantly again until the fluence exceeds $\sim 10^{22}$ n/cm². The expansion rate in this latter stage is seen to be considerably less than that shown by conventional graphites after turnaround in this temperature regime. Above 1200 °C the POCO graphite contracts slightly with irradiation until a fluence is reached of $\sim 10^{22}$ n/cm², after which the trend appears to be toward slow expansion. The maximum linear contraction of the various POCO grades is usually on the order of 1%.

COMPUTER CALCULATIONS OF GRAPHITE OXIDATION IN HTGR

R. C. Giberson

Additional data have been received from Gulf General Atomic concerning the 1100 MW_e HTGR. This data indicates a leak rate of 4.2×10^{-2} lbs of H₂O/hr and a purification factor of 9.9×10^{-5} fraction per cycle. Using this new data we have reevaluated the steady-state gas compositions to be anticipated in the 1100 MW_e reactor. Table 7.1 presents the results of these evaluations compared with the 330 MW_e (PSC) reactor.

The steady-state water vapor in an HTGR is given approximately by:

$$PH_2O_{ss} \approx \frac{PH_2OLK}{\Delta W + PF}, \quad (1)$$

where: PH_2OLK = water in leakage rate.

PF = purification factor (fraction of gas purified per cycle).

ΔW = change in water vapor in passing through the reactor core.

For the reactions considered in the present computer program,⁽¹⁾ any change in water vapor concentration gives an equal but opposite change in hydrogen concentration; therefore, the hydrogen steady state is:

$$PH_{2ss} \approx \frac{\Delta W (PH_2O)_{ss}}{PF} \quad (2)$$

and
$$\left(\frac{PH_2}{PH_2O} \right)_{ss} \approx \frac{\Delta W}{PF} \quad (3)$$

TABLE 7.1. Steady-State Gas Compositions in HTGR's

Thermal Reactions only, K = 1.0K^(a)

Reactor	Concentrations in mm					
MW _e	H ₂ O	CO	CO ₂	H ₂	H ₂ /H ₂ O	ΔW
330 ^(b)	7.24 × 10 ⁻²	2.86 × 10 ⁻¹	6.1 × 10 ⁻⁴	2.87 × 10 ⁻¹	3.96	1.06 × 10 ⁻³
1100	1.16 × 10 ⁻²	3.25 × 10 ⁻¹	9.2 × 10 ⁻⁴	3.28 × 10 ⁻¹	28.3	2.80 × 10 ⁻³

a. Refers to normal TSX graphite rate constants.

b. Values reported for the 330 MW_e are slightly different than given in the previous quarterly. This is a result of a change in the temperature simulation equation.

As shown in Table 7.1, calculated by the computer program, ΔW is significantly higher in the 1100 MW_e than in the 330 MW_e reactor. This is a result of the higher graphite temperatures in that reactor.

To a first approximation, Equation (3) can be used to determine the effect of increased purification system capacity on the hydrogen/water vapor ratio. It also suggests that this ratio is relatively independent of leak rate. However, precise effects must be calculated because ΔW is not immune to water leakage.

The results given in Table 7.1 are calculated neglecting radiation-induced reactions. It is noted, however, that radiation effects increased the H_2/H_2O ratio in the 330 MW_e HTGR,⁽¹⁾ and a similar effect is expected for the 1100 MW_e. Data have also been obtained from GGA on some postulated accident conditions. These will be analyzed and reported in the next quarterly.

RADIOLYSIS OF H_2 - CO_2 -He MIXTURES

D. E. Clark and G. L. Tingey

The apparent stability of CO_2 toward ionizing radiation can be attributed to the rapid recombination of intermediates.⁽²⁾ However, in the presence of chemical scavengers which interfere with the recombination reactions, decomposition of CO_2 does occur to a significant extent. As an example of such a substance, hydrogen gives the overall reaction $H_2 + CO_2 \rightarrow CO + H_2O$. We have studied the reaction of equimolar amounts of H_2 and CO_2 in helium, the reactive gas concentrations ranging from ppm levels to 50 mole%. Both static and single-pass flow methods of irradiation were used and gave comparable results.

The relative yield of CO, $Y(CO)$, is shown in Figure 7.3 as a function of reactant concentration. The dose rate must be known in order to calculate the G-value for the reaction (the number of molecules formed per 100 electron-volts absorbed). We have determined the dose rate with two different chemical dosimeters, the formation of N_2 from N_2O [$G(N_2) = 9.3$] and H_2 from CH_4 [$G(H_2) = 5.7$]. Agreement between the two was good, giving a value for the current dose rate of 5.5×10^{17} electron-volts per second per mole of electrons. With reference to Figure 7.3, then, this leads to the expression $G(CO) = 0.018 Y(CO)$.

FIGURE 7.3. Irradiation of Flowing Gas Stream Yield of CO (Relative Units)
Versus CO₂ Concentration

In mixtures containing less than about 10,000 ppm of reactants, energy is absorbed mainly by helium which results in energy-rich forms of He*.

These may be de-excited through the emission of radiation

or through the transfer of energy to reactive species

which then undergo chemical change to form the observed products. The latter reaction occurs at a significant rate down to the ppm level of reactive gas concentration, thus implicating relatively long-lived metastable states of atomic or molecular helium as the energy-transfer species. At these concentration levels, the rate of direct activation of the reactants, such as by the reaction $\text{CO}_2 \xrightarrow{\gamma} \text{CO}_2^*$, is relatively slow and can be ignored. Our results also indicate that transfer of energy to H_2 does not significantly affect the reaction rate, but rather that the rate depends primarily on the transfer of energy to CO_2 .

We are presently studying the effect of pressure and temperature on the observed reaction. Assuming a steady-state concentration of He*, the following expression is obtained:

$$\frac{d}{dt} (\text{CO}) \approx \frac{k_1 k_4 (\text{He}) (\text{CO}_2)}{k_2 + k_4 (\text{CO}_2)} + k_3 (\text{CO}_2)$$

Thus, depending on the relative values of k_1 , k_2 , k_3 , and k_4 , the overall reaction rate should depend on the pressure of He, P_{He} , or on $P_{\text{He}} \times P_{\text{CO}_2}$ in the concentration range where one mode of de-excitation predominates over the other. The yield was shown to increase with pressure in the range of ~ 1000 ppm but does not obey the simple P or P^2 relationship. At lower relative concentrations, it is expected that a linear dependence on P will be observed.

REFERENCES

1. R. C. Giberson and G. L. Tingey. Reaction of Gaseous Impurities in a High Temperature Gas-Cooled Reactor, BNWL-974. Battelle-Northwest, Richland, Washington, December 16, 1968.
2. A. R. Anderson and L. A. Dominey. "The Radiolysis of Carbondioxide," Radiation Research Reviews, vol 1, no 3, pp. 269-300. October 1968.

8.0 NONDESTRUCTIVE TESTING

EDDY CURRENT METHODS

H. L. Libby

A simulated practical tube testing problem has been solved using the multiparameter eddy current tester. When tubes must be tested after they are installed in a system, large signals are often received from known causes such as tube supports or iron inclusions in concrete surrounding the tube. It is often impossible to test for flaws on the inner (ID) and outer (OD) surfaces of a tube which are near or under a tube support by using a conventional single frequency eddy current tester. This study shows that the multiparameter eddy current tester can be used to test for flaws on both the inner and outer surface of a tube even though the flaws are under simulated tube supports. However, this report does not intend to imply that the study shows that all eddy current testing problems can be solved by using the multiparameter eddy current tester.

A Hanford prototype multiparameter eddy current tester was used to demonstrate its advantage over the use of a conventional single frequency tester for detecting flaws in tubing which occur coincident with tube supports. The same tester, flaw standard, probe and simulated tube supports were used for all of the tests described in this study. For the single frequency test, the tester was operated at a single frequency (125 kHz). For the multiparameter tests, the tester was operated at 125 kHz and 250 kHz, simultaneously. The flaw standard was a piece of 0.5 in. ID, 0.050 in. wall, 304 SS tubing with a 0.023 in. deep OD notch, 0.015 in. deep ID notch, a kink, and an area of suspected ID intergranular corrosion. Two simulated tube supports were used: (1) a piece of flat, 3/8-in. thick mild steel, and (2) a piece of 1/2-in. thick, flat stainless steel, each with a 41/64 in. diameter hole in it. The simulated tube supports fit the tube loosely enough

to be moved along the tube, but were not a sloppy fit. The test probe was a standard encircling type, inside differential coil arrangement.

The in-phase and quadrature signal component channels of a single frequency (125 kHz) eddy current tester are shown in Figures 8.1 and 8.2. The signals of each channel, from left to right, are caused by:

- 0.023 in. deep OD notch.
- Mild steel simulated tube support.
- Stainless steel simulated tube support.
- Suspected ID intergranular corrosion.
- Kink.
- 0.015 in. deep ID notch.

The tester was adjusted to produce minimum signal output for the mild steel tube support on the lower trace in both Figures 8.1 and 8.2. In Figure 8.1 all of the signals are separate, distinct signals. Notice that even though discriminated against, the mild steel tube support produces a large residue signal on the lower trace. Next, the mild steel tube support was physically placed over the OD notch giving the results shown in Figure 8.2. Now, the iron tube support signal and the OD notch signal appear together, and the OD notch signal is unreadable.

Figures 8.3 and 8.4 are strip chart recordings of the output signals from the multiparameter eddy current tester operated simultaneously at 125 and 250 kHz. The same tube standard, simulated tube supports and test probe were used for these tests as were used for those related to Figures 8.1 and 8.2. The top trace in Figures 8.3 and 8.4 is a reference trace and is used only to show the relative position of the tube supports with respect to the notches. Notice in Figure 8.3 that all flaws and supports occur at distinct (separate) positions and that both of the tube supports signals have been discriminated

FIGURE 8.1. Two-Channel Output of a Single-Frequency Eddy Current Tester Showing Null Against the Mild Steel Tube Support Signal on the Lower Trace

FIGURE 8.2. Two-Channel Output of a Single-Frequency Eddy Current Tester When the OD Notch is Under with the Mild Steel Tube Support

FIGURE 8.3. Multiparameter Eddy Current Tester Output Showing Discrimination Against Mild Steel and Stainless Steel Tube Support Signals on the Lower Trace

FIGURE 8.4. Multiparameter Eddy Current Tester Output Showing OD Notch Signal and the ID Intergranular Corrosion Signal when the Mild Steel Tube Support is over the OD Notch and the Stainless Steel Tube Support is over the ID Intergranular Corrosion Flaw

against on the lower trace, leaving small residues. In comparing the lower trace of Figure 8.4 to the lower trace in Figure 8.3, notice that the OD notch signal and ID corrosion signal in Figure 8.4 are essentially the same as in Figure 8.3, even though the mild steel tube support has been moved over the OD notch and the stainless steel tube support has been moved over the intergranular corrosion area on the tube.

Figures 8.5 and 8.6 were made with the same test equipment, standards and probe as were Figures 8.3 and 8.4. The top traces in each figure are again only reference traces used for showing the position of the tube supports with respect to the notch signals. The lower trace in Figure 8.5 shows that both tube support signals and the OD notch signal have been discriminated against. The residues from these signals are very small. Here again the tube support signals and the notch signals are separate. In Figure 8.6 the stainless steel support has been moved over the intergranular corrosion flaw. Notice that the intergranular corrosion signal appears essentially the same on the lower trace in Figure 8.6 as it did in Figure 8.5.

Figures 8.7 and 8.8 are photographs of the complex voltage waveforms for all of the signals shown in Figures 8.1 through 8.6. Figure 8.7 was made using a test frequency of 125 kHz and a differential probe. Figure 8.8 was made using a test frequency of 250 kHz with the same probe. The probe motion axis is horizontal in each of these figures. The two largest signals in Figures 8.7 and 8.8 are from the tube supports. The stainless steel tube support signal is the largest in both figures. The magnitude of the stainless steel tube support signal is approximately 15 times larger than the largest notch or intergranular corrosion signal at 125 kHz. All of the notch, kink, and corrosion signals appear clustered at the center of the photograph. These two figures are intended to show the relative magnitudes and nonlinear nature of the signals.

FIGURE 8.5. Multiparameter Eddy Current Tester Output Showing Discrimination Against OD Notch Signals, Mild Steel Tube Support Signals and Stainless Steel Tube Support Signals on Lower Trace When Signals Occur Separately

FIGURE 8.6. Multiparameter Eddy Current Tester Output Showing the Intergranular Corrosion Signal When the Stainless Steel Tube Support Is Over the Intergranular Corrosion Flaw

FIGURE 8.7. Complex Voltage Waveforms for Single Frequency Eddy Current Test at 125 kHz

FIGURE 8.8. Complex Voltage Waveforms for Single Frequency Eddy Current Test at 250 kHz

ULTRASONIC METHODSPhase Measurements of Reflected Ultrasonic Waves near the Rayleigh Critical Angle

F. L. Becker

The nature of the reflected ultrasonic wave at the Rayleigh critical angle has been the subject of much research. Several researchers^(1,2,3) have measured the sharp minimum which occurs at the Rayleigh critical angle but have been unable to explain this effect with a suitable model. Others⁽⁴⁾ have investigated the phase of the reflected wave for cases of low material attenuation or low frequencies. Through our research of the effects of attenuation on the propagation of ultrasound,⁽⁵⁾ we have developed a mathematical model⁽⁶⁾ which describes both the phase and amplitude of the reflected wave. Verification of the model's accuracy in predicting the amplitude has been previously reported.^(7,8) This work presents phase measurements which fully confirm the predictions of our model. These predictions are in agreement with those formulated by V. M. Merkulova⁽⁹⁾ and G. Mott.⁽¹⁰⁾

These predictions differ substantially from nonattenuating theories in that the phase and rate of change in phase with incident angle are sensitive to attenuation and frequency in the reflecting medium. Above the frequency of least reflection, the phase no longer shifts from approximately 0 to +360°, rather it shifts from 0 to +60 then -60° and back to zero again. The frequency of least reflection (FLR) is the frequency for which the reflection at the Rayleigh critical angle is a minimum, approximately zero. This frequency has been found to be dependent on the shear wave attenuation per wave length in the reflecting media.⁽⁸⁾

The phase and amplitude of a plane wave reflected at a liquid solid interface (as in References 6-8) can be written as follows:

$$\frac{z_{112}}{z_{111}} = \frac{\left(\frac{C_{22}}{C_{12}}\right)^2 \sin 2T_{221} \sin 2T_{121} + \cos^2 2T_{221} + \frac{\rho_1 C_{11}}{\rho_2 C_{12}} \frac{\sin T_{121}}{\sin T_{111}}}{\left(\frac{C_{22}}{C_{12}}\right)^2 \sin 2T_{221} \sin 2T_{121} + \cos^2 2T_{221} - \frac{\rho_1 C_{11}}{\rho_2 C_{12}} \frac{\sin T_{121}}{\sin T_{111}}} \quad (1)$$

Equation (1) is very similar to those found in existing theories^(1,11) except that the velocities C_{ij} and angles T_{ij} can be complex, which results in a complex reflection factor. The amplitude and phase are the modulus and phase, respectively, of this complex number. The development of and solutions for this equation are given in detail in other reports.^(6,8)

Measurements of the change in phase of a reflected wave with angle requires that the ultrasonic path length be very stable; for example, an error of 0.001 in. would result in a phase change error of 92° at 15 MHz. To overcome this limitation, a dual angle goniometer was designed and constructed which has an error of less than ±80 μin. between 30 and 35°, which is equivalent to ±7° phase change at 15 MHz. This error was evaluated by measuring the phase shift from a brass sample which has a phase change of less than 1° over this angle range. The physical details and operation of this device are similar to that described by Fountain.⁽¹²⁾

The phase was measured by the same technique described by VanBuren and Breazeale,⁽⁴⁾ with two exceptions: first, single frequency pulse bursts of 10 μsec duration were used and second, a vector voltmeter was used for phase readout. The use of the voltmeter has two advantages: (1) a calibrated analog output is obtained and (2) the actual phase lead or lag is measured, rather than the difference in phase (as in the case where Lissajous patterns are used). In this method, the phase of a cw reference signal is adjusted to match the received pulse,

and this shifted signal is then measured relative to an unshifted reference signal, point by point for each incident angle.

The feasibility of using a cw ultrasonic signal was also investigated and found to be equally suitable and much faster than the pulse method. In this technique, the phase of the cw signal, relative to a fixed reference, was read directly by the voltmeter and recorded automatically as the incident angle was varied. Within the limits of the stability of the measurement (approximately $\pm 5^\circ$ due mostly to vibration and electrical noise), the two methods gave the same results.

The results of the phase measurements for frequencies of 10, 14.5 and 18 MHz are shown in Figure 8.9 for Type 304 SS. The cw method was used for these curves. The reflected amplitude was also recorded to show its relationship to the phase at the Rayleigh critical angle (minimum amplitude point). At 10 MHz the phase changes from approximately 0 to 360° and is 180° at the critical angle. These results are similar to existing theories. Significant differences occur at higher frequencies. For a frequency of 14.5 MHz which is slightly below the FLR (note amplitude minimum), the slope becomes very steep. Above the FLR, 18 MHz, the phase rises to approximately $+60^\circ$, falls to -60° and then approaches zero as the incident angle passes through critical.

It is easy to see that the phase change is indeed a phase lead by observing a pulse signal on an oscilloscope relative to a fixed reference. Figure 8.10 depicts these phase changes for small changes in the incident angle for frequencies of 10 and 18 MHz. Although the changes are difficult to determine from this figure, it is possible to show that the transient portion of these signals (first one or two cycles) does not move forward in time but that the steady-state portion does. Previous negative phase predictions⁽⁸⁾ are actually consistent

Neg 701343-4

FIGURE 8.9. Reflected Phase (ϕ) and Amplitude for a Water Type 304 SS Boundary. Frequencies of 10, 14.5 and 18 MHz Are Shown

Neg 701343-1

FIGURE 8.10. Received Ultrasonic Signal Showing Phase Advance as the Incident Angle is Varied Through Critical. Both 10 and 18 MHz Pulses Are Shown Relative to a Fixed cw Reference Signal

with these results, if the definition of the traveling wave used is considered. The wave, $\exp i\omega\left(\frac{\vec{N}\cdot\vec{R}}{c} - t - \phi\right)$, was used and in this system a negative ϕ actually represents a phase lead in time.

The anomalies (slope changes and reversals) noted above can be explained by using a model such as Equation (1) which considers the attenuation in both media. The predicted and measured phase changes in the area of the critical angle are shown in Figure 8.11 for Type 304 SS in water. The attenuations and other material constants used in these calculations are given in Reference 8. Two facts should be noted from this figure. First, the general shapes of the predicted and measured phases are very similar. Second, the slope of the phase curve decreases for frequencies away from the FLR. The difference in angular position of the measured curves is caused by a thin surface layer of work-hardened material (which occurred during the sample preparation) and results in a critical angle shift.⁽¹³⁾ This shift is dependent on the frequency, as the depth of penetration of these waves depends on the wave length.⁽¹⁴⁾

The otherwise good agreement between measured and predicted phase along with the previously verified amplitude relationships substantiates the accuracy and applicability of the model. The sharp phase change for frequencies just below the FLR present several possibilities for materials testing or characterization. The Rayleigh critical angle has been shown^(8,13) to be sensitive to changes in material properties such as elastic modulus and density. The steep slope of the phase curve would be capable of much higher resolution than existing methods which use amplitude detection only.

Neg 701343-3

FIGURE 8.11.

Calculated and Measured Phase of Reflected Ultrasonic Waves at a Water Stainless Steel Interface. Frequencies of 10, 12, 14, 15, 18 and 20 MHz Are Shown.

Ultrasonic Measurement of Cold Work

R. L. Trantow

Our preliminary studies of the application of Rayleigh critical angle techniques to cold work and grain size determinations in Type 316 SS have established this test as a very sensitive indicator of a material's metallurgical state. The studies have also shown, however, that the Rayleigh critical angle and reflected beam attenuation are sensitive to specimen alignment and to changes in the transmitted beam configuration. Because of the high degree of curvature in the actual fuel cladding, the effects of alignment and beam geometry become very critical. Therefore, to ensure the accuracy of the critical angle measurement on our present flat plate specimens, and eventually on the actual fuel cladding, a two-part program was initiated. First to be developed was a reliable alignment procedure to be used with our present dual angle goniometer. The second part of the program, which is continuing presently, involves the study of the actual transmitted ultrasonic beam to determine its spatial energy distribution and to study the effect of that distribution on the reflected critical angle beam shape. This second part of the program will be discussed more fully here.

During early critical angle measurement of our characterized Type 316 SS flat specimens, we noticed that in some combinations of transmitter and receiver transducers, the critical angle null region of our data became distorted. In some cases, the distortion would reach proportions where multiple or false null points could be obtained. Our initial studies into the causes of this distortion were directed toward scanning the transmitter beam to determine if anomalies in beam symmetry or uniformity were present. One of our 1-in. diameter transducers (one that had presented some problems in system alignment and data uniformity) was selected for beam scanning. An isometric

representation of the energy distribution in a plane normal to the beam axis and 3.9 in. from the transducer face is shown in Figure 8.12. In this representation, the high amplitude peaks have been clipped to more clearly show the lack of symmetry and beam nonuniformity. This anisometric effect is probably due to areas of reduced sensitivity caused by lense unbonds or dead spots in the transducer material. Near-field effects and internal reflections are highly probable contributors as this scanning distance is well within the near field region. However, their effects would be expected to produce a more uniform beam interference. Although the reflected critical beam resulting from this transducer has not been scanned as yet, it appears safe to assume that the perturbations in the transmitted beam will be present in the reflected beam. Therefore, their presence will partially explain the distortion and multiple nulls in the critical angle data which were taken. An example of a uniform and symmetric beam energy distribution is shown in Figure 8.13.

FIGURE 8.12. Energy Distribution Scan of a 15 MHz 1 in.-Diameter Transducer with Tone Burst Excitation

FIGURE 8.13. Symmetrical Energy Distribution of a 15 MHz 1/4 in.-Diameter Transducer with Tone Burst Excitation

This photograph shows an isometric representation of the energy distribution from a smaller, 1/4-in. diameter PZT transducer driven by a 15 MHz tone burst. As the scanning distance is approximately 4 in. away from the transducer face coinciding with the first beam maximum, this scan is in the far-field zone. The critical reflection resulting from bouncing this beam off a stainless steel block has also been scanned (see Figure 8.14). The notch or saddle between the two peaks is the result of critical reflection, and as the critical angle of incidence is approached, it can be seen to sweep through the peak of reflected energy. The null occurs over a very small range of incident beam angle and is, therefore, a very good indicator of the critical reflection condition. It is interesting to note that there are a series of secondary peaks or interferences scattered about the bases of the two major peaks. These are possibly caused by reflections within the specimen and/or by transducer near-field effects since in this experiment the beam-plate interception point occurred well within the transmitted beams near-field zone.

Since the possibility still exists that these side lobes may interfere with the saddle position and with the minimum amplitude through superposition of energy, we attempted to minimize the near-field effect by applying a pulse rather than a tone burst to the transmitter transducer. The energy distribution resulting from a 10 MHz period pulse applied to the same 1/4-in. transducer discussed above is shown in Figure 8.15. The energy distribution now became much broader and very smooth. The scan of critical reflection energy distribution of the initial beam is shown in Figure 8.16. The scale of this scan is the same as all previous scans shown in Figures 8.13 through 8.15. A broad smooth saddle point appeared and seemed to be free of extra interfering radiation lobes. Although the pulsed critical angle technique appears

FIGURE 8.14.

Energy Distribution of
the Critical Reflection
of the Beam Described
in Figure 8.13

FIGURE 8.15.

Energy Distribution of
the Transducer with a
10 MHz Period Pulse
Excitation

FIGURE 8.16.

Energy Distribution of
the Critical Reflection
of the Beam Described
in Figure 8.15

to yield the most uniform and interference-free data, it must also be realized that the position and depth of the critical angle null point is frequency sensitive and that a pulse has high harmonic content.

Flaw Identification and Characterization

N. E. Dixon

An ultrasonic signal reflected from an object is believed to contain all the necessary geometric properties of the object such as size, shape, contour, etc. and that by processing the data in an appropriate manner (unknown as yet) this information can be extracted. The present approach to accomplish this consists of utilizing broad bandwidth pulses for interrogation of the objects and computerized spectral analysis techniques for identification and characterization.

In order to determine if the size, shape, etc. of parameters of an object can be extracted from the data, a series of experiments were undertaken which made use of three very simple flaws. These were a series of flat disks, spheres, and ribbons. The diameter of the disks were 0.030, 0.050, and 0.075 in. in diameter. The spheres were 0.150 in. in diameter and the ribbons were 0.025×0.300 and 0.050×0.300 in. Each of the flaws was embedded in the center of a 2-in. glass cube.

Figures 8.17 through 8.23 show a partial set of data from a pulse with a relatively broad bandwidth reflected from each of three types of flaws. In Figure 8.17, the photo insert on the computer spectral content graph shows the received pulse from a back surface of a glass block without a flaw and is intended as a reference signal. This type of reflector reflects the entire beam from which a normalized spectrum can be obtained. The spectral frequency coordinate (horizontal) is from 1 through 40 MHz. The peak spectral amplitude, (vertical coordinate) was normalized to one on the spectral distribution graph.

Neg 700548-1

FIGURE 8.17. Received Pulse Displayed on a Sampling Scope from the Back Surface of a 1 in. Glass Plate and the Computer Analyzed Power Density Function from 1 Through 40 MHz

Neg 700548-3

FIGURE 8.18. Received Pulse from 0.030 in. Diameter Disc 1 in. Deep in Glass
and the Power Density Function from 1 Through 40 MHz

Neg 700548-2

FIGURE 8.19. Received Pulse from 0.050 in. Diameter Disc 1 in. Deep in Glass
and the Power Density Function from 1 Through 40 MHz

Neg 700548-6

FIGURE 8.20. Received Pulse from 0.075 in. Diameter Sphere 1 in. Deep in Glass
and the Power Density Function from 1 Through 40 MHz

Neg 700548-7

FIGURE 8.21. Received Pulse from 0.150 in. Diameter Sphere 1 in. Deep in Glass and the Power Density Function from 1 Through 40 MHz

Neg 700548-4

FIGURE 8.22. Received Pulse from 0.025 by 0.300 in. Ribbon 1 in. Deep in Glass
and the Power Density Function from 1 Through 40 MHz

Neg 700548-5

FIGURE 8.23. Received Pulse from 0.050 by 0.300 in. Ribbon 1 in. Deep in Glass and the Power Density Function from 1 Through 40 MHz

Figure 8.18 shows the signal from a 0.030-in. diameter disc. Note the shape of the spectral distribution including the relative increase in the high frequency components as compared to Figure 8.18. Figure 8.19 shows the signal from a 0.050-in. diameter disc. Note the relative increase in amplitude in the lower spectral frequency components compared to Figure 8.18. The relative signal amplitude between Figures 8.18, 8.19 and 8.17 are noted as an attenuation factor just beneath the photo insert -- the higher attenuation (Figure 8.17) being the stronger signal.

Figure 8.20 shows the signal from a 0.075 in. diameter sphere. Note the spectral peaks at approximately 3.6 MHz intervals and the overall spectral pattern change compared to the flaws of Figures 8.17 through 8.19. Figure 8.21 shows the signal from a 0.150-in. diameter sphere. Note the difference in the spectral peaks which are now approximately doubled those in Figure 8.20. Also, the lower frequency spectral components of Figure 8.21 have increased in the overall curve shape compared with Figure 8.20.

Figure 8.22 shows the signal from a 0.025×0.300 -in. ribbon. Note the change in spectral pattern shape compared to the disc, (Figures 8.18, 8.19) and the sphere, (Figure 8.20, 8.21). Also, the lower spectral frequency content increases compared to a disc or sphere. Figure 8.23 shows a 0.050×0.300 -in. ribbon. Note the broader spectral peaking bands compared to Figure 8.22 and the increase in relative amplitude.

These and associated data indicate the following:

- Spectral information from broadband-width ultrasonic pulses reflected from flaws appears to be a basis for determining flaw size and geometry.

- Further development and optimization of pulse techniques and understanding of broadband-width pulse-wave propagation behavior is mandatory for practical implementation of the spectral analysis techniques for flaw identification.

THERMAL METHODS

D. R. Green

Research and development on new thermal and infrared techniques is continuing, and further applications of the techniques have been made. These applications include testing FFTF fuel pin simulators and carbon/carbon-composite cones. A full-scale experimental prototype of a thermal-image transducer for testing conical specimens was fabricated for Sandia Laboratories.

Figure 8.24 shows the full-scale, conical thermal transducer. Air pressure is used to press the transducer surface into close contact with the conical test specimens. Testing the area covered by the transducer requires 3 sec, not including the time required to get the cone into position. A bank of ultraviolet lamps and three solenoid-operated cameras that are used to record the thermal image are not shown in Figure 8.24 so that the details of the transducer itself can be seen.

Automatic analysis of data from thermal-image transducers requires rapid return to equilibrium upon terminating the heating pulse. Previous attempts to perform automatic image analysis were made on an air-convection cooled transducer. Because equilibrium was attained too slowly with this type of transducer, it was redesigned to incorporate water cooling. In addition to allowing automatic data analysis, the water cooling allows a test to be repeated sooner, if desired. Excellent images of defects were obtained with the water-cooled transducer. However, the thermally sensitive phosphor used to produce the image darkened after a few tests. We believe

Neg 70022-3

FIGURE 8.24. Full-Scale, Experimental Prototype of a Thermal-Image Transducer for Testing Conical Specimens

that this darkening is due to volatile materials, contained in the binder used with the phosphor, that are vaporized by the heat during a test and condense on the water-cooled, optically-transparent transducer backing. Phosphor applied to the transducers is presently being outgassed at elevated temperatures in a vacuum to reduce the concentration of volatile vapors.

High frequency response is desired in the thermal image transducer so that it can be applied to test bonds under thin coatings. A special low-thermal inertia foil for use in the transducers has been obtained. This foil should improve the frequency response of the thermal transducer so that it exceeds 25 Hz. Methods of mounting the foil and coating it with thermally sensitive phosphor are being developed.

Another application for the thermal transducer method is emittance-independent, noncontacting measurement of surface temperature. The thermal transducer comprises a thin, blackened foil suspended between the specimen surface and a backing as shown in Figure 8.25. Blackening of the foil is not essential, but increases sensitivity. The backing surface is water cooled to provide a constant temperature of 25 °C. We will assume values of surface temperatures and emittances as shown in Figure 8.25 for the purpose of computations to demonstrate the feasibility. In a practical surface temperature transducer, such assumptions would not be necessary, as we shall see later.

FIGURE 8.25. Physical Arrangement of the Thermal Transducer and Specimen of Which the Surface Temperature is to be Measured

If $T_1 = T_3$, all of the heat flow, q , must pass through thermal resistance R_1 (Figure 8.25) to the backing, since the temperature drop across R_2 is zero and thus no heat flows through it. Let us consider the foil temperature that results from various amounts of electrical power injected into the foil, assuming for the moment that the specimen temperature is always somehow adjusted to be equal to the foil temperature. (The reason for this apparently trivial assumption will become clear later.) Then, at equilibrium

$$\frac{\text{Electrical Power Into Foil per Unit Area}}{\text{Radiant Energy Lost per Unit Area From Foil to Cold Backing}} = 1 \quad (1)$$

assuming the foil is large enough and close enough to the backing and to the specimen that edge effects do not occur in the center of the foil (where the foil temperature is monitored). Values of foil temperature that result from some various amounts of injected electrical energy are tabulated in Table 8.1.

TABLE 8.1. Foil Temperatures Resulting from Electrical Heating, q , When the Specimen Temperature and Foil Temperature are Equal

$q, \text{ W/cm}^2$	$T_1, ^\circ\text{F}$
0.100	716
0.115	755
0.135	807
0.152	850
0.175	894

These temperature values are approximate; they were calculated on a radiation slide rule to save time.

Now, consider the foil temperature resulting from electrical power input when the specimen temperature is not equal to the foil temperature. In this case,

Energy Injected Energy Absorbed by the Energy Lost Radi-
Electrically Into + Foil Radiantly from antly Into Back-
Foil per Unit Area Specimen per Unit Area ing per Unit Area
or

$$q + \frac{1}{\frac{1}{\epsilon_1} + \frac{1}{\epsilon_3} - 1} \sigma (T_3^4 - T_1^4) = \frac{1}{\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} - 1} \sigma (T_1^4 - T_2^4). \quad (2)$$

Again, as above, we tabulate the resulting values of foil temperature resulting from electrical heating. However, this time we must assume a specimen temperature and surface emittance as indicated in Tables 8.2 and 8.3.

TABLE 8.2. Foil Temperatures Resulting from Electrical Heating, q , When the Specimen Temperature and Emittance are 824 °F and 0.10, Respectively

$q, \text{ W/cm}^2$	$T_1, \text{ °F}$
0.100	770
0.115	796
0.135	822
0.152	841
0.175	862

TABLE 8.3. Same as Table 8.2, but Specimen Surface Temperature and Emittance are 716 °F and 0.10, Respectively

$q, \text{ W/cm}^2$	$T_1, \text{ °F}$
0.100	716
0.115	739
0.135	767

Figure 8.26 shows a plot of the foil temperatures as a function of electrical heating for the loaded case (the case where specimen temperature is not necessarily equal to foil temperature) and nonloaded case (where specimen temperature is always equal to foil temperature). The shaded areas represent a band of error of about ± 4 °F resulting from the use of a radiation

slide rule in the calculations. More precise computations could easily be carried out, but would not add anything to the discussion here.

FIGURE 8.26. Foil Temperature Versus Electrical Power Injection

As we would expect, the loaded and nonloaded transducer curves in Figure 8.26 cross at the point where the foil temperature and specimen temperature are equal. Hence, if the electrical power input to the foil is swept through a range of values, the corresponding loaded foil temperature will only arrive at an unloaded value when the foil temperature is passing through the sample temperature. The unloaded values of foil temperature are, of course, predetermined by placing the transducer foil near a specimen that is always maintained at a temperature equal to the foil temperature while the electrical input to the foil is swept through its range of values. In this manner, the transducer is calibrated according to its own internal resistance. The point at which the foil passes through the unloaded transducer response curve can either be determined graphically or by an automatic electronic method which uses an analog curve follower or a digital memory.

Specimen emittance has no effect on the temperature measurement, except for sensitivity, since the emittance influences only the slope of the loaded curves and not the crossover points between the unloaded curve and the loaded curves. In practice, the unloaded curve would be best determined experimentally rather than computed, since real materials do not always behave according to the Plank radiation function that was used in the computed example above

The computed curves in Figure 8.26 are given only as evidence of the sensitivity that could be expected. Somewhat lower values of specimen emittance than those used in the computations are possible, but the sensitivity of the transducer should be more than adequate since the foil temperature should be measurable with an accuracy of better than ± 1.0 °F.

NEW TECHNIQUES

Holographic Methods

F. R. Reich

Applications of holography in nondestructive testing have been successfully demonstrated, using both cw (continuous wave) and pulsed ruby laser illumination. The main effort has been directed toward reflection illumination holography and the ability to do classical interferometry with complexly shaped objects not optically smooth or flat. All holograms were made using either a cw helium-neon laser or a pulsed ruby laser with maximum power outputs of 50×10^{-3} W and 0.5 joules, respectively. The ruby laser was operated only in the free lasing mode as the "Q" switched output power was not sufficient for film exposure.

Both real time and double exposure interferometric holograms were used in vibration analysis of a membrane and an audio speaker. Real time interference holograms refers to optical reconstructions where interference occurs between the real object and a reconstructed image stored on a hologram. In double exposure interferometric holography, the interfering images are both stored on the hologram plate. The reconstructed holograms of the speaker and membrane contained fringes produced by motion at the specific area marked by the fringe. The shape and intensity gradient of the node pattern indicate both amplitude and frequency response which may be used to evaluate the material or dimension of the vibrating object. In real time interferometric holography, the dynamic properties of the vibrating object can be studied quickly as the fringe structure is presented in real time.

Double exposure and real time holography was demonstrated to show strain induced by pressure in a vessel and by mechanically loading a sample. Both mechanical and material

properties can be studied under stress. Weld defects in FFTF reactor cladding pin weld samples were located with pressure-induced stress. Areas which appeared to be weakened or annealed by the welding process were also visible. The holographically noted defect region correlated with other NDT analyses previously applied to the cladding samples. By heating a sample, both thermal expansion and conductivity of the sample material can be studied with holography. The expansion, induced by heating, was studied with both real time and double exposure holography. Thermal gradients in a honeycomb constructed panel indicated bonding discontinuities. The thermal gradient was induced by heating and cooling the panel surface between exposures of a double exposure hologram. Fringes from an overall panel warping due to the localized heat source produced a fringe grid on the panel. This enhanced some of the smaller thermal gradients. Bonding discontinuities noted with holography correlated with other thermalgraphic testing methods.

REFERENCES

1. L. M. Brekhovskikh. Waves in Layered Media. Academic Press, New York, 1960. pp. 28-44.
2. F. R. Rollins Jr. Mater. Evaluation, vol 24, p 683. 1966.
3. O. I. Diachok. Ultrasonic Reflectivity At a Liquid-Solid Interface at the Rayleigh Angle, ONR-4. Georgetown University, Washington, D.C., 1969.
4. A. L. VanBuren and M. A. Breazeale. J. Acoust. Soc. Am., vol. 44, no. 4, p. 1014. 1968.
5. C. E. Fitch and R. L. Richardson. Progress in Applied Materials Research edited by E. G. Stanford. J. N. Fearon and W. J. McGounagle, Iliffe Books, London 1967. Vol. 8, p. 79.
6. F. L. Becker, C. E. Fitch and R. L. Richardson. Ultrasonic Reflections and Transmission Factors for Materials Attenuation, BNWL-1283. Battelle-Northwest, Richland, Washington, 1970.

7. F. L. Becker and R. L. Richardson. J. Acoust. Soc. Am., vol. 45, no. 3, p. 793. 1969.
8. F. L. Becker and R. L. Richardson. Research Techniques in Nondestructive Testing, edited by R. S. Sharp. (Accepted for publication by Academic Press, London) Vol. 1, Chapter 4, p. 91.
9. V. M. Merkulova. Soviet Phys.-Acoust., vol. 15, no. 3. March 1970.
10. Unpublished Data. Westinghouse Research Laboratories, Pittsburgh, Pennsylvania, 1970. (Personal Communication with G. Mott)
11. A. Schoch. Ergeb. Exakt. Naturw., vol. 23, p. 127. 1950.
12. L. S. Fountain. J. Acoust. Soc. Am., vol. 42, no. 1, p. 242. 1967.
13. F. L. Becker and R. L. Trantow. Trans. Am. Nucl. Soc., vol. 12, no. 2, p. 540. 1969.
14. I. A. Viktorov. Rayleigh and Lamb Waves. Plenum Press, New York, 1967. p. 5.

9.0 ATR WATER LOOP OPERATION AND MAINTENANCE

R. S. Hope

The Advanced Test Reactor reached full power of 185 MW for Cycle 1 on December 26, 1969 and completed the first half of the cycle on February 8, 1970. Accumulated exposure was 6833 MWd or 36.94 equivalent full power days. The reactor was restarted for Cycle 1B on February 12, 1970 and has accumulated 3700 MWd at 185 MW. Cycle 2 shutdown occurred March 4, 1970. Eighteen of the 20 full power days during Cycle 1B were without a reduction in power.

No changes were made in the ATR 1D-N pressurized water loop during reactor shutdown. The loop is being operated at 2000 psig, 520 °F, 70 gal/min, pH 10.00 ± 0.2 and oxygen concentration at less than 0.05 ppm. The loop contained 168 miniature stainless steel Fermi tensile specimens and 84 Zircaloy corrosion coupons. Total accumulated exposure at a peak fast flux of 1.2×10^{14} nv was 5.9×10^{20} nvt.

DISTRIBUTIONNo. of
CopiesOFFSITE

1	<u>AEC Division of Naval Reactors, HQS</u> R. H. Steele
22	<u>AEC Division of Reactor Development and Technology</u> Director, RDT Asst. Dir for Project Management Chief, Water Projects Branch Chief, Gas Cooled Projects Branch FFTF Project Manager EBR-II Program Manager LMFBR Program Manager Asst. Dir. for Reactor Engineering Asst. Dir. for Program Analysis Asst. Dir. for Engineering Standards Asst. Dir. for Nuclear Safety Asst. Dir. for Plant Engineering Chief, Facilities Branch Chief, Components Branch Chief, Liquid Metal Systems Branch Chief, Water/Gas Systems Branch Asst. Dir. for Reactor Technology Chief, Reactor Physics Branch Chief, Fuels & Materials Branch Chief, Special Technology Branch Chief, Fuel Recycle Branch Chief, Coolant Chemistry Branch
2	<u>AEC Division of Research</u> P. W. McDaniel D. K. Stevens
6	<u>AEC Division of Space Nuclear Systems</u> D. Beard J. F. Griffo C. E. Johnson J. A. Powers F. C. Schwenk E. S. Wilson
5	<u>AEC Division of Technical Information Extension</u>

No. of
Copies

1	<u>AEC Library, Washington</u>
10	<u>AEC RDT Site Representatives</u> M. E. Jackson, ANL D. J. Wille, APDA G. U. Merckel, CE R. L. Morgan, CPAI J. V. Levy, GE, San Jose J. R. Weissenberg, GE-NMPO R. H. Ball, GGA J. E. Hebert, ID D. F. Cope, OR M. Napack, UNC
11	<u>Argonne National Laboratory</u> R. Bane S. Greenberg J. H. Kittel K. Kuczen M. V. Nevitt P. G. Shewmon F. Smith R. C. Vogel LMFBR Program Office A. Amorosi J. R. Humphreys L. R. Kelman
1	<u>Idaho Falls Office</u> C. M. Walter
1	<u>Allis-Chalmers</u> <u>Atomic Energy Division</u> 5401 Westbard Ave. Bethesda, Maryland 20016 L. Phillips
3	<u>Atomic Power Development Associates, Inc. (AEC)</u> E. C. Kovacic A. A. Shoudy Technical Director

No. of
Copies

4	<u>Atomics International (AEC)</u> J. Balif (2) R. W. Dickinson H. Pearlman
5	<u>Babcock & Wilcox Company (AEC)</u> C. Baroch S. P. Grant C. Johnson P. B. Probert L. Weissert
1	<u>Baldwin-Lima-Hamilton Corporation</u> <u>Eddystone, Pennsylvania 19013</u> P. S. Otten
3	<u>Battelle Memorial Institute (AEC)</u> D. Keller S. Paprocki E. M. Simons
2	<u>Bettis Atomic Power Laboratory (AEC)</u> E. J. Kreh Laboratory Library
3	<u>Brookhaven National Laboratory (AEC)</u> O. E. Dwyer D. H. Gurinsky C. Klamut
1	<u>Bureau of Mines, Albany (INT)</u> H. Kato
1	<u>Chalk River Nuclear Laboratory</u> E. C. W. Perryman
2	<u>Combustion Engineering, Inc. (AEC)</u> W. P. Chernock S. Christopher
2	<u>du Pont Company, Aiken (AEC)</u> <u>Savannah River Laboratory</u> Technical Information Service

No. of
Copies

1	<u>General Electric Company</u> <u>Nuclear Systems Programs</u> Cincinnati, Ohio 45215 E. E. Hoffman
2	<u>General Electric Company, San Jose (AEC)</u> R. Duncan H. Klepfer
5	<u>General Electric Company, Sunnyvale</u> K. P. Cohen F. Comprelli R. Skavdahl C. Spalaris E. L. Zebroski
4	<u>Gulf General Atomic Incorporated</u> D. B. Coburn A. J. Goodjohn S. Jaye J. F. Watson
1	<u>Idaho Nuclear Corporation</u> W. C. Francis
1	<u>IIT Research Institute</u> D. J. McPherson
4	<u>Iowa State University</u> O. N. Carlson W. L. Larsen M. Smutz F. H. Spedding
1	<u>Jet Propulsion Laboratory (NASA)</u> J. Davis
1	<u>Knolls Atomic Power Laboratory (AEC)</u> Document Library
1	<u>Lawrence Radiation Laboratory, Livermore (AEC)</u> J. S. Kane

No. of
Copies

7	<u>Los Alamos Scientific Laboratory (AEC)</u> R. D. Baker D. B. Hall J. A. Leary R. Perkins R. Peterson G. Waterbury L. E. Whinery
1	<u>Mound Laboratory (AEC)</u> R. Grove
1	<u>MSA Research Corporation</u> <u>Evans City, Pa.</u> R. C. Andrews
2	<u>NASA Lewis Research Center</u> J. J. Lombardo C. M. Scheuermann
1	<u>National Aeronautics and Space Administration</u> <u>Space Administration</u> <u>Lewis Flight Propulsion</u> <u>Laboratory, Cleveland, Ohio 44135</u> C. A. Barrett
3	<u>Naval Research Laboratory</u> J. R. Hawthorne C. Serpan L. E. Steele
1	<u>North Carolina University</u> J. Beeler
2	<u>Nuclear Materials and Equipment Corporation (AEC)</u> C. Caldwell K. Puechl
15	<u>Oak Ridge National Laboratory (AEC)</u> G. M. Adamson J. E. Cunningham J. H. DeVan W. P. Eatherly D. Gardiner W. O. Harms W. R. Martin R. E. MacPherson H. E. McCoy C. J. McHargue P. Patriarca O. Sisman M. S. Wechsler J. R. Weir J. White

No. of
Copies

2	<u>Ohio State University</u> M. G. Fontana R. W. Staehle
1	<u>Rensselaer Polytechnic Institute</u> C. B. Willingham, Jr.
2	<u>Sandia Corporation, Albuquerque (AEC)</u> J. Jacobs R. P. Stromberg
1	<u>Stanford Research Institute</u> <u>Menlo Park, California</u> D. Weinstein
2	<u>United Nuclear Corporation (AEC)</u> K. Goldmann A. Strasser
1	<u>USAEC Scientific Representative</u> <u>Chalk River, Ontario, Canada</u> R. W. Ramsey, Jr.
1	<u>Westinghouse Astronuclear Laboratory</u> <u>Materials Department</u> Pittsburgh, Pa. Leonard France
5	<u>Westinghouse Electric Corporation (AEC)</u> R. Miller T. Stern J. H. Wright (3)
6	<u>Westinghouse Electric Corporation</u> <u>Atomic Power Division</u> Advanced Reactor System Waltz Mill Site P. O. Box 158 Madison, Pa. 15663 E. C. Bishop M. Bleiberg A. Boltax J. C. R. Kelly P. J. Levine P. Murray

No. of
Copies

2 Westinghouse Electric Corporation (WAL) (AEC)

R. T. Begley
D. C. Goldberg

ONSITE-HANFORD

1 AEC Chicago Patent Group

R. K. Sharp (Richland)

2 AEC Richland Operations Office

C. L. Robinson (2)

3 Douglas United Nuclear Inc.

P. A. Carlson
D. W. Peacock
L. H. Rice

2 RDT Assistant Director for Pacific
Northwest Programs

71 Battelle-Northwest

F. W. Albaugh
H. J. Anderson
E. R. Astley
D. E. Baker
J. M. Batch
J. L. Bates
A. L. Bement
C. A. Bennett
T. K. Bierlein
S. H. Bush
J. J. Cadwell
T. D. Chikalla
J. A. Christensen
T. T. Claudson
P. D. Cohn
R. E. Dahl
J. L. Daniel
F. G. Dawson
R. L. Dillon
D. G. Doran
K. Drumheller
E. A. Eschbach
E. A. Evans
J. J. Fuquay

Battelle-Northwest (contd)

R. L. Gibby
S. Goldsmith
J. E. Hanson
L. A. Hartcorn
H. Harty
B. R. Hayward
E. D. Jenson
S. S. Jones
D. C. Kaulitz
R. S. Kemper
R. L. Knecht
H. A. Kornberg
G. A. Last
R. D. Leggett
D. C. Lehfeldt
C. E. McNeilly
J. E. Minor
R. P. Nelson
R. E. Nightingale
D. P. O'Keefe
L. T. Pederson
W. D. Richmond
W. E. Roake
W. F. Sheely (2)
R. C. Smith
J. C. Spanner
R. W. Stewart
H. A. Taylor
G. L. Tingey
J. C. Tverberg
E. E. Voiland
R. E. Westerman
R. G. Wheeler
E. T. Weber
D. C. Worlton
H. H. Yoshikawa
Patent Section (2)
Technical Information File (5)
Technical Publications (3)