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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

By

A. L. Bement

G. J. Dau

E. A. Evans

R. E. Nightingale

November 1968

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SUMMARY

NUCLEAR GRAPHITE

Prestressing of graphite, in either tension or compression, up to ultimate strength causes a reduction in elastic moduli. Stress-induced changes in moduli begin to anneal out at temperatures as low as 200 °C. This change is reflected in the observed annealing effect on fractional expansion of precompressed samples.

ELECTRON MICROSCOPY STUDIES OF GAS - GRAPHITE REACTIONS

Spherical, shell-like arrangements of graphite crystallites, appearing as rosette structures, have been found in binder regions of TSX and AGOT graphite. Rosette structures occur singly, as fused duplex arrangements, and in various kinds of

clusters of three or more units. Electron diffraction data confirm the spherical symmetry of rosette structures and indicate that they are surrounded by smaller less orderly crystallites than those within the rosettes. Oxidation experiments indicate that rosettes are quite resistant to attack by thermal reaction but are easily consumed by microwave-excited oxygen. Various stages in the development of cone structures and impurity networks have been examined in TSX and AGOT graphite oxidized by microwave-excited oxygen. After 5% burnup, cone heights range from 100 to 5000 Å, and the size distribution does not appear to change with burnup level up to at least 30%.

FUEL DEVELOPMENT FOR GAS-COOLED REACTOR -- HTGR GRAPHITE STUDIES

Water vapor is a common contaminant in otherwise inert atmospheres of graphite-moderated reactors. To investigate the reaction, ampoules of pyrex and quartz were exposed to gamma radiation in a cobalt-60 facility. The complete removal of unknown carbonaceous from the graphite ampoules was not possible. Gas analysis of eight quartz ampoules is in progress.

Previously, it had been assumed

that the major source of impurity in Peach Bottom HTGR was the ingress of steam from the heat exchangers, but other possible sources have been considered also including oil leakage, proton diffusion from steam corrosion, and water ingress followed by preferential oxygen cleanup. Additional information is needed before a computer assessment can be made.

NUCLEAR CERAMICS

Several experimental techniques are being developed or considered for measurement of the properties of molten uranium-plutonium oxides.

The change in volume on melting and liquid expansion coefficient is measured at temperatures to 3000 °C by use of radiography to observe a change in liquid column height as a function of temperature.

Two methods that have been developed for measurement of shear viscosity are the falling sphere method using Stokes' Law and the oscillating torsional pendulum technique. Conventional-pendant and sessile-drop techniques for measurement of surface tension will be investigated.

Studies of sample holder design for measurement of thermal diffusivity by the laser-pulse method have been initiated, and plans are underway to determine liquid compressibility by measurements of ultrasonic velocity.

The SNOUT facilities in the Hanford K Reactor are to be used for (U,Pu)O₂ creep experiments. The initial experiments will be short-time bend-test irradiations of rectangular beams of fuel. The tests will be run in the temperature range of 800 to 1200 °C.

Final modifications to the high-temperature creep apparatus have been completed. The plutonium glove-box in which the apparatus was installed was subsequently leak-checked and placed on plutonium control. Several runs have been made on UO₂ to identify problems peculiar to glove-box

operation. Samples of UO₂-25 wt% PuO₂ have been prepared, and initial creep measurements have begun.

A high-temperature sample holder was fabricated to be used with the phase-comparison technique of determining the velocity sound in solids, and work has begun on finding a good high-temperature bonding material.

Consideration is now being given to the design of a cell for performing ultrasonic velocity measurements on molten oxides.

The thermal diffusivity, α , of sintered mixed oxide was measured from 100 to 1200 °C as a function of O/M from 1.93 to 2.00. Thermal conductivity, λ , was calculated from the thermal-diffusivity data. The general expressions for α and λ in terms of the amount of missing oxygen, x , (moles oxygen atoms/mole mixed oxide) are, according to this study

$$1/\alpha = 0.110 T + 1.08 \times 10^3 (x) + 4 \quad (\text{sec/cm}^2)$$

$$1/\lambda = 0.0312 T + 3.55 \times 10^2 (x) + 1.5 \quad (\text{cm } ^\circ\text{K/W}),$$

with temperature in units of °K. The defect thermal resistance resulting from oxygen removal from the mixed-oxide lattice was found to be in agreement with values predicted from Klemen's phenomenological models for phonon scattering by point defects.

The electrical resistivity of high-energy-rate-impacted uranium mononitride has been measured from 25 to 2550 °C. Changes in resistivity during thermal cycles are described.

The melting points of UO_2 were measured as a function of neutron irradiation to approximately 3×10^{19} f/cm³. The melting points of the

irradiated UO_2 (6 exposures) were the same as the melting point of non-irradiated UO_2 .

FAST REACTOR NITRIDES RESEARCH

Melting studies in the UN-PuN system at nitrogen pressures from 1 to 100 atm have been reported previously. The most recent work has included measurements on samples of sintered mixed-nitride fuel compositions.

Measurements of the nitrogen decomposition pressure for these fuel materials, as well as on several specially prepared compositions at

various U/Pu ratios, have been extended to pressures between 0.01 and 1 atm. A cold-wall tungsten-filament furnace was used which was similar to the high pressure furnace previously described. The system was capable of reaching high vacuum, with an aneroid-type absolute pressure gauge used to measure nitrogen pressures down to 2mm Hg.

NONDESTRUCTIVE TESTING

EDDY CURRENT METHODS

The published literature of eddy-current nondestructive testing techniques uses several systems of units and different ways of presenting the same data. As an aid in clarifying one confusing situation, three different ways of treating the normalized impedance diagrams for a solenoid containing an electrically conducting bar are shown and discussed. The relationships between different units are shown. The significance of the ratio a/δ (where a = radius of bar and δ = plane wave skin depth) in these impedance diagrams is explained.

THERMAL METHODS

Work was done to adapt a 30 W carbon dioxide laser for use with the transient test system. A suitable shutter and optical system for creating short output pulses and a large collimated beam size were investigated. A draft of a journal article describing the system was completed, and work continues on the interim progress report.

Thermal transducer methods have a number of potential practical applications and are completely independent of surface emittance (emissivity) of the test specimen. Some of the most important near-future

nuclear applications will involve cylindrical test specimens, and will require scanning with high-frequency transducers. Attempts this quarter to produce a high-frequency transducer were partially successful, with a frequency response of about 20 Hz obtained. Present techniques for detecting the transducer temperature, however, are not completely satisfactory, and other possible techniques are being investigated.

ULTRASONIC METHODS

Ultrasonic critical angle method measurements were taken at central positions on all irradiated and unirradiated specimens. Ultrasonic longitudinal velocity measurements

taken on samples of 304 SS irradiated from 3×10^{20} to 1.1×10^{22} nvt > 1 MeV at 900 to 1000 °F indicate an increasing velocity with fluence.

There are several problems to consider when one fabricates a high-resolution lithium sulfate transducer. These problems include the ability to monitor the thickness, the sealing of the crystal from water, and the application of the thin electrodes.

The "Ultrasonic Wave Model with Attenuation" has been shown to predict the distortion of Ultrasonic pulses in aluminum with good accuracy. Accuracy through highly attenuating material such as stainless steel is considerably reduced.

BASIC SWELLING STUDIES

FUELS DEVELOPMENT SECTION

Fabrication of the experimental fuel elements for the Canadian Whiteshell Reactor is complete. These elements will allow the evaluation of twelve variables with three different uranium enrichments. The fuel cores are metallic and the cladding is Ozhenite 0.5. Some of the elements are coextruded with a small-diameter coaxial void, the others use metal pellets--some with dished ends and the remainder with a coaxial hole.

Irradiation of unalloyed uranium and dilute uranium base alloy specimens was conducted at 5000 psi and

480 to 550 °C to burnups of 0.27 to 0.99 at.%. Radiometallurgical observations reveal:

- A hydrostatic pressure of 5000 psi reduces swelling in high purity uranium by a factor of 30 to 40.
- Dilute alloy additions combined with this pressure reduce swelling to the theoretical minimum.

Irradiation at 1000 psi and ~ 625 °C to burnups in the range 0.3 to 0.6 at.% revealed no evidence of "break-away" swelling. Pressure and alloying additions are additive in reducing swelling almost to the theoretical minimum.

Thorium-uranium alloys were in excellent condition after being irradiated at 780 to 830 °C and 50 psi to burnups as high as 1 at.%. Ura-

nium and uranium alloys in the same capsule exhibited R values $\left(\frac{\% \Delta V/V}{\text{at.}\% \text{ BU}}\right)$ of 50 to 270.

ATR GAS LOOP OPERATION AND MAINTENANCE

Problems associated with the Hastelloy-X material used in the Gas Loop regenerative heat exchangers were resolved. Discussions with consultants on the requirements of Section III of the ASME Code required replacement of some of the forgings because the direction of high stresses coincided with the

direction of the lowest tensile properties.

The analytical equipment for the Gas Loop is in the final check out stage. PNL has had full responsibility for the inspection to determine compliance with the specifications. Shipment of the unit to the ATR site will be arranged during October.

FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS

The PNL-ANL cooperative dosimetry experiment was prepared for irradiation in the Experimental Breeder Reactor-II (EBR-II) during October-November 1968. A compact (0.328 in. OD x 1 in. long) activation-monitor container capable of holding 15 to 20 different materials was designed and constructed for the experiment.

The IRL experiment to simulate a pressure-vessel spectrum will include an effort to determine the change in neutron spectrum during penetration through various thicknesses of water. The schedule for the test now calls for long-term irradiations to start in the latter part of September.

To assure satisfactory service of cladding and structural materials

in the FTR, some method is needed for estimating the damage expected in FTR components during their service life. Such a method is being studied, and the analysis of damage in pressure-vessel steels has established its feasibility. Preliminary results also indicate that the low-energy neutrons are more important in their contribution to damage than predicted by current damage models.

The technique for simulating the annealing of displacement spikes has been refined and applied to three 20 keV spikes in α -iron. Several thousand time-steps were used (corresponding to $\sim 10^{-6}$ sec real time), during which 80% of the defects were annihilated and the number of large vacancy clusters were decreased.

Also, the surviving interstitials formed small clusters.

The results of point-defect calculations in metals depend on the type of defect considered. For both fcc copper and bcc α -iron, divacancies separated by 1st and 2nd neighbor distances were calculated to be the most stable independent of potential type and range. For interstitial defects, the $\langle 100 \rangle$ split interstitial was calculated to be the most stable configuration in copper for all potentials used. In bcc α -iron, the modified Morse potentials of this study produced the result that either the $\langle 111 \rangle$ split or the activated crowdion could be the stable interstitial configuration. This result is dependent on the

potential used since earlier calculations identified the $\langle 100 \rangle$ split as the stable interstitial.

GRAINS, a newly completed pseudodynamic program used in fcc nickel-atom arrangements at simulated grain boundaries, calculated formation and migration energies for vacancies, interstitials, and carbon impurity atoms near grain boundaries. Comparable calculations for the perfect lattice disclose significant differences for these quantities between the values in the crystal and at grain boundaries. Therefore, the rates of production and annealing of defects during and after irradiation in the vicinity of the grain boundary are expected to differ from the rates in the perfect crystal.

IRRADIATION DAMAGE TO REACTOR METALS

REACTOR METALS RESEARCH

Irradiation Facilities Operation

A total of 36 tensile, creep, and corrosion specimens was discharged from the ETR at the conclusion of Cycle 96.

The ATR Pressurized Hot Water Loop is scheduled for start-up on January 6, 1969. Specimens of 304 and 316 SS for the initial loading are being machined and the support hardware designs are complete.

Microstructural Modifications to Austenitic Stainless Steels and Nickel-Base Alloys

Thermomechanical "stress-aging" treatments lead to improved strength with high ductility retention at 1200 °F in 304 SS. Moreover, to achieve a given high strength level requires less "work", in terms of reduction in thickness, than is required in ordinary warm-working. Both of these facts might signify

decoration of dislocations by precipitates that, if true, could indicate reduced irradiation-induced precipitation, leading to improved irradiation-stability.

In-Reactor Measurements of Mechanical Properties

An analysis of creep-rupture data for Type 304 SS showed that rupture lives during in-reactor and post-irradiation testing are essentially the same for comparable conditions of stress and temperature. The effect of neutron irradiation, as reflected in both the in-reactor and postirradiation tests, is to reduce the rupture life or the rupture strength as compared to the unirradiated control data.

An analysis of in-reactor and unirradiated control-creep data for Zr-2.5 Nb using the yielding creep model showed that there is a greater internal stress in Zr-2.5 Nb than in Zircaloy-2, which is consistent with the greater strength of Zr-2.5 Nb.

Grain Boundary Stabilization

Three-mil-thick nickel foils of very small grain size and containing inert gas have been produced by sputtering techniques. Annealing at 450 °C for 30 hr did not appreciably increase the apparent grain size (as measured by X-ray line broadening). Electron microscopy indicates a highly cold-worked type of cell structure with only slight mis-orientation between the cells or grains.

Irradiation Damage to Heavy Section Vessel Steels

The plane strain fracture toughness, K_{Ic} , is determined for ASTM A533B pressure vessel steel between -160 and -80 °C using various size Double Cantilever Beam (DCB) fracture specimens. Initial validity criteria for fracture testing low-yield strength steels using the DCB specimen are established, but additional studies of the DCB specimen are indicated. Initial designs of fracture specimen irradiation capsules have been completed.

Fracture Studies

Calculations of the orientation factors for the two twin systems, $\{10\bar{1}2\} \langle 10\bar{1}1 \rangle$ and $\{11\bar{2}1\} \langle 11\bar{2}\bar{6} \rangle$, and the slip system, $\{10\bar{1}0\} \langle \bar{1}2\bar{1}0 \rangle$, in the vicinity of a crack tip in α -zirconium have been completed. Based on rolling textures in zirconium plate, the results indicate that more deformation should be expected from the two twin systems for cracking perpendicular to the rolling direction than parallel to the rolling direction. This is in agreement with the observed anisotropy in the fracture toughness, K_{Ic} , of rolled Zircaloy-2 plate.

Damage Mechanisms

Evidence is presented showing a change in activation volume in iron at 295 °K after irradiation. New results indicate a Cottrell

atmosphere-drag influences the activation volume in this temperature range, and that the changes produced by irradiation are a consequence of impurity-defect interactions that remove the impurities from solid solution. Straining the materials after irradiation destroys the impurity-defect complex, releases the impurity, and the Cottrell atmosphere returns.

Oversaturation or softening effects are reported in Ferrovac E and Battelle zone-refined iron irradiated to fast fluences in excess of $8 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$). Several mechanisms for this effect are considered, and it is concluded that a dynamic rearrangement of defect aggregates into larger aggregates with an attendant decrease in the number of point defects surviving the damage event is the most likely explanation of oversaturation.

High Pressure Studies

A fission fragment-induced phase transition in ZrO_2 has been found to be accelerated by the application of pressure during the irradiation. Zirconia irradiated to 1.1×10^{15} fissions/ cm^3 under 24 kbars of pressure showed approximately a 30% conversion while powder at ~ 0.001 kbar showed no detectable transition.

In-Reactor Corrosion of Zirconium Alloys

Hydrogen absorption in irradiated Zircaloy-2, lots AT, CT, and HT, is reported and supplements data published in a previous report. Hydro-

gen pickup was relatively small in specimens exposed in the as-etched condition or with thin prefilms. Thick prefilm oxides caused large increases in hydrogen absorption, but the amount differed from lot to lot in agreement with previous G-7 loop results.

Evaluation of Zirconium-2.5 wt% Niobium Pressure Tubing

Creep testing of heat-treated zirconium-2.5 wt% Nb tubing has shown that, at 400°C , the addition of a 15% cold reduction between the water quench and aging does not provide added creep strength nor extend rupture time.

FAST REACTOR SUPPORTING STUDIES

Swelling Studies on Structural Materials

A study of the effects of fast-reactor irradiation on volume changes of austenitic stainless steels has been initiated. Preliminary results suggest that as much as 100% swelling may be found at fluences in excess of $4 \times 10^{23} \text{ n/cm}^2$.

Transmission Electron Microscopy of Irradiated Austenitic Stainless Steel

The electron micrographs of Type 304L SS samples cut from an EBR-II control rod thimble are being analyzed. The specimens have been subjected to fast fluences ranging from 3×10^{20} to $5 \times 10^{22} \text{ n/cm}^2$.

(>0.1 MeV) at irradiation temperatures from 700 to 870 °F (371 to 466 °C). Cavity distributions of Type 304L SS irradiated to 5×10^{22} n/cm² at 790 °F (421 °C) have been determined. These distributions appear to be dependent on the thickness of the foil examined.

EBR-II Irradiations - Tensile Testing

Tensile tests have been conducted on AISI Type 316 SS specimens irradiated in the EBR-II at about 900 °F to a total fluence of 0.8×10^{22} n/cm². Increased yield strength due to irradiation was observed at all test temperatures (RT to 1400 °F). However, the yield strength values at the lower temperatures were much less than those previously observed for 304 SS specimens irradiated in the EBR-II. Tensile specimens and uniaxial and biaxial stress-rupture specimens are being aged in sodium at temperatures from 800 to 1400 °F as part of the aging studies program. Aging at these temperatures also provides controls for the irradiation experiment.

Uniaxial Creep - Rupture Studies

Creep-rupture testing of as-fabricated 316 specimens is continuing. Tests on 304 specimens irradiated in EBR-II and ETR at fast fluences of 4×10^{21} n/cm² ($E > 0.1$ MeV) and 8×10^{20} n/cm² ($E > 1$ MeV) were performed at 1000 °F. Specimens irradiated to a fast fluence of 1.1×10^{22}

($E > 0.1$ MeV) have been creep tested at 1250, 1300, 1350, and 1400 °F to provide data for determining the temperature dependence of the creep rate.

Biaxial Creep - Rupture Studies

The disassembly of biaxial stress-rupture specimen irradiation sub-capsules in an in-cell glove-box is being conducted at Idaho Nuclear Corporation. The effort is currently being directed toward improving the quality of the inert atmosphere of the glove-box. Initiation of biaxial stress-rupture tests on irradiated materials is expected by October 15, 1968. The fuel-clad test facility for testing these specimens is virtually complete.

Efforts directed toward insertion of a fuel cladding irradiation experiment in EBR-II is progressing on schedule for mid-December 1968.

Creep Test Facilities Development

The construction of the facility to test irradiated and unirradiated creep-rupture specimens of candidate structural and cladding materials for the FFTF is complete except for the controls to the heating and ventilation equipment. Eight test frames from the CANEL equipment pool have been moved into place and are being activated. Twelve creep tests performed on four temporarily installed units were completed and represent 2820 actual test hours. The support section of the liquid metal loop has undergone a series of

repairs and short-term tests. The Prototype Hot-Cell was given a final radiation leak test and was found satisfactory.

LMFBR Fuel Cladding Information Center

An information center has been established at Battelle-Northwest to collect and store all information pertinent to the analysis of fuel cladding behavior for LMFBR fuel pin

irradiations. The initial phase of the program has been designed and data are being stored. Selection of appropriate statistical analysis techniques are being made on the basis of several demonstration cases with data from REM (Radiation on Materials Program). The REM system now contains five master files, one for each alloy type. Plotting routines have been written and the data retrieval can now be displayed in graphic form.

NUCLEAR GRAPHITE
R. E. Nightingale

ELECTRON MICROSCOPY STUDIES OF GAS -
GRAPHITE REACTIONS

S. S. Jones

This task involves research on the microstructure of polycrystalline graphite and the structural changes occurring because of gas-graphite reactions. Graphite oxidation is being considered with respect to:

- Crystallite structure and binder-filler relationships
- Impurities
- Radiation-produced defects
- Nature of the oxidizing species
- Temperature
- Ionization and electronic excitation of the gas.

The objectives are to collect data concerning microstructural changes caused by gas-graphite reactions related to the nuclear environment and to develop models for predicting reaction rates and the nature of the associated structural changes.

Spherical shell-like arrangements of graphite crystallites, appearing as rosette structures, have been found in the binder regions of TSX and AGOT graphite. These structures are believed to be caused by carbon blacks--originally present in the coal-tar-pitch binder--that have been graphitized in the process of graphite manufacture. Rosette structures have been found to occur singly, as fused duplex arrangements, and in various kinds of clusters of three or more units. The appearance of these structures occurring in a

binder region of TSX graphite is shown in Figure 3.1. Individual rosetts are roughly spherical in shape and occur with diameters between 1000 and 5000 Å. Sometimes, in the preparation of specimens for microscopic examination, rosettes are brushed from their original position in the graphite binder leaving vacant cavities where they were initially located. Such an area in AGOT graphite is shown in Figure 3.2.

The crystallites in a rosette are arranged in shell-like configurations with the c-axis directed radially to the center of the rosette. Crystallites in the outer shells fit together rather smoothly, but space limitations near the rosette centers appear to prevent inner crystallites from conforming fully with the outer array. It appears that graphitization occurs first in the outer shell, with the process resulting in voids being created in the center of the rosettes. The details of void and crystallite arrangement for a typical rosette are shown in Figure 3.3.

While electron diffraction patterns of individual graphite crystallites vary greatly in orientation to the electron beam, the diffraction patterns of rosette structures do not change in general characteristics with the angle of the incident beam. A typical rosette electron diffraction pattern is shown in Figure 3.4. Such electron diffraction characteristics provide further evidence of the spherical symmetry

FIGURE 3.1. Rosette Structures in a Binder Region of TSX Graphite.

64,000X

FIGURE 3.2. Vacant Cavities in a Binder Region of AGOT Graphite Where Rosettes Were Once Located.

FIGURE 3.3. Rosette Structure Showing Details of Crystallite Arrangement and Internal Voids.

180,800X

FIGURE 3.4. *Typical Rosette Electron Diffraction Pattern.*

of rosette structures. Selected-area electron diffraction patterns of areas surrounding rosettes indicate that the crystallites are smaller and arranged in a less orderly manner than those within the rosettes.

Graphite binder regions containing rosettes are affected differently by thermal and microwave-induced reactions with oxygen. Thermal oxidation attack, occurring primarily at the edges of graphite basal planes, removes the small disordered crystallites around rosette structures, thus, leaving the rosettes isolated from most of the surrounding graphite (Figure 3.5). The resistance of rosettes to thermal oxidation indicates that crystallites in the outer shells of these structures are joined so that (1) layer edges are not appreciably exposed, and (2) the shells are sufficiently gas-tight to prevent oxidation of exposed layer edges within the rosettes. The more aggressive species produced in microwave-induced oxidation reactions readily attack all graphite crystal faces so that rosettes are removed along with other graphite structures as they become exposed to the oxidizing agent. This type of experimental result is shown in Figure 3.6.

Cone-shaped structures, described in earlier reports as saw teeth, are produced in TSX and AGOT graphites by oxidation in the glow region of microwave-excited oxygen. The cone axes are perpendicular to graphite basal planes; impurities, which appear to affect cone formation,

are found in the cone tips. Some details in the growth process of these cones and the associated impurity movements have recently been studied at both low and high graphite burnup.

The initial stages of cone formation can be observed at less than 1/2% burnup. While the location of impurities are believed to determine the cone tips, it has not been possible to predict where cones will appear because impurities are not visible until cone formation has started. Repeated examination of the same specimen areas during cone development has shown that cone concentration, as well as impurity concentration, is highly variable from one area to another. In some cases, great numbers of cones and impurities become visible within a small area, which suggests that the impurities may have been in solution. In other cases, cones and impurities are first seen in linear formations; therefore, the impurities may have been deposited at grain boundaries during the manufacturing process. Cones and impurities become visible initially when a cone height of about 100 Å is attained. One example of cone development in a single area of an AGOT graphite specimen is shown in three successive stages in Figures 3.7, 3.8, and 3.9. In Figure 3.7, the specimen has been oxidized to about 1% weight loss to reveal an early stage of cone growth. The same area is shown in Figures 3.8 and 3.9 after each of two additional oxidation treatments with graphite burnup of

64,000X

FIGURE 3.5. Thermal Oxidation Attach Upon a Binder Region in AGOT Graphite Showing Almost Complete Removal of All Graphite with the Exception of Rosette Structures.

64,000X

FIGURE 3.6. Attack of Microwave-Excited Oxygen upon a Binder Region in AGOT Graphite Showing Nonselective Removal of All Graphite Including Rosette Structures.

64,000X

FIGURE 3.7. AGOT Graphite Oxidized to 1% Weight Loss by Microwave-Excited Oxygen Showing an Early Stage in Cone Development and the Appearance of Impurities at the Cone Tips.

64,000X

FIGURE 3.8. AGOT Graphite Oxidized by Microwave-Excited Oxygen (same area as in Figure 3.7 but after 0.1% further weight loss) Showing Further Development of Cone and Impurity Structures.

64,000X

FIGURE 3.9. AGOT Graphite Oxidized by Microwave-Excited Oxygen (same area as in Figure 3.7 but after 0.2% further weight loss) Showing Later Stage in Growth of Cone and Impurity Structures.

0.1% per treatment. The three figures illustrate how cones form and grow to various heights and, finally, are removed by the oxidation process except for the impurity residue at the cone tip. Depending upon the initial distribution of cones and impurities, the impurity residue will take the form of clusters, chains, and/or networks. Such forms give rise to the extended lace-like structures that have been reported earlier.

While many small cones are removed in the oxidation process, some cones grow to heights of about 5000 Å when 5% burnup has occurred. At this burnup level, there is a wide distribution in cone sizes including cones which are just large enough to be observed. This distribution does not appear to change significantly with burnup level to at least 30%. The principal change at high burnup is the increase in lace-network impurity structures, which is proportional to degree of burnup and graphite purity.

Impurities affect not only the growth of cones during graphite oxidation but probably cause formation of various kinds of imperfect cone-like structures when the impurity level is high. However, relatively perfect cones are formed when the general level of impurities is low. The angular span of these cones, measured with the axes of the cones perpendicular to the electron microscope beam, has been found to vary between 45 and 120 degrees. It is believed

that these angles may be related to the catalytic influence of various impurities at the cone tips involved in the mechanism of cone formation.

COMPATIBILITY STUDIES OF SODIUM WITH GRAPHITE

D. E. Baker

The compatibility of liquid sodium with various types of irradiated and unirradiated graphite was investigated. Conclusions from this study were:

- Irradiated graphite expands more than unirradiated graphite when exposed to liquid sodium at 1000 °F (538 °C).
- Graphite irradiation temperature is a major factor in sodium-graphite compatibility.
- The degree of graphite crystallinity influences the compatibility with sodium.

The purpose of the sodium-graphite study was to identify gross effects of several variables not previously investigated, such as irradiation temperature, fluence, annealing, and stored energy.

Screening tests to determine the compatibility of various graphites with sodium were conducted by encapsulating graphite samples and sodium in stainless steel cans, and heating them at 1000 °F (538 °C) for 100 hr. The capsule welding and opening operations were performed in a helium-filled glove-box where oxygen and water-vapor impurities were less than 20 ppm. After heating, the capsules were

cut open in the glove-box and excess sodium was removed from the surface of the samples before length measurements were made with a micrometer. The sodium-graphite samples had a violet coloration that disappeared after 2 to 8 hr. Colored lamellar compounds of graphite with the alkali metals of sodium, potassium, rubidium, and cesium have been reported in the literature.

The interaction of unirradiated types TSX and TSGBF nuclear graphite with sodium caused an expansion in the samples ranging from 0.44 to 1.05%, the largest expansion occurring in transverse samples. Pyrolytic graphite samples delaminated and cracked into several pieces. An isotropic graphite sample graphitized at 2825 °C also cracked from exposure to molten sodium. The coefficient of thermal expansion (CTE) of type CSF graphite decreased 20% after exposure to sodium. The CTE was measured before the sample was heated with sodium and again after sodium had been removed from the sample by distillation.

Five unirradiated graphite samples containing various amounts of carbon-black were heated with sodium for 100 hr at 1000 °F (538 °C). The samples, graphitized at 3000 °C, were identical except for the carbon-black content, which was 0, 20, 25, 33, and 50%. The only sample not cracked after exposure to sodium contained 25% carbon-black, and this sample expanded 1.4%. Graphite Type N-10-28, which is a Texas-coke graphite containing 30% carbon-

black and graphitized at 2450 °C, was also severely cracked and eroded. Although the results of this test were not conclusive, graphite containing carbon-black is expected to be less stable in the presence of contact with sodium.

The growth of unirradiated CSF graphite in contact with sodium was observed at several temperatures. A cylindrical sample with a central reservoir for sodium was placed vertically in a thermal expansion apparatus, and length changes were observed as a function of time. At 400 °C, graphite expansion was essentially completed after 1 1/2 hr, with no additional length changes occurring during the following 3 hr. The times required to complete the expansion at 350 and 300 °C were 4 hr and 7 hr, respectively. At 250 °C, expansion was continuing after 12 hr.

Transverse samples of unirradiated CSF, TSX, and CSGBF were heated to 1000 °F (538 °C) in contact with sodium in stainless-steel capsules, and sample lengths were measured after 2, 4, 8, and 16 hr. Length changes were similar (0.85 to 1.12%) for the four exposure periods and are in the same range as samples heated for 100 hr.

Graphite samples were irradiated at several temperatures and fluences and then heated in contact with sodium at 1000 °F (538 °C for 100 hr). Test results are given in Table 3.1. A comparison of the data indicates that graphite expansion from interaction with sodium is reduced at higher graphite irradiation temperatures, while graphite expansion

TABLE 3.1. Graphite Expansion from Interaction with Sodium at 1000 °F (537 °C)

Graphite Type, $\perp$	Irradiation Temperature, °C	Exposure, nvt (E > 0.18 MeV)	Length Change, %
CSF(a)	30	1×10^{19}	+0.97
CSF(a)	30	1.2×10^{20}	+1.02
CSF(b)	550	1.5×10^{20}	+0.73
CSF(b)	550	4.5×10^{20}	+0.67
TSX	600	1.4×10^{21}	+1.17
NC8	600	1.4×10^{21}	+1.07
CSF	600	1.4×10^{21}	+1.07
TSX	550	1.6×10^{21}	+1.74
CSF(b)	550	1.7×10^{21}	+0.95
TSGBF	550	1.7×10^{21}	+1.17
TSGBF	700	1.8×10^{21}	+2.40
CSF(b)	550	2.2×10^{21}	+0.98
CSGBF	550	2.3×10^{21}	+0.90
TSX	600	2.45×10^{21}	+2.6
TSX	450	2.6×10^{21}	+4.1
CSF	450	2.85×10^{21}	+3.1
CSF	775	3.06×10^{21}	+2.1

(a) Annealed at 600 °C before exposure to sodium.

(b) Annealed at 1000 °C before exposure to sodium.

increased with increasing neutron exposure. TSX graphite, which is more anisotropic than CSF or TSGBF, also expanded more for equivalent irradiation conditions. When irradiated samples are annealed at high temperatures prior to exposure to sodium, length changes are about the same as for unirradiated samples.

A sample of CSF graphite irradiated at 80°C to 2.2×10^{20} nvt, expanded to about 2 1/2 times its original length upon exposure to molten sodium at 120 °C (Figure 3.10). X-ray diffraction measurements were made before and after

the sample was treated with sodium. The interlayer spacing (002) was unchanged from 7.410 Å, but line broadening decreased after exposure to sodium.

The exothermic reactions of sodium and potassium with irradiated graphite were observed in a differential scanning calorimeter. As reported in the literature, the heat of reaction of graphite with potassium is 81 ± 2 cal/g at 66 to 95 °C. The heat of reaction of an unirradiated graphite sample with potassium was 83 cal/g in this study. Several measurements were

Neg 43642-1

FIGURE 3.10. Irradiated Graphite Samples Before and After Reaction with Sodium at 120 °C. Samples were irradiated at 80 °C to 2.2×10^{20} nvt.

made with CSF graphite irradiated at 80 °C to 2.2×10^{20} nvt with an estimated total stored energy of 300 cal/g. When the irradiated CSF graphite was heated with potassium, the exothermic energy release was 138 cal/g. Annealing the graphite at 250 °C reduced the energy release to 90 cal/g. The reaction of sodium with CSF graphite irradiated to 2.2×10^{20} nvt released 136 cal/g for an unannealed sample and 65 cal/g for a sample annealed at 250 °C. Since all of the stored energy would not be removed by annealing at 250 °C, the heat of reaction of graphite and sodium is probably less than 65 cal/g.

Boronated graphite samples containing 7 wt% Boron as B_4C particles were irradiated at five temperatures between 301 and 473 °C and fluences ranging from 2.2 to 3.5×10^{20} nvt ($E > 0.18$ MeV). Expansions of 5.6 to 8% were measured after the irradi-

ated samples were heated with sodium for 100 hr at 1000 °F (538 °C). No correlation with irradiation temperature or fluence was obtained. Expansion of the boronated graphite was larger than nonboronated graphite as would be expected because of the greater damage from the $B^{10}(n,\alpha)Li^7$ reaction.

GRAPHITE-RESTRAINT IRRADIATION TEST

A. L. Pitner

The second capsule in the graphite-restraint irradiation test series, H-3-51, was discharged from the General Electric Test Reactor (GETR) in September. The purpose of the experiment is to determine the effect of sample size and low compressive stress on the turnaround behavior of graphite. Small-sample irradiations have indicated that the rapid expansion following turnaround may limit lifetimes of future graphite reactors.

This program will determine whether the expansion can be delayed or restrained by a larger sample size (due to possible different stress relief and creep behavior) or by a low compressive stress (50 psi).

Sample sizes in the test are nominally 1.75 in. diam $\times$ 2.25 in. long. Previous irradiation data have been collected primarily on samples 0.25 in. diam $\times$ 1.75 in. long. Sample types in the experiment are mostly conventional graphites (CSF, NC8, and TSGBF) on which extensive small-sample data have been accumulated, thus a good comparison of irradiation behavior can be made. Some isotropic graphites, which have shown improved dimensional behavior, are also included in the test. A total of 13 samples are included in the capsule, 5 unstressed and 8 compressed at 50 psi by a bellows. Sample irradiation temperatures range from 600 to 1100 °C.

The foil monitors, from which fluence will be determined, are now being counted. However, it is estimated that the maximum will be about 6×10^{21} nvt ($E > 0.18$ MeV), which is beyond the high-temperature turnaround point for transverse samples. To the present doses, it appears that the large unstressed samples are displaying behavior similar to that of small samples. Turnaround has been evidenced in several of the conventional graphites, and some have again expanded beyond their virgin length. It was reported in the last Quarterly Report⁽¹⁾ that the large samples might have been starting to expand at a lower fluence than small samples,

but the exposures at that time were overestimated. The corrected exposures resulted in good agreement between large and small sample data. The large-sample isotropic graphites, which do not contract but only expand with irradiation, also show good agreement with small-sample behavior up to the maximum exposure of 4×10^{21} nvt attained on these types.

The stressed samples again display behavior similar to that of unstressed small samples up to turnaround. One stressed sample, a TSGBF at 900 °C, which has surpassed the small-sample turnaround exposure, shows no indication of turning around. However, further exposure is needed to firmly define the effect of the 50 psi stress on the postturnaround irradiation behavior of this and other sample types.

This is the last capsule in this test to be irradiated in the GETR. Transfer of all or part of the test to the ETR is being evaluated.

SIZE-EFFECT STUDIES

W. J. Gray

Analysis of the neutron flux monitors from one of the Hanford size-effect experiments⁽²⁾ has been completed, and these data together with the dimensional change data are being analyzed.

An experiment is being designed to determine the effect of cold-seeding on the irradiation-induced contraction of nuclear graphite. In this case, cold-seeding is a term used to indicate the effect on graphite of a low neutron influence ($\sim 10 \times 10^{19}$ nvt

$E > 0.18$ MeV) at relatively low temperatures. There is some evidence that cold-seeding may result in a factor of two to four increase in the contraction rate of graphite when it is subsequently irradiated at higher temperatures. Cold-seeding might partially explain some of the early size-effect results from this laboratory.

THE EFFECT OF STRESS ON THE ELASTIC MODULUS OF GRAPHITE

P. E. Hart

Models have been proposed for the thermal-expansion behavior of anisotropic materials.⁽³⁾ In these models, crystallite failure during cooling from elevated temperatures is considered to result in a reduction of thermal expansion. To assess proposed models, the effect of prestressing on thermal expansion has been previously determined.⁽¹⁾ In this work, the effect of stress and annealing on the elastic modulus of graphite samples has been determined. Two graphites were used: ATJ, a graphite of moderate isotropy; and POCO grade PE-1924, a higher density, finer-grained, isotropic graphite.

Samples prestressed in compression were 0.5 in. diam $\times$ 3 in. long while those prestressed in tension were 0.426 in. diam $\times$ 4 in. long. Samples were annealed at 2400 °C to relieve machining stresses. Preloading was performed as discussed previously while elastic moduli were calculated from resonance frequencies measured at room temperature.

The effect of heat treatment on the moduli of samples prestressed at

80% of compressive strength was also determined. ATJ specimens were annealed for 1/2 hr in 200 °C intervals, starting at 200 °C after prestressing. Annealing was performed in air below 600 °C and in vacuum at higher temperatures. Resonance frequencies were measured after each annealing.

The effect of prestressing in tension and compression on elastic moduli is shown in Figure 3.11 as a function of applied load. The effect of heat treatment on the moduli of ATJ transverse samples loaded in compression is shown in Figure 3.12.

Preloading in either tension or compression resulted in a reduction in moduli. Samples loaded to 90% of their ultimate tensile strength exhibited lower reduction in moduli than those loaded in compression to only 30% of their compressive strength. In terms of actual load, tensile-loading data fit well with compressive-loading data.

The decrease in elastic moduli caused by prestressing in either tension or compression indicates that both modes of loading cause crack formation. Previously, the observed effect of prestressing in compression was interpreted in light of crack closure. Although compressive loading may cause some crack closure, crack formation appears to be the predominant effect.

Heating of prestressed samples to only 200 °C caused an increase in the moduli. Annealing-induced change was found to continue up to about 1400 °C. This change in moduli is reflected in the observed annealing

FIGURE 3.11. Effect of Stress on Elastic Modulus

FIGURE 3.12. Effect of Annealing on Elastic Modulus

effect of fractional expansion of pre-compressed samples, an effect attributed to the annealing out of stress-induced damage.

It is concluded that prestressing of graphite in either tension or compression up to ultimate strengths causes a reduction in elastic modulus. Uniaxial compressive loading, therefore, does not appear to cause significant crack closure.

Elastic moduli from this work will be used to test the validity of the models predicting thermal-expansion behavior of anisotropic materials.⁽⁴⁾

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FUEL DEVELOPMENT FOR GAS-COOLED REACTOR - HTGR GRAPHITE STUDIES

R. E. Nightingale

THE RADIOLYTIC REACTION OF GRAPHITE
WITH WATER VAPOR

R. E. Woodley

The radiolytic reaction of graphite with water vapor is of considerable importance to the technology of graphite-moderated reactors because water vapor is a common contaminant in otherwise inert reactor atmospheres. To investigate the reaction, ampoules of either Pyrex or quartz containing graphite, helium, and water vapor have been exposed to gamma radiation in a ^{60}Co facility. In addition, ampoules containing only helium and water vapor have been irradiated to determine what reaction products occur in the absence of graphite. While the reaction products CO_2 and H_2 are produced in greater concentration in ampoules containing graphite, it has been found generally that both gases are also produced in significant amounts in ampoules containing no purposely added carbon.⁽¹⁾

It has not been possible to completely remove the unknown carbonaceous impurity from the Pyrex ampoules, even with freshly prepared chromic acid. Because Pyrex cannot be heated to high temperatures to oxidize the source of carbon, the use of Pyrex ampoules has been discontinued and quartz ampoules have been used instead. To date, eight

ampoules, which were heated in air at 900 °C for about 8 hr prior to their filling, have been prepared, irradiated, and their contents analyzed. Six of the ampoules contained graphite samples weighing about 7.5 g, roughly 20 torr of water vapor, and about 0.5 atm of helium. The remaining two ampoules contained water vapor and helium but no graphite. The radiolytic yields resulting from the irradiation of these ampoules are presented in Table 4.1.

It is readily apparent from Table 4.1 that even such a drastic treatment as air oxidation at 900 °C is not sufficient to remove the carbonaceous impurity. However, as previously found, $G(\text{CO}_2)$ and $G(\text{CH}_4)$ are larger in the case of ampoules which contain graphite samples.

An additional eight quartz ampoules, which were given somewhat different preliminary treatments, have been prepared and irradiated. Of the six ampoules which did not contain graphite samples, two were heated overnight in air at 900 °C, two were soaked overnight in chromic acid, and two were given both of these treatments prior to their filling. The remaining two ampoules did contain graphite samples and also underwent both treatments prior to being filled. Gas analysis of these eight ampoules is in progress.

TABLE 4.1. Radiolytic Yields from Irradiated Ampoules

Ampoule No.	Contents	Dose x 10 ⁻¹⁷ , eV/cm ³			
			G(CO ₂)	G(H ₂)	G(CH ₄)
1	C+He+H ₂ O	1.08	7.59	15.2	1.13
2	C+He+H ₂ O	1.08	2.69	4.92	0.43
3	C+He+H ₂ O	2.03	2.41	2.94	0.12
4	C+He+H ₂ O	2.03	5.60	7.96	0.34
5	C+He+H ₂ O	3.00	2.42	4.00	0.19
6	C+He+H ₂ O	4.00	1.70	4.27	0.14
7	He+H ₂ O	0.99	1.30	4.70	--
8	He+H ₂ O	2.96	1.28	3.64	0.10

ANALYSIS OF GRAPHITE OXIDATION AND GAS COMPOSITIONS IN AN HTGR

G. L. Tingey and R. C. Giberson

The Peach Bottom HTGR is a 40 MW_e prototype of a high-temperature helium-cooled system. Previously, it has been assumed that the major source of impurity was ingress of steam from the heat exchangers and that reaction would lead to equal concentrations of H₂ and CO from the reaction

However, gas analysis of the coolant gas shows H₂ concentrations larger than CO by a factor of 20.⁽²⁾ There are a number of possible sources for the excess hydrogen including the following:

- Nuclear processes where hydrogen is formed by neutron reactions
- Outgassing of reactor components
- In-leakage of oil followed by cracking with the formation of hydrogen

- Proton diffusion as a result of steam-side corrosion of the heat exchanger
- Water ingress followed by preferential oxygen cleanup.

Of these, the first source listed is not considered further since a simple calculation of hydrogen buildup caused by nuclear processes clearly shows that the hydrogen formed is insignificant relative to the amount observed in the reactor.

Although outgassing of graphite in other reactor components was undoubtedly a significant process in the early stages of reactor operation, the following calculations show it to be a very small effect after several hundred hours of operation. Originally HLM-85 graphite contained about 20 ppm of hydrogen by weight. Currently a sample from the Peach Bottom Reactor contains about 5 ppm. This level of hydrogen appears to be in equilibrium with the hydrogen in the gas phase of the reactor. If the desorption rate required to maintain the hydrogen is assumed to be 10 ppm, the calculated time required to remove all of the hydrogen from the graphite is only 2000 hr of reactor operation. The rate of hydrogen generation from graphite by the outgassing mechanism would decrease with time as the hydrogen adsorbed on the graphite decreased. A decrease with time of the hydrogen concentration has not been observed. After 5000 hr of Peach Bottom operation, the hydrogen levels due to

outgassing of the graphite must be well below the levels observed in the gas coolant.

Although many precautions have been taken to avoid oil leakage into the primary coolant of the reactor, some potential still exists for this process. Several attempts⁽³⁾ have been made recently to detect traces of oil in the primary coolant or of hydrocarbon deposits in the graphite itself. After evaluating these data, it appears quite unlikely that this mechanism is contributing significantly to the hydrogen impurity.

For proton diffusion to be responsible for the hydrogen observed, either a very rapid corrosion rate of the heat exchanger is required or nearly 100% of the hydrogen generated by corrosion must diffuse through the heat exchanger tubes into the primary circuit. This mechanism appears to have potential since hydrogen diffusion rates are probably sufficiently rapid to admit essentially all of the hydrogen into the primary circuit at the temperatures of interest.

The fifth possible mechanism considered was that of in-leakage of water into the reactor system, followed by preferential removal of oxygen-containing species in the cleanup system. The cleanup system of the Peach Bottom Reactor is significantly different from that of the Fort Saint Vrain Reactor. Whereas in Fort Saint Vrain all reacting species are removed from the helium coolant at an equal rate,

the Peach Bottom system removes species at varying rates depending on their absorbance in the various sections of the purification system. For example, the fractional removal of carbon dioxide and water vapor are about five or six times that of hydrogen. Likewise, the rate of removal of CO is possibly two or three times the rate of removal of hydrogen from the gas coolant.

The GOP (Graphite Oxidation Program) computer code has been modified to allow for variable purification factors, and also to permit a hydrogen in-leakage rate. Some computer analyses have been made to assess the effects of proton diffusion and water ingress.

Steady-state calculations of the gas compositions in Peach Bottom have been made at two levels of graphite chemical reactivity designated 0.1K and 1.0K. The 0.1K is typical of nuclear graphite which has had no previous oxidation and has a low impurity level (~ 10 ppm). The 1.0K level is typical of the reactivity of the same graphite after 5% oxidation. The calculated steady-state levels are given in Table 4.2.

The H_2/CO ratios, as expected, are the same as the purification ratios for these species. When the radiation reactions are included, the ratios change substantially. Presently, we have not been able to determine steady-state concentrations for these conditions, but it appears that the H_2/CO ratio at steady-state would be greater than 10. The calculated steady-state

TABLE 4.2. Steady-State Gas Compositions in Peach Bottom for Thermal Reactions Only

Level	Concentration in Torr				
	H ₂ O	CO	CO ₂	H ₂	H ₂ /CO
0.1K	1.39×10^{-2}	2.10×10^{-3}	3.52×10^{-7}	3.30×10^{-3}	1.57
1.0K	1.31×10^{-2}	5.14×10^{-3}	5.00×10^{-7}	8.08×10^{-3}	1.57

hydrogen level, however, is less than 2 ppm compared with the measured value of 9 ppm. The possible reason for this discrepancy is that the assumed water leak rate or the assumed graphite reactivity is too low. Computer runs to determine the leak rate or reactivity necessary to obtain concentrations in the range observed will be made.

The other possible mechanism, i.e., hydrogen in-leakage from the steam-side corrosion could also explain the results. Additional information is needed before the magnitude of this effect can be assessed on the computer.

Another unique feature of the Peach Bottom HTGR is that it has a "purge" flow where helium flows slowly down a channel adjacent to the fuel to purge out fission products. The temperature of the graphite in this area is as much as 300 °C higher than the maximum moderator temperature. It is likely that water vapor in the purge stream

is quantitatively removed via the steam-carbon reaction. The outlet from this stream feeds directly into a special cleanup system, and any removal of CO or conversion of CO to CO₂ in this system will give rise to a high H₂/CO ratio. The computer code is currently being modified to test this condition.

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NUCLEAR CERAMICS

R. E. Nightingale

MICROSTRUCTURE AND TRANSMISSION
ELECTRON MICROSCOPY OF NUCLEAR
SINTERED UO_2 -25 wt% PuO_2

S. J. Mayhan and J. L. Daniel

Transmission-electron microscopy studies of polycrystalline nuclear ceramics have continued with the examination of sintered UO_2 -25 wt% PuO_2 . Although large areas of the specimen were sufficiently thinned for electron diffraction, only numerous small edge projections were suitable for transmission microscopy (Figure 5.1).

The specimen was thinned to electron transparency by a combined mechanical-thinning and chemical-polishing technique similar to that

reported for sintered UO_2 .⁽¹⁾ The recently developed dilute HNO_3 -HF-cerium nitrate etchant was used for chemical polishing.⁽²⁾ A fine pattern of interference striations with 50 Å spacing was evident in all transparent areas, resembling the "fringes" observed in some specimens of single-crystal UO_2 . No pronounced internal defects were evident. The spots comprising the well-ordered crystalline diffraction pattern were each composed of multiple reflections, consistent with the appearance of interference striations.

The difficulties experienced in thinning coprecipitated UO_2 -25 wt%

Neg J-4933

160,000X

FIGURE 5.1. Transmission Electron Micrograph of UO_2 -25 wt% PuO_2 Mechanically Thinned and Chemically Polished. Interference Striations Are Spaced at 50 Å.

$\text{PuO}_2^{(3)}$ were again encountered, resulting in the loss of some thinned edges. It appears that extreme brittleness and a tendency to spall are characteristic of the unirradiated mixed oxides.

PROPERTIES OF MOLTEN OXIDE FUELS

R. P. Nelson

Mixed uranium-plutonium oxide fuels may experience power transients which cause partial or, in extreme cases, total melting of the fuel. Adequate safety analyses of such events require data for such fundamental liquid properties as volume expansion on melting, expansion coefficient, compressibility, heat of fusion, heat capacity, thermal conductivity, viscosity, surface tension, and other related properties. This information is nonexistent for mixed uranium-plutonium oxides, and only limited data are available for uranium dioxide.

Previous work at PNL⁽⁴⁾ has shown that substantial increases in volume (~10%) occur during melting of UO_2 . The extent to which typical LMFBR oxide fuel compositions expand during melting must be known to either provide special accommodation in the fuel-pin design or to predict the extent of pin deformation associated with a given partial-melting transient. A proper cladding stress-deformation analysis also requires data on compressibility. Fuel distribution during such a transient is also of concern and is related most directly to liquid oxide density, viscosity, and sur-

face tension. Only an approximate value for the surface tension of liquid UO_2 has been measured.⁽⁵⁾ Energy release during severe overpower transients is expressed by high-temperature state equations in terms of critical volumes, temperature, and pressures. These equations, fundamental to safety analyses of severe overpower transients, require special measurement methods for their development.

Several experimental techniques have been developed or considered for measurement of the properties of molten uranium-plutonium oxides relating to mechanical design and safety of LMFBR fuels. Common to all techniques is the control of stoichiometry through the employment of oxide samples contained in closed refractory-metal capsules.

Expansion

Change in volume on melting and liquid expansion coefficient is measured at temperatures to 3000 °C by using radiography to observe a change in liquid column height as a function of temperature. Self-resistance-heated capsules of tungsten or rhenium containing the oxide are radiographed by using either an isotopic source (^{60}Co , ^{137}Ce , ^{192}Ir , etc.) or 150 kV X-rays (Figure 5.2). The high X-ray and gamma mass-absorption coefficients of tungsten and rhenium require long exposures and still radiography. A 300 kV X-ray source and a high-resolution video-tape recorder will be obtained to permit continuous observation

and image recording. Preliminary experiments to 3000 °C on UO_2 have been completed to test the apparatus.

Viscosity

Two methods have been developed for measurement of shear viscosity: the falling-sphere method using Stokes' Law⁽⁶⁾ and the oscillating torsional pendulum technique.⁽⁷⁾ In the first method, a 0.030 to 0.040 in. tungsten sphere is suspended by a small UO_2 - PuO_2 "fuse" above the liquid in a closed tungsten or rhenium capsule which is self-

resistance heated (Figure 5.2). The liquid temperature is essentially uniform, and the sphere (located near the upper end of the capsule) is at a lower temperature. As the liquid temperature is slowly increased, the sphere is released to fall when its UO_2 - PuO_2 support melts. Adjustment of the position of the "fuse" and sphere allows measurement at different temperatures. The velocity of fall in the liquid is measured by continuous (300 kV) X-radiography and stop-motion playback of the video-tape record.

FIGURE 5.2. Schematic of Radiographic System for Liquid Expansion, Viscosity, and Surface Tension Measurements

The torsional pendulum technique consists basically of measuring the time-rate of decrease in amplitude of a freely oscillating closed tungsten or rhenium crucible (containing the liquid) suspended from a torsion wire. The system (Figure 5.3) is similar to one developed by Wittenberg et al.⁽⁸⁾ The amplitude decrease is measured by replaying a video-tape record of the scale movement in stop action mode, and the total period of a number of successive oscillations is recorded

on an electronic counter activated by a photo-cell circuit. The system is currently being calibrated by using known viscosity liquids.

Surface Tension

Conventional pendant and sessile-drop techniques⁽⁹⁾ for measurement of surface tension will be investigated. Configuration of liquid drops contained in closed capsules will be determined by X-radiography. Methods based on equilibrium meniscus shape⁽¹⁰⁾ will also be investigated.

Thermal Conductivity

Studies of sample holder design for measurement of thermal diffusivity of mixed oxide liquids by the laser-pulse method⁽¹¹⁾ have been initiated. Values of thermal diffusivity will be combined with measured values of heat capacity to yield thermal conductivity.

Compressibility

Liquid compressibility may be determined by measurements of ultrasonic velocity. A measuring system which may be used for measurement of the velocity of a longitudinal acoustic wave by a phase-comparison method has been assembled.* Design of a tungsten cell for containment of the liquid oxide and pulse transmission has been initiated.

FIGURE 5.3. Oscillating Cup Viscometer

* O. D. Slagle. High Temperature Velocity Measurements in Fast Reactors Fuels. p. 5.6 of this report.

IN-REACTOR CREEP OF (U,Pu)O₂

J. A. Basmajian

Swelling of mixed-oxide fuels caused by fission gases and solid fission products results in high stresses being applied to the clad of tubular fuel elements. The fuel operating in the temperature range 800 to 1500 °C, where fission-gas release does not occur, is primarily responsible for the phenomenon. Data on in-reactor creep properties of mixed-oxide fuels in this temperature range are required for fuel design so that clad-strain limitations of high-performance fuel elements will not be exceeded. Tests designed for the Hanford K-Reactor Snout facilities will be used to obtain the creep data.

The Snout facilities are horizontal positions in the Hanford K-Reactor which can be utilized for uncomplicated capsule assemblies. Capsules can be charged and discharged at controlled rates during reactor operation. Since in-reactor instrumentation is available, well-characterized tests can be run. The working arrangements will allow experiments to be completed from concept to operation in a short time. Initial experiments will be short-time bend-test irradiations of rectangular beams of (U,Pu)O₂ and UO₂. Tests of high-burnup fuel will be run later in the program.

The initial experimental capsules are being designed, and reference fuel compositions for FTR will be used for sample material. The 800 to 1200 °C temperature range will be investigated initially. Stress levels will

be chosen on the basis of out-of-reactor creep studies currently in progress. Comparison of the results of these in-reactor experiments with the results from duplicate out-of-reactor bench tests will permit determination of fission-rate effects on creep.

HIGH-TEMPERATURE CREEP BEHAVIOR OF FAST-REACTOR OXIDES

O. D. Slagle

Final modifications to the high-temperature creep apparatus have been completed, and the plutonium glove-box enclosing the equipment was subsequently leak-checked and placed on plutonium control. The apparatus consists of an Astro high-temperature graphite furnace mounted on a table-model Instron loading frame. The furnace is capable of temperatures up to 2500 °C, but is currently being used with an alumina muffle so that maximum temperatures will be restricted to less than 1700 °C. The Instron loading frame, on which the creep apparatus is mounted, is a special frame which has no strain rate controls itself, but is driven by a power take-off from a normal frame. The two frames are coupled by a rotary vacuum seal through the hood wall.

Deformation of the sample is by four-point bending in an alumina sample holder. Alumina was chosen as the most suitable material because of its stability to high temperatures in both reducing and oxidizing atmospheres, as required for UO₂ and PuO₂, respectively. Four-point bending was

selected over compressive creep because of the lower stress involved, thus minimizing the amount of plastic deformation occurring in the load train.

The load on the sample is measured by a low-capacity (250 lb) compressive load cell. A constant load can be applied to the sample by the use of limiting switches controlling the up and down movement of the cross-head. A linear displacement transducer is used to monitor the amount of sample deflection. With the above capabilities, the creep properties of a material can be determined either by applying a constant load and measuring the strain rate or by applying a constant strain rate and measuring the resultant equilibrium stress.

The sample size required for measurement is 1.25 in. $\times$ 0.25 in. $\times$ 0.1 in. To obtain samples of this size, cylindrical pellets 1 1/2 in. diam $\times$ 1/2 in. high are pressed and sintered. Four samples of sufficient length can be cut from the resultant pellet. After being cut to size, the samples are mounted on a special holder, and the sides are ground parallel over the 0.1 in. thickness.

Samples of UO_2 have been successfully prepared by dry-pressing the UO_2 powder at 50,000 psi using butyl stearate as a die lubricant. Initial attempts at pressing UO_2 -20 wt% PuO_2 coprecipitated mixture by this procedure were unsuccessful. Suitable pellets of this material have been obtained by the following procedure:

1. Ball-milling in water for 6 hr
using a tungsten-carbide mill
2. Addition of 2 wt% carbowax

3. Pressing at 15,000 psi

4. Sintering at 1650 for 8 hr.

Mechanically mixed UO_2 -25 wt% PuO_2 containing 4 wt% carbowax has also been successfully pressed and sintered under the above conditions.

The determination of the creep properties of mechanically mixed UO_2 -25 wt% PuO_2 in the temperature range from 1200 to 1500 °C has begun. This study will involve the dependence of creep properties on temperature, stress, porosity, and stoichiometry. Materials to be studied in the future include UO_2 -(20, 40, 60, and 80) wt% PuO_2 and PuO_2 .

HIGH-TEMPERATURE ULTRASONIC VELOCITY MEASUREMENTS IN FAST-REACTOR OXIDES

O. D. Slagle

Setting up the basic equipment for ultrasonic velocity measurements using the phase-comparison technique⁽¹²⁾ and initial measurements on UO_2 and CaF_2 were discussed in the previous quarterly report.⁽²⁾ During this quarter, the high-temperature sample holder was fabricated, and work was begun on finding a good high-temperature bonding material.

Figure 5.4 is a schematic diagram of the apparatus to be used for the high-temperature ultrasonic velocity measurements in solids. Presently, fused silica is being used for the buffer rod to transmit the ultrasonic waves from the transducer to the sample; this limits temperatures to less than 1200 °C, although the furnace is capable of temperatures up to 1700 °C. The upper fused silica rod, in conjunction with the adjustable

FIGURE 5.4. Apparatus for High-Temperature Ultrasonic Velocity Measurements

spring, helps to hold the sample firmly on the end of the buffer rod. Platinum foil is used between the sample and the upper fused silica rod to prevent a reaction from occurring. The sample holder is sealed from the atmosphere by silicone O-rings so that a reducing atmosphere as needed by UO_2 can be used. Bonding materials being considered are thin gold and platinum foils and ceramic adhesives such as Aremco Ceramabond 503.

Extending this method to higher temperatures requires a material other than fused silica for a buffer

rod. Preliminary tests indicate that single-crystal tungsten rod may be suitable. In addition to making possible measurements in solids at high temperatures, the temperature capabilities of tungsten make it appear possible to make longitudinal velocity measurements in molten nuclear oxides. Since the shear modulus in a liquid is zero, the longitudinal modulus is all that is necessary to determine bulk modulus. Consideration is now being given to the design of a cell for performing such measurements on molten nuclear oxides.

EFFECT OF OXYGEN STOICHIOMETRY ON THE THERMAL DIFFUSIVITY AND CONDUCTIVITY OF $\text{U}_{0.75}\text{Pu}_{0.25}\text{O}_{2-x}$
R. L. Gibby

The thermal diffusivity of sintered mixed oxide was measured from 100 to 1200 °C as a function of O/M from 1.93 to 2.00. Thermal conductivity was calculated from the measured thermal diffusivity and estimated values for heat capacity. Measurements were made with a laser heat-pulse apparatus.

The properties of the specimens are given in Table 5.1. The samples were prepared by dry blending PuO_2 and UO_2 powders for 72 hr in a ball mill, dry pressing the powder into pellets, and sintering the pellets at 1650 °C for 24 hr in a cover atmosphere of argon-8% hydrogen.

The O/M of the stoichiometric sintered pellet was adjusted to the desired hypostoichiometric value by heating them between 1150 and

TABLE 5.1. Properties of Mechanically Mixed and Sintered ($U_{0.75}Pu_{0.25}O_{2+x}$)

Lattice Parameter a_o :	(O/M = 2.000)	5.4515 Å
	(O/M = 1.97)	5.4601
	(O/M = 1.95)	5.4649
	(O/M = 1.93)	5.4730
Plutonium, wt%:	21.6	
Theoretical Density	95.5	
Spectrochemical Analysis; ppm:		
Ag	<1	Na 2
Al	50	Ni 50
B	<0.1	P <50
Ca	<1	Pb 2
Cr	<5	Si 12
Cu	20	Sn 5
Fe	120	V <20
K	20	Zn 50
Mg	2	
Mn	2	
Mo	<5	

1600 °C in an ultrapure hydrogen atmosphere. An O/M of 1.93 was obtained after a 2 hr heat treatment at 1600 °C, while an O/M of 1.97 required only 1 hr at 1200 °C.

The O/M of the stoichiometric specimen was controlled during thermal-diffusivity measurements with a cover gas of argon-8% hydrogen passed through 0 °C water. For hypostoichiometric compositions, a dry argon-8% hydrogen cover gas was employed.

Only two test specimens were used for the study so that O/M was essentially the only variable between runs.

Thermal conductivity was calculated by using the relationship

$$\lambda = 4.183 \alpha \rho C_p \text{ (W/cm } ^\circ\text{C)}$$

where α is the thermal diffusivity, ρ is the measured density, and C_p is the heat capacity. Values of heat

capacity were calculated by combining (according to mole fraction in the solid solution) the heat capacities of PuO_2 ⁽¹³⁾ and UO_2 ⁽¹⁴⁾

The results of this study for each O/M obeyed the empirical relationships

$$1/\alpha = A' + B'T$$

$$1/\lambda = A + BT$$

where the temperature coefficients B' and B were independent of the oxygen content while the terms A' and A increased linearly with x for the compositions $U_{0.75}Pu_{0.25}O_{2-x}$.

The general expressions for α and λ in terms of oxygen content and temperature are, according to this study,

$$1/\alpha = 0.110T + 1.08 \times 10^3 \text{ (x)} \\ + 4 \text{ (sec/cm}^2\text{)}$$

$$1/\lambda = 0.0312T + 3.55 \times 10^2 (x) \\ + 1.5 \text{ (cm } ^\circ\text{K/W)}$$

with temperature units of $^\circ\text{K}$.

The linear variation of A with x is in agreement with Klemens' theory⁽¹⁵⁾ for phonon scattering by point defects. The theoretical value of A as a function of x was calculated by using Ambegaokar's⁽¹⁶⁾ relationship for point defect scattering at high temperatures

$$A = \frac{4 \theta V_o \pi^2 \Gamma}{h v^2}$$

where θ is the Debye temperature, V_o is the atomic volume, h is Plank's constant, and v is the phonon velocity. Γ is equivalent to a scattering cross section and is given by the expression⁽¹⁷⁾

$$\Gamma_i = \frac{V_o \epsilon_i}{12} \left[\left(\frac{\Delta M_i}{M} \right)^2 + n \left(\frac{\Delta R_i}{R} \right)^2 \right]$$

$$n = 32(1 + 1.6\gamma)^2$$

where ϵ_i is the concentration of the i^{th} defect per unit volume, ΔM_i is the mass difference and ΔR_i is the size difference of the i^{th} defect, and γ is the Gruneisen constant.

Calculations using the above expressions indicate that the defect thermal resistance of hypostoichiometric mixed oxides arises from phonon scattering from the lattice strain surrounding oxygen vacancies and Pu^{+3} ions, both of which are larger than the host site which they occupy. The thermal resistance from

the mass difference of the oxygen vacancies is negligible according to these calculations. The experimental results are compared with the theoretical calculations in Figure 5.5.

Neg 0683413

FIGURE 5.5. The Defect Thermal Resistance, A , as a Function of Number of Moles of Missing Oxygen Atoms/Mole Mixed Oxide, x .

ELECTRICAL RESISTIVITY OF URANIUM MONOTRIDE

J. Lambert Bates

The electrical resistivity of high-energy-rate impacted uranium monitride was measured from 25 to 2550 $^\circ\text{C}$ using a four-contact technique.⁽¹⁸⁾ To prevent the formation of the higher uranium nitrides at temperatures

below 1150 °C, low-temperature measurements were made in either vacuum ($<10^{-4}$ torr) or purified 8% hydrogen-92% argon (1 atm). Above approximately 1150 °C, an atmosphere of 5% hydrogen and 95% nitrogen (1 atm) was used to prevent decomposition of the nitride and to provide a low-oxygen partial pressure. The hydrogen mixtures of argon and nitrogen contained less than 5 ppm H_2O and 1 ppm oxygen (inlet compositions).

Measurements were made on heating and cooling during three thermal cycles (Table 5.2).

The UN specimen was fabricated by high-energy-rate impaction in an evacuated stainless steel can at 1200 °C. A 0.635 cm diam and 1.91 cm long specimen was machined from the larger compact. External examination of the specimen indicated presence of some small cracks near the ends of the specimen, outside the positions of the voltage probes and measurement section.

Significant changes in electrical resistivity occurred during heating

the UN to 2550 °C. Abrupt changes in resistivity were observed at 400 to 600 °C and 1000 to 1400 °C. The electrical resistance of the as-impacted sample at room temperature was approximately 3 times greater than the resistivity of sintered UN.^(19, 20) During heating, the resistivity increased rapidly to 400 °C, decreased rapidly from 400 to 600 °C, and increased again between 600 and 900 °C. From 900 to 1300 °C, the resistivity decreased by approximately 50%. During cooling, the resistivity decreased slowly with temperature with values significantly less than those observed on heating.

The electrical resistivity on heating from 25 to 1200 °C during the second cycle reproduced closely the values measured during cooling in the previous thermal cycle. Continued decreases in resistivity occurred above 1400 °C, matching closely the first heating cycle. The resistivity increased slightly with temperatures above 1800 °C. The resistivity values during cooling were slightly less than during the heating cycle.

The resistivity during both cooling and heating in thermal Cycle 3 were essentially the same as the cooling curve values of Cycle 2.

Some densification of the specimen occurred during the second cycle, and the diameter of the specimen decreased from 0.635 to 0.605 cm. The surface of the specimen also exhibited a craze-like appearance; however, this was readily removed by grinding prior to the third thermal cycle. No changes were measured after the third cycle.

The reasons for these changes in electrical resistivity on heating are

TABLE 5.2. *Atmosphere During Thermal Cycles*

	Heating, °C	Vacuum	Atmosphere
Cycle 1	25 to 1400		10^{-4} torr
Cycle 2	25 to 1200	8% H_2 -92% Ar	1 atm
	1200 to 2550	5% H_2 -95% N_2	1 atm
	2550 to 1190		
	1190 to 25	8% H_2 -92% Ar	1 atm
Cycle 3	25 to 1150	8% H_2 -92% Ar	1 atm
	1150 to 2375	5% H-95% N_2	1 atm
	2375 to 1225		
	1225 to 25	8% H_2 -92% Ar	1 atm

not presently understood. Sintering, gas release, grain boundary changes, microcrack healing, strain and stress relief and other structural changes may be responsible. Characterization of the UN before and after measurements will be made to aid in the understanding of the results.

The final electrical resistivity values (to 1200 °C) are higher (Figure 5.6) than the reported values for UN. (19, 20) However, the shape of the temperature versus resistivity curves of the three experiments are similar.

FIGURE 5.6. Electrical Resistivity Values

IRRADIATION DAMAGE IN LOW-EXPOSURE URANIUM DIOXIDE: MELTING POINT

J. L. Bates, J. L. Daniel, and
S. J. Mayhan

The inter-relations of property changes induced during neutron irradiation of UO_2 are being investigated. Sintered UO_2 specimens and selected UO_2 single crystals were irradiated in a single capsule under controlled conditions with maximum oxide temperatures less than 100 °C. Four of the eight specimens had been previously irradiated to exposures of 0.4 to 1.4×10^{20} f/cm³. The final cumulative maximum exposure is estimated to be 3.8×10^{20} f/cm³.

The four previously irradiated sintered UO_2 specimens had been part of a study to investigate the effects of neutron irradiation on the thermal and electrical conductivity.^(21, 22) The irradiated specimens included in this present study were surplus samples from the earlier investigation. The two unirradiated sintered specimens were also part of this earlier program, but had not been irradiated.

Each of the UO_2 specimens were characterized prior to the final irradiation. Each sintered specimen was broken into two sections, one section being irradiated and the other being retained as a reference. The single crystals were from the same lot, with additional single crystals from the same source being retained as references. Preirradiation examination of the eight specimens include

- Knoop microhardness
- Lattice parameters
- Melting points

- Room-temperature fracture characteristics
- Microstructure.

The UO_2 specimens were 0.635 cm in diameter and varied in length from approximately 2.4 to 4.0 cm. The specimens were fitted into holes in split aluminum cylinders which were inserted into a 20 cm long, 4.81 cm OD container. The capsule was filled with purified helium and welded shut. Aluminum rods were used as spacers.

Melting-Point Changes

The melting point of the irradiated UO_2 was measured as a function of irradiation to approximately 3.8×10^{20} f/cm³. No change in the melting point from nonirradiated UO_2 was observed. Previous measurements of sibling irradiated UO_2 showed a significant increase in the melting point with increased exposure.⁽²³⁾ The melting point of UO_2 was observed to increase from 2800 to 2895 °C after an exposure of 1.1×10^{20} f/cm³. However, other studies⁽²²⁾ using similar sibling specimens stored for four years in air showed only relatively small changes after exposure to 1.4×10^{20} f/cm³.

Melting points were measured in 8% hydrogen and argon and pure argon atmospheres using a brightness pyrometer and a tungsten "V" filament furnace. This was the same apparatus used in the previous investigations.^(22, 23) Four to five melting points were measured for each exposure, with a precision of better than ± 10 °C for each measurement. An unirradiated oxide specimen from the

original UO_2 lot was used as the reference melting point. The furnace was calibrated with the use of materials with known melting points, i.e., copper, platinum, Al_2O_3 , and molybdenum. All irradiated samples were sintered UO_2 .

Results are summarized and compared with the previous measurements^(22, 23) in Table 5.3 and shown in Figure 5.7.

The data do agree with more recent melting-point measurements of high exposure UO_2 conducted by Christensen, Allio, and Biancheria.⁽²⁴⁾ These measurements were conducted at a different laboratory but with a similar technique and equipment to those developed at Battelle-Northwest. Although exposures were much larger, no

increase in melting points were observed and a linear decrease in melting point was measured at very high burnups.

The discrepancy between the data at low exposures is not readily explained. With the exception of the pyrometer, the same equipment was used for all of the melting point determinations. Although the environmental gases may have been changed, previous experience indicates that a small variation in gas purity and tungsten filament geometry does not significantly alter the observed melting points. Since this study is concerned primarily with the difference between nonirradiated and irradiated UO_2 , absolute values of the melting point are not as significant.

TABLE 5.3. Summary of Melting Points for Irradiated
Melting Points, °C

Exposure, f/cm ³	Specimen Designation	Christensen ⁽²³⁾	Bates and Daniel ⁽²²⁾	Present Study
0	Reference UO_2	2800	2800	2805
1.4×10^{19}	13		2784	
	232	2816		2804
4.1×10^{19}	27		2802	
	289	2876		
5.6×10^{19}	308,504	2884		
1.1×10^{20}	55		2791	
	610	2895		
$1.4 \times 10^{20(a)}$	63		2791	
$\sim 1.7 \times 10^{20(a)}$	R-2			2802
$\sim 1.9 \times 10^{20(a)}$	13			2795
$\sim 2.2 \times 10^{20(a)}$	27			2804
$\sim 2.8 \times 10^{20(a)}$	55			2805
$\sim 3.8 \times 10^{20(a)}$	63			2810

a. Exposure estimated; burnup analysis not completed.

FIGURE 5.7. Changes of UO_2 Melting Point After Irradiation (Exposure Estimated)

It was initially suggested that the four to five year storage period between the removal of the specimens from the reactor and the experimental measurements by Bates and Daniel⁽²²⁾ resulted in physical, structural, and compositional changes in the oxide--specifically, a change in O:U ratio and the isotopic fission-product composition. The increase in melting point at low exposure suggested a stabilization of hypostoichiometric UO_2 which is reported to have a

higher melting point⁽²⁵⁾ than stoichiometric UO_2 . However, the most recently irradiated UO_2 samples which were studied less than five months after removal from the reactor and less than two months after removal from the capsule did not confirm the increased melting points. Thus, increases in melting point by nonstoichiometric stabilization by irradiation can be questioned.

The reproducible results of the present study and the former study of

Bates and Daniel⁽²²⁾, coupled with the results from Christensen et al.⁽²⁴⁾ on high exposure UO_2 , suggest that irradiation exposure to 3×10^{19} f/cm³ at temperatures greater than 15 °C but less than 100 °C does not significantly alter the melting point of sintered, nearly stoichiometric UO_2 .

Differences were visibly observed in the vaporization and grain growth rates between the nonirradiated and irradiated specimens. Irradiation appeared to inhibit the vaporization and subsequent grain growth from the vapor. This difference was of sufficient magnitude to be observed directly through the optical pyrometer. These effects are similar to those reported by Christensen during the initial melting point studies.⁽²³⁾

MATERIALS AND INFORMATION EXCHANGE

H. J. Anderson

During this reporting period, nuclear materials and information exchanges were continued. The exchange involves the procurement for stockpiling of basic supplies, listing of purity and nuclear properties, purification and preparation of specimens of various geometries, and shipment of the material with known values to selected sites.

During this quarter, UO_2 single-crystal buoles were selected from stocks, annealed in a hydrogen atmosphere at 1750 °C for 8 hr, cooled, and characterized. These UO_2 buoles were then selected for use in several basic studies. Three large UO_2 buoles were packaged and shipped

to Battelle-Columbus and eight were sent to Barkeley Nuclear Laboratories (England) for use in basic studies. Two UO_2 single-crystal buoles (natural and depleted) were selected, characterized, and sent to Westinghouse Research Laboratories for basic studies use. In addition, UO_2 -20(wt%) PuO_2 pellets were selected, characterized, and sent to Oak Ridge National Laboratory (ORNL) to be used in fission gas release studies.

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FAST REACTOR NITRIDES RESEARCH

R. E. Nightingale

MELTING BEHAVIOR OF URANIUM-
PLUTONIUM MONONITRIDE FUEL
COMPOSITIONS

E. T. Weber

Melting studies in the UN-PuN system at nitrogen pressures from 1 to 100 atm have been reported previously.⁽¹⁾⁽²⁾ The most recent work has included measurements on samples of sintered mixed-nitride fuel compositions. The three different compositions used represented a standard metal-derived nitride and two compositions obtained by a carbothermic reduction synthesis scheme. The latter two were a standard or "best reproducible" composition (carbon and oxygen each <0.2%) and a high-carbon (~0.6%) high-oxygen (~0.9%) composition.

Measurements of the nitrogen decomposition pressure for these fuel materials, as well as on several specially prepared compositions at various U/Pu ratios, have been extended to pressures between 0.01 and 1 atm. A cold-wall tungsten filament furnace was used which was similar to the high pressure furnace previously described.⁽¹⁾ The system was capable of reaching high vacuum with an aneroid-type absolute pressure gauge used to measure nitrogen pressures down to 2mm Hg.

The results from the observation of the temperature of liquid formation in the sintered nitrides as a function of nitrogen pressure over four decades are shown in Figure 6.1.

The independence of the decomposition temperature on nitrogen pressure extends down to a pressure of 0.3 atm. A major departure of some data points at 1 atm from the trend of measurements at higher and lower pressures is anomalous. However, the lower pressure data are consistent with the high pressure measurements in that no significant effect of composition differences between samples was seen. Since these materials did not show the typical (for mononitride) pressure dependence to an identifiable congruent melting region, ceramographic examination of melted and quenched samples was used to distinguish between congruent and incongruent behavior. Below nitrogen pressures of 4 to 6 atm, a typical "peritectic"-type structure containing a metal phase was observed; above this pressure, melting occurred congruently. An explanation for the observed behavior of sintered nitrides is not yet clear. However, it possibly involves the effect of an appreciable but uncontrolled CO partial pressure for the compositions involved.

Several compositions in the UN-PuN pseudo-binary system were investigated, and Figure 6.2 shows that they conform to the typical nitrogen pressure dependence for decomposition. These samples were prepared by nitriding arc-cast U-Pu alloys and annealing the nitrides at 1600 °C. The temperature for

Neg 0683627-5

FIGURE 6.1. Pressure-Temperature Behavior for the Melting of Sintered $(U_{0.80}Pu_{0.20})$ Nitrides Over Four Decades of Nitrogen Pressure.

congruent melting (2760 °C) for the $(U_{0.75}Pu_{0.25})N$ composition was in good agreement with that obtained for the sintered fuel materials. A departure from ideal solidus-

liquidus configuration is indicated by the overlapping of the curves in the congruent melting region for the 25% and 50% PuN contents.

Neg 0683627-4

FIGURE 6.2. Pressure-Temperature Behavior for the Melting of Compositions in the UN-PuN Psuedo-binary System

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NONDESTRUCTIVE TESTING

G. J. Dau

EDDY CURRENT METHODS

Multiparameter Studies

H. L. Libby

A review of the literature of eddy-current nondestructive testing techniques shows the use of several systems of units and different ways of presenting the same data. For example, the normalized impedance or voltage plane diagrams for a solenoid containing an electrically conducting bar are treated differently by different authors as shown in Figure 7.1. Each of these has its special advantage and usage. Although they are related in a straightforward manner, the nature of the relationships is not obvious.

An often used diagram, in Figure 7.1a is shown one using mixed units, introduced by Foerster.^(1,2) A second representation using MKS units presented by Hochschild⁽³⁾ is shown in Figure 7.1b. A third little known representation⁽⁴⁾ (Figure 7.1c) is also included here because it helps to show the physical relationship between skin depth δ , and the impedance plane diagram.

In Figure 7.1, Foerster used, as the argument in the related Bessel functions, the quantity:

$$A = \frac{f \mu_{rel} \sigma d^2}{5066} \quad (1)$$

where f = frequency of the exciting field

σ = electrical conductivity of test material in meter/ohm-mm²

d = diameter of test object, cm

μ_{rel} = relative permeability of bar

He then defined the variable $\frac{f}{f_g}$,

$$\frac{f}{f_g} = \frac{f \mu_{rel} \sigma d^2}{5066} \quad (2)$$

and labeled computed impedance positions along the "conductivity" locus with their corresponding $\frac{f}{f_g}$ reference values.

Hochschild has used

$$ka = a \sqrt{\omega \mu \sigma} \quad (3)$$

as the argument of the related Bessel functions where

a = radius of bar, meter

ω = $2\pi \times$ frequency

μ = permeability of free space
space = $4\pi \times 10^{-7}$ H/m

σ = electrical conductivity, mhos/meter.

In Figure 7.1c the impedance positions have been labeled with reference numbers corresponding to the related ratios of radius to skin depth for the plane wave semi-infinite plane conductor.

When the conversion of the mixed units used in Figure 7.1 is made, it can be shown that the numbers on the diagram in Figure 7.1b are simply equal to the square root of the reference numbers associated with corresponding points on the diagram in

(a)
$$\frac{f}{f_g} = \frac{f_{\mu_{rel}} \sigma d^2}{5006}$$

f = Hertz
 σ = Meters/ohm-mm²
 d = Diameter of Bar, cm.

For the Same Bar and Frequency

(b)
$$Ka = a \sqrt{\omega \mu \sigma}$$

$\omega = 2 \pi f$
 f = Hertz
 $\mu = 4 \times 10^{-7}$ Henry/Meter
 σ = mhos/Meter
 a = Radius of Bar, Meters

$$Ka = \sqrt{\frac{f}{f_g}} = \sqrt{2} \frac{a}{\delta}$$

(c)
$$\frac{a}{\delta} = a \sqrt{f \mu \sigma} = \frac{a \sqrt{\omega \mu \sigma}}{\sqrt{2}} = \frac{Ka}{\sqrt{2}}$$

δ = Skin Depth, Plane Wave Case
 Units Same as in (b)

FIGURE 7.1 Three Impedance Diagrams for Solenoid Encircling Cylindrical Bar-Fill Factor = 1.0

Figure 7.1a. The Number 5066 is the factor relating the two systems of units used and appears as follows:

$$\frac{4}{(2\pi)(4\pi \times 10^{-7})(10^6)(10^{-4})} \omega \mu \sigma a^2 = \frac{f \mu_{rel} \sigma d^2}{5066} \quad (4)$$

$$\frac{5 \times 10^4}{\pi^2} = 5066 \omega \mu \sigma a^2 = \frac{f \mu_{rel} \sigma d^2}{5066} \quad (5)$$

$$ka = \omega \mu \sigma a^2 = \frac{f \mu_{rel} \sigma d^2}{5066} \quad (6)$$

The relationship between the numbers at corresponding impedance points of Figure 7.1b and Figure 7.1c is that the latter are equal to the former divided by $\sqrt{2}$. This follows from the relation:

$$\frac{a}{\delta} = a \sqrt{f \mu \sigma} = \frac{a \sqrt{\omega \mu \sigma}}{2}$$

The representation in Figure 7.1c shows how the solenoid impedance varies with the ratio of the radius of the bar to the eddy-current depth of penetration (or skin depth). Here, following the usual custom, the depth of the penetration is that depth in the plane wave, semi-infinite conductor case at which the eddy current density has decreased to $\frac{1}{e}$ times its value at the surface.

THERMAL METHODS

Transient Infrared Technique

J. D. Jensen

Work continued this quarter on the remote infrared transient method

for obtaining surface thermal-impedance values for a wide range of materials. Work has continued on overcoming the malfunctioning sample and hold circuit in the $f(t)$ simulation system. The present circuit is being severely loaded by successive stages and, in addition, the driver stage ahead of the sample and hold circuit is causing premature discharge of the storage capacitor. A search of manufacturers' literature has resulted in location of a commercial sample-and-hold module which might be used as a direct replacement for the malfunctioning circuit.

A 20 W carbon dioxide laser is now available in the laboratory and is being evaluated for use as a test-specimen excitation source. The laser emits at a wavelength of 10.6μ , in the far infrared. It operates in the continuous wave mode and generates a 0.25 in. diam beam. To adapt the laser for use with the transient test system, a shutter must be used to generate a short pulse as its output rather than the continuous wave output. Initial tests, with a commercial camera shutter operating at 1/400th of a second, resulted in partial destruction of the shutter vanes caused by intense local heating by the laser beam. The shutter vanes were removed and gold-plated to increase their reflectivity to the 10.6μ infrared emitted by the laser. In this way, it is hoped that further destruction of the shutter vanes will be avoided.

An investigation of a refracting optical system to diverge and collimate the 0.25 in. diam laser beam has

shown that design and fabrication of a suitable lens system would be difficult and expensive. Lenses fabricated from Irtran-II would cost approximately \$5,000, with germanium lenses about twice as expensive. As a result, reflecting optics are now being considered as an alternate approach. It is possible to diverge and collimate the beam by directing it against a small reflecting hemisphere located at the focus of a parabolic reflector. The beam would pass through a small hole in the center of the parabola and would be reflected by the hemisphere. The emerging beam from the face of the parabolic reflector would then have the desired 4 in. diam required to irradiate the entire sample surface uniformly, and would be reasonably well collimated.

Failure of the laser exciter has curtailed further experiments with the laser for the present time. The repaired exciter will probably be returned to the laboratory early in October, and further experiments will begin at that time.

Work began this quarter on an interim progress report on the transient system covering work performed on the system from August 1966 to the present time. A draft of a journal article on the system has been completed and will be submitted to the Journal of Applied Physics for publication.

Thermal Transducer Studies

D. R. Green

A thermal transducer system for nondestructively testing metals, graphite, ceramics, and composites has been developed. The thermal transducer method is completely independent of sample surface emittance (or emissivity) and has a number of potential practical applications such as nondestructively determining thermal uniformity in high-performance fuel elements, measuring thermal properties, and testing composite materials. Tests last quarter demonstrated the feasibility of making accurate thermal property measurements with the system by using a plane transducer at low frequencies. Operating experience gained in these tests also showed that higher frequency operation is needed not only to test small samples, but also to speed up measurements of thermal properties; measurements using the present transducer design are more time consuming than is desirable for routine measurement of thermal properties. In addition, cylindrical samples are considered prime candidates for practical applications of thermal-wave testing. Future work is being aimed at development of high-frequency and cylindrical transducers. To enhance the research and development aimed at specific areas of application, the

required personnel were transferred to the Nondestructive Testing Methods Development Section. Attempts to produce a high-frequency transducer have been partially successful, and development is continuing with emphasis on transducers that can be used to perform tests on samples of practical importance. High-speed scanning methods are being investigated.

An initial attempt to make thermal measurements at high frequencies was carried out by using the old-style rubber-backed transducer with high-amplitude oscillation in the input power. This was not successful and resulted in failure of the thermistor in the transducer.

A newly designed transducer, using "Hysol Epoxi-Patch 1C-White" epoxy resin to bond the exciter foil to the cold plate, was found to respond to frequencies up to about 20 Hz. Extended testing of this transducer to determine whether the bond will withstand long term thermal cycling has not yet been completed.

The smallest commercially made thermistors are too large to use in high frequency transducers. For this reason, a method using the self-resistance of the transducer foil as its own temperature monitor is being investigated. Thus far, difficulties with noise have prevented optimum results; however, shielding and filtering should eliminate this problem.

Coupling high-frequency transducers to samples requires a thin couplant layer to prevent exces-

sive attenuation of the thermal wave. Solid materials acting as an electrical insulator between the sample and transducer have been used in conjunction with gas and liquid couplants. A solid material normally used for coating cookware was tried, but was not adherent enough and finally flaked off. Glyptal was found to have satisfactory wear resistance, thermal and electrical properties, and adherence and is now being used as the insulating material.

Development of the high-frequency transducer system for application to small, cylindrical fuel pins is continuing. The first transducers will be plane high-frequency units fabricated to develop the required techniques, and to investigate high-speed scanning methods. Cylindrical transducers will then be developed for testing practical test specimens such as LMFBR fuel pins.

ULTRASONIC METHODS

Detection of Irradiation-Induced Shift in Nil-Ductility-Transition Temperature

D. O. Hunter

Ultrasonic shear velocity, longitudinal velocity, and critical angle signal response measurements have been made on irradiated A-302-B, A-542-B, and 304 SS, with neutron fluence as the independent variable and temperature of irradiation as a parameter. The similarity of the curves of the three ultrasonic measurements indicates that

changes in velocity are responsible for the critical-angle signal response.

Some additional measurements were taken during the first quarter. Most of these were intended primarily to help establish error caused by scatter in the material and by measurement methods. Other measurements were taken on a few irradiated specimens to determine if previously taken anomalous data points were valid. In addition, critical angle measurements were taken at the center position of all four sides of each irradiated and each control specimen. Previously reported critical angle measurements data points were the average of the integrated readings over about 1 in. length on each of the two sides. Although the new data have not yet been analyzed, they are expected to help determine error caused by scatter and, also, to provide a more valid comparison to velocity measurements taken at the center of the specimen.

Change-in-longitudinal-velocity measurements were made on eight 304 SS miniature tensile specimens irradiated at the EBR-II in a hard flux spectrum. Exposure range is estimated to be from about 3×10^{20} to 1.1×10^{22} nvt >1 MeV, with irradiation temperature estimated at 900 to 1000 °F. Because of small size and unfavorable geometry of the specimens, neither shear-velocity nor critical-angle measurements were possible. Results of the longitudinal velocity measurements must be accepted only with considerable reservation since ΔV_L was determined

with respect to only one unirradiated control specimen which had not been heat treated to correspond to the in-reactor heat treatment. Also, it was previously found that ultrasonic properties scatter in 304 SS can be exceptionally severe. However, results indicate that velocity increases with increasing fluence. With three anomalous data points out of eight measurements, V_L increased from

$$\frac{\Delta V_L}{V_L} = -0.327\% \text{ to } +0.089\%$$

as fluence increased. The slope of the resultant curve is approximately twice that of $\Delta V_L/V_L$ taken on 304 SS specimens irradiated at a lower temperature T , where $550 < T < 800$ °F, and in the opposite direction.

A topical report about measurements on irradiated low alloy steels is now being prepared.

Lithium Sulfate Transducer

N. L. Johnson

High-resolution transducers have been fabricated in the transducer laboratory by using lithium sulfate transducer elements. Transducers in the range of 40 to 90 MHz fundamental frequency have been assembled and tested.

To make the 40 to 90 MHz transducers, one must be able to monitor the thickness of the crystal as it is thinned. This is done by using a microscope and focusing on the front and back surface of the crystal and then measuring the distance between the focal points.

Because the transducers have electrodes of silver paint, it has been difficult to obtain a thin, even, conductive surface.

Another problem is the sealing of the lithium sulfate crystal from the water, which is necessary because it dissolves in water.

The crystal is first thinned and then attached by a silver paste to a tungsten-loaded epoxy backing member. If a thin vapor-deposited electrode could be used, a ringing of the transducer would probably be a decrease. Also, the pulse would be cleaner because the 1 to 2 mil thick layer of silver would not be present to cause interfering reflections. Vapor-deposited aluminum, gold on nickel have been deposited on lithium sulfate for the back electrode. The crystal is then attached to the backing member by uncured tungsten-loaded epoxy.

Humiseal has been used, but the layer is usually too thick and not uniform, and thus results in a distorted ultrasonic beam because of random interfering reflections. It is hoped that the humiseal layer can be eliminated by using vapor-deposited silicon monoxide or silicon dioxide to seal the lithium sulfate crystal.

To date, some of the transducers have been able to determine the thickness of 3 mil steel and 1 mil brass.

Wave Propagation Studies

F. L. Becker

The distortion of ultrasonic pulses caused by attenuation has been investigated by C. E. Fitch and R. L. Richardson.⁽⁵⁾ The wave model and computer program that they developed was not entirely successful in predicting pulse shape changes due to attenuation. The goal of this investigation is to evaluate present capabilities and limitations of this model and to define the problem areas. In addition, new methods of analysis and testing will be employed as well as extending the measurements to new materials.

The first step in this study was to measure the attenuation versus frequency curves for each material. The method of Reference (6) was used to obtain diffraction corrected attenuation measurements. Results of these tests are shown in Figures 7.2 and 7.3 for 304 SS and aluminum, respectively. The straight line through the data points is the least squares fit which is used in the calculations that will follow. The results shown in Figure 7.2 agree closely with those measured in Reference (6).

Three basic differences in technique were employed in this study:

- Both attenuation and pulse distortion measurements were accomplished at the same incident angle and position on the sample. This was done to eliminate possi-

FIGURE 7.2. Diffraction Corrected Attenuation Measurements for 304 SS

FIGURE 7.3. Diffraction Corrected Attenuation Measurements for Aluminum

ble effects caused by localized material anisotropies.

- The high attenuation and steep slope of the stainless steel curves result in drastic pulse shape changes. A more realistic test of the systems capabilities could be accomplished by using a material with less attenuation, thinner specimens or lower frequencies.
- Test signals of a more narrow bandwidth were used. Very broad signals have an irregular power spectrum; whereas, the more narrow band types have a bell shaped nature and are less difficult to analyze mathematically.

To date, two cases have been investigated: stainless steel at 15 MHz and aluminum at 8 MHz. Procedure and apparatus similar to that in Reference (5) were employed. In the first case, the pulse through quartz was chosen to give the same diffraction as the sample⁽⁶⁾ (Figure 7.4). This pulse was then digitized in increments of 5 nsec and analyzed by the "waves" program.⁽⁵⁾ This program fits this information to a 25-term Fourier series, and its real, imaginary and power spectrums are calculated. By knowing the attenuation in quartz and the water couplant, the shape of the signal at the transmitter is then calculated. The attenuations (Figure 7.2) are then used to calculate the shape of the pulse through the specimen. The received pulse and the calculated values are shown in Figure 7.5. The peak value is within 3% of the measured value while the first negative cycle is considerably less accurate.

This measurement represents only a slight improvement over previous measurements.

Power spectra of the pulse through quartz and stainless steel are shown in Figure 7.6. It is easy to see that very little of the original signal remains after passing through 1 in. of stainless steel. This in turn makes the measurement extremely sensitive to noise as the low frequency portion remaining was only slightly above the noise level.

The second case studied is that of a pulse with a center frequency of 8 MHz through 1 in. of aluminum. The same procedure as before is employed. Measured and calculated signals are shown in Figure 7.7; the correspondence is almost exact. Accuracy of the vertical scale is 0.09 divisions, and only the first negative and sixth positive peak exceed this limit. The power spectra of the pulse through quartz and aluminum is presented in Figure 7.8

From these examples, it is clear that the model and computer program is extremely accurate where attenuation is not too great. In cases of higher attenuation, further refinement is needed to improve the accuracy of the system. One such refinement is the possibility of using two or more Fourier series to represent the signal rather than one. This is a likely choice as broadband signals appear to be a composite of two or more signals. Further testing of the model and development of composite pulse analysis will be conducted during the next quarter.

FIGURE 7.4. Pulse Through Quartz
Center Frequency 15 MHz

FIGURE 7.5. Pulse Through 1 in. of
Stainless Steel Measured and Calculated Values

FIGURE 7.6. Relative Power Spectra of Pulse Through Quartz and Stainless Steel

FIGURE 7.7. Measured and Calculated Pulse Through 1 in. of Aluminum

FIGURE 7.8. Relative Power Spectra of Pulse Through Quartz and Aluminum

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METALLIC FUELS

J. C. Tverberg

WHITESHELL EXPERIMENTAL FUEL
ASSEMBLIES

W. E. Gurwell

Certain properties of metallic uranium make it an attractive fuel material, especially in organic-cooled reactors where the rupture of an unalloyed uranium fuel element is much less serious than in a water-cooled reactor. Metallic uranium offers many advantages over nonmetallic loadings such as high density, high thermal conductivity, and low fabrication costs.

Personnel at the Pacific Northwest Laboratory recently completed fabrication of metallic uranium fuel rods designed to eliminate fuel rod swelling by incorporation of a void in the fuel material. Techniques for fabricating small axial holes as small as 1/8 in. diam within coextruded fuel rods were demonstrated to be economically feasible. Another procedure for fuel fabrication included the use of metallic uranium pellets with small axial holes produced by extrusion or pellets with end dishes.

The purpose of this irradiation test is to develop an optimum fuel design for operation with fuel center temperatures in the 500 to 700 °C range and to average exposures of 7000 MW/T. Experience with hollow-core rods in the ETR at the low temperature and high

pressures of a PWR has shown that these fuel rods can easily exceed burnups of 15,000 MW/T with very small and usually slightly negative volume changes.

This irradiation test will examine the effectiveness of various combinations of void volume, cladding thickness, and void geometry in reducing diametral growth of the fuel rods in the higher temperature and lower pressure environment of an organic-cooled reactor. The performance of bonded and unbonded clad fuels also will be compared. These fuels will be irradiated in the WR-1 Reactors at the Whiteshell Nuclear Research Establishment. The irradiation test will begin shortly after the approval for shipping is received from the Division of International Affairs.

Fuel Design

The twelve basic fuel permutations are listed in Table 8.1. All rods have identical exterior dimensions. So all rods would have the same central temperature, the fuel enrichment was required to be 2.1, 2.3, and 2.54% for void volumes of 0, 5, and 10%, respectively.

All coextruded fuels have axial holes while pellets in some of the unbonded elements also have axial holes. The remaining unbonded elements use pellets with conical

TABLE 8.1. Twelve Uranium Alloy Fuel Permutations

<u>Bonded - Coextruded</u>				
<u>Clad,</u> <u>in.</u>	<u>Void Volume, %</u>	<u>Axial Hole</u> <u>Diameter, in.</u>		
0.020	0	None		
0.020	5	0.125		
0.020	10	0.177		
0.035	0	None		
0.035	5	0.119		
0.035	10	0.168		
 <u>Unbonded - Pellet</u>				
<u>Clad,</u> <u>in.</u>	<u>Void Volume, %</u>	<u>Conical End Dishes</u> <u>Diam</u> <u>Depth</u>		<u>Axial Hole</u> <u>Diameter, in.</u>
0.020	0	None	None	None
0.020	5	0.451	0.091	0.125
0.020	10	0.451	0.182	0.177
0.035	0	None	None	None
0.035	5	0.434	0.087	0.119
0.035	10	0.434	0.175	0.168

end dishes as voids. Figure 8.1 shows the different void geometries in the pellets, as well as tubular and end-cap components, and a completed fuel rod.

The fuel alloy is adjusted uranium containing 600 ppm carbon, 350 ppm iron, 800 ppm aluminum, 120 ppm silicon. This alloy has

been shown to have good swelling resistance.

The cladding alloy is Ozhenite 0.5, a zirconium base alloy having a low hydriding rate. Its composition is: 0.2% tin, 0.1% iron, 0.1% niobium, 0.1% nickel and the balance zirconium.

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FIGURE 8.1. Mock-Up of Pellet Fuel Showing Tube and End Caps Pellets (both Axial hole and conical dished ends finished autoclaved element)

BASIC SWELLING STUDIES

J. C. Tverberg

U₃Si U-Pu Th-U AND URANIUM METAL
SWELLING STUDIESC. R. Hann, R. K. Marshall, and
B. Mastel

Irradiation of fissionable materials causes a number of changes in the ordered metallic structure. High-energy neutrons and fission fragments knock the metal atoms out of their lattice positions leaving vacancies, interstitials, and dislocations. The purpose of this program is to provide a systematic investigation of the influence of irradiation on the specimen temperature, burnup, burnup rate, and external pressure as a function of such metallurgical variables as composition, structure, and geometry. To date data from this program indicate that swelling in alpha uranium is caused by mechanical processes associated with the irradiation process and from fission event defects rather than the agglomeration of fission gas pores. A slight amount of alloying element or application of external pressure has been found effective in overcoming this mechanical type of swelling.

Materials under evaluation at the present time are all metallic. These materials include unalloyed uranium, dilutely alloyed uranium, and thorium uranium alloys. During this quarter, two additional materials have been added; namely, the uranium-plutonium-zirconium alloys and uranium silicide (U₃Si).

The former is considered a potential fast-reactor fuel, while uranium silicides show potential for the Canadian Power Reactor Program.

The fundamental information obtained in this program will provide the fuel designer with a more accurate basis for fuel design and will allow a better understanding of the fission-induced swelling behavior.

Irradiation Program

Two tandem assemblies, one containing the three temperature-pressure controlled capsules P-12, P-13, P-17 and the other containing the capsules P-19, P-20, are now operating in KW reactor. These capsules contain high-purity and dilutely alloyed uranium specimens with three different ²³⁵U levels; two uranium-silicide (U₃Si) alloys; and three thorium-uranium alloys enriched with 93% ²³⁵U. A single capsule, P-21, is operating at 450 °C and 1000 psi in KE reactor. Capsules P-22 and P-23, to operate at 550 °C and 1000 psi and 625 °C and 1000 psi, respectively, are at KW reactor awaiting charging. Capsules P-21, P-22, and P-23 contain specimens of uranium, U-5 Pu, and U-14 Pu-12 Zr in the as-cast and heat-treated condition, as well as specimens of U₃Si. These irradiation capsules will provide data needed to evaluate:

- The effects of alpha, beta, and gamma phase temperatures in uranium base alloys.
- Hydrostatic pressure.
- Burnup rate.
- The effect of minor alloying additions on the irradiation behavior of uranium, uranium-plutonium and thorium-uranium alloys.

The status of these capsules is indicated in Table 9.1.

Postirradiation Examination

Three controlled temperature-pressure capsules, P-11, P-16, and

P-18, were opened in the radio-metallurgy facility, with the specimens recovered for density measurements and metallographic analysis. Table 9.2 gives the composition of the alloys irradiated in these capsules. Metallurgical histories, irradiation parameters, chemical compositions, and measured volume changes for each capsule are summarized in Tables 9.3, 9.4, and 9.5. Irradiation temperatures are given as average temperatures, and the error limits represent the range of occasional temperature fluctuations caused by flux perturbations and changes in reactor power levels.

TABLE 9.1. Capsule Status

Capsule Number	Control Temp, °C	Control Pressure, psi	Goal ^(a) BU, at.%	Status
P-11	550	5000	0.2 - 0.8	Under examination in Radiomet.
P-12	625	5000	0.2 - 0.8	Under irradiation
P-13	700	5000	0.2 - 0.8	Under irradiation
P-14	700	5000	0.2 - 0.8	Under examination in Radiomet.
P-16	625	1000	0.35 - 0.7	Under examination in Radiomet.
P-17	825	5000	0.2 - 0.8	Under irradiation
P-18	825	50	0.1 - 0.7	Under examination in Radiomet.
P-19	825	50	0.2 - 1.5	Under irradiation
P-20	725	50	0.2 - 1.5	Under irradiation
P-21	450	1000	0.1 - 0.2	Under irradiation
P-22	550	1000	0.1 - 0.2	Ready to charge
P-23	625	1000	0.1 - 0.2	Ready to charge

(a) Different burnups are achieved in a single capsule by including specimens of various enrichments.

TABLE 9.2. Compositions of Specimens in Swelling Capsules P-11, P-16, and P-18

Material	Alloy Designation	Composition (additions in ppm)
High-Purity Uranium	HiP	U+64Fe-25Al-25Si-6C
High-Purity Uranium	E-1	U+98Fe-11Al-34Si-56C
High-Purity Uranium	E-2	U+109Fe-30Al-36Si-53C
Dilute-Uranium Alloys	U+Fe-Si	U+140Fe-25Al-95Si-400C
Dilute-Uranium Alloys	U+Fe-Al-Si	U+407Fe-645Al-95Si-500C
Dilute-Uranium Alloys	WEB	U+393Fe-807Al-32Si-402C
Thorium-Uranium Alloys	Th-1 wt% U	Analysis Incomplete
Thorium-Uranium Alloys	Th-3.85 wt% U	Analysis Incomplete
Thorium-Uranium Alloys	Th-5.4 wt% U	Analysis Incomplete

The specimen number is coded so that the first number refers to the capsule. The letter in the specimen number refers to the position in the capsule, and the number following the letter refers to either half 1 or half 2 of the specimen pair at a particular position. All

specimens were identical in geometry, i.e., a split tube, and each material was represented in several metallurgical states. However, no specimen represents an optimum heat treatment with regard to swelling resistance.

TABLE 9.3. Irradiation History and Swelling of Specimens in Capsule P-11 Irradiated at 550 °C and 5000 psi

Sample	Alloy Designation (a)	Metallurgical Condition (b)	235U%	Irr. Temp., °C		at.%	Burnup ^(c) fiss/cm ³	As-Irr. Density, g/cm ³	% Swell., $\frac{p_i - p_f}{p_i} \times 100$	"R" % Swell., at.% BU
				Up	Down					
P11-A1	E-2	AE	2.88	480 $\pm$ $\frac{20}{10}$	240 $\pm$ 5	0.99	4.76 x 10 ²⁰	18.08	5.3	5.3
P11-A2	E-2	BQ	2.88	480 $\pm$ $\frac{20}{10}$	240 $\pm$ 5	0.99	4.76 x 10 ²⁰	17.71	7.6	7.7
P11-B1	Fe-Si	AE	0.72	530 $\pm$ 5	310 $\pm$ 5	0.27	1.30 x 10 ²⁰	18.63	1.8	6.7
P11-B2	Fe-Si	BQ	0.72	530 $\pm$ 5	310 $\pm$ 5	0.27	1.30 x 10 ²⁰	18.57	2.2	8.0
P11-C1	HiP	AE	0.72	550 $\pm$ 2	350 $\pm$ 2	0.27	1.30 x 10 ²⁰	18.46	3.3	12.0
P11-C2	HiP	BQ	0.72	550 $\pm$ 2	350 $\pm$ 2	0.27	1.30 x 10 ²⁰	18.54	2.8	10.4
P11-D1	Fe-Al-Si	AE	0.72	535 $\pm$ $\frac{5}{10}$	325 $\pm$ 5	0.27	1.30 x 10 ²⁰	18.64	1.3	4.8
P11-D2	Fe-Al-Si	BQ	0.72	535 $\pm$ $\frac{5}{10}$	325 $\pm$ 5	0.27	1.30 x 10 ²⁰	18.53	1.7	6.4
P11-E1	WEB	BQ	1.47	575 $\pm$ 5	315 $\pm$ 5	0.54	2.59 x 10 ²⁰	18.48	2.0	3.7
P11-E2	WEB	BQ	1.47	575 $\pm$ 5	315 $\pm$ 5	0.54	2.59 x 10 ²⁰	18.50	2.0	3.7
P11-F1	E-1	AE	1.44	530 $\pm$ 10	270 $\pm$ 5	0.52	2.50 x 10 ²⁰	17.99	5.8	11.2
P11-F2	E-1	BQ	1.44	530 $\pm$ 10	270 $\pm$ 5	0.52	2.50 x 10 ²⁰	18.26	4.2	8.1

(a) See Table 9.3 for compositions

(b) AE: As extruded
BQ: 730 °C, 15 min, oil quench

(c) Based on reactor data

Capsule P-11 was the second capsule in a series designed to irradiate fissile material under controlled hydrostatic pressures of 5000 psi. Irradiation temperatures ranged from 480 to 575 °C which placed specimens near the maximum on the "R" versus temperature curve described by Leggett, et al.⁽¹⁾ A range of burnups, 0.27 to 0.99 at.% was investigated by using samples of different enrichment. The notches and

holes apparent in the macroviews of the specimens in Figure 9.1a and 9.1b are identification marks cut prior to irradiation. Most of the specimens, with the exception of P11-A2, P11-C2, P11-E1, and P11-F2, were in very good condition considering the relatively high burnups in the temperature range where uranium is susceptible to fission-event-induced volume changes rather than fission-gas swelling.

**TABLE 9.4. Irradiation History and Swelling of Specimens
in Capsule P-16 Irradiated at 625 °C and 1000 psi**

Sample	Alloy Designation (a)	Metallurgical Condition (b)	235U%	Irr. Temp., °C		at.%	Burnup (c) fiss/cm ³	As-Irr. Density, g/cm ³	% Swell. pi-pf pf x 100	"R" % Swell. at.% BU
				Up	Down					
P18-A1	Th-5.4 U	A	93	780 ± 15 20	415 ± 5	1.03	3.19 x 10 ²⁰	10.70	12.5	12.1
P18-A2	E-2	AE	2.88	780 ± 15 20	415 ± 5	0.59	2.84 x 10 ²⁰	12.56	52	88
P18-B1	Th-1U	B	93	785 ± 10	470 ± 5	0.19	0.57 x 10 ²⁰	11.59 ?	0.4 ?	2.1 ?
P18-B2	Th-1U	C	93	785 ± 10	470 ± 5	0.19	0.57 x 10 ²⁰	11.32 ?	2.9 ?	15.3 ?
P18-C1	HiP	AE	0.72	825 ± 5	500 ± 5	0.16	0.77 x 10 ²⁰	17.33 ?	9.9 ?	62 ?
P18-C2	HiP	BQ	0.72	825 ± 5	500 ± 5	0.16	0.77 x 10 ²⁰	17.53 ?	8.7 ?	54 ?
P18-D1	Fe-Si	AE	0.72	835	500	0.16	0.77 x 10 ²⁰	17.17 ?	10.5 ?	66 ?
P18-D2	Fe-Al-Si	AE	0.72	835	500	0.16	0.77 x 10 ²⁰	16.68 ?	11.9 ?	74 ?
P18-E1	E-1	AE	1.44	880 ± 10 30	480 ± 5 10	0.31	1.49 x 10 ²⁰	11.10 ?	71 ?	230 ?
P18-E2	WEB	BQ	1.47	880 ± 10 30	480 ± 5 10	0.32	1.54 x 10 ²⁰	10.16 ?	86 ?	270 ?
P18-F1	Th-3.85U	D	93	830 ± 10 20	440 ± 5	0.74	2.27 x 10 ²⁰	10.92	8.2	11.1
P18-F2	Th-3.85U	C	93	830 ± 10 20	440 ± 5	0.74	2.27 x 10 ²⁰	10.94	8.1	10.9

(a) See Table 9.2 for compositions

(b) A: Extruded and cold swaged
B: Hot rolled, cold rolled, and cold swaged
C: 850 °C, 2 hr, FC
D: Hot rolled and cold swaged
AE: As extruded
BQ: 730 °C, 15 min, oil quench

(c) Based on reactor data

**TABLE 9.5. Irradiation History and Swelling of Specimens
in Capsule P-18 Irradiated at 825 °C and 50 psi**

Sample	Alloy Designation (a)	Metallurgical Condition (b)	235U%	Irr. Temp., °C		at.%	Burnup (c) fiss/cm ³	As-Irr. Density, g/cm ³	% Swell., pi-pf pf x 100	"R" % Swell., at.% BU
				Up	Down					
P16-A1	E-1	AE	1.44	~565	~335	0.63	3.03 x 10 ²⁰	14.77	28.9	46
P16-A2	E-1	BQ	1.44	~565	~335	0.63	3.03 x 10 ²⁰	14.43	32.0	51
P16-B1	E-1	AE	1.44	~615	~390	0.63	3.03 x 10 ²⁰	17.09	11.4	18.1
P16-B2	E-1	BQ	1.44	~615	~390	0.63	3.03 x 10 ²⁰	16.76	13.7	21.8
P16-C1	Fe-Al-Si	AE	0.72	630 ± 10	395 ± 5	0.33	1.59 x 10 ²⁰	18.44	2.2	6.7
P16-C2	Fe-Al-Si	BQ	0.72	630 ± 10	395 ± 5	0.33	1.59 x 10 ²⁰	18.44	2.3	7.0
P16-D1	WEB	AE	1.47	622 ± 5	400 ± 5	0.65	3.12 x 10 ²⁰	18.22 ^(d) ?	3.6 ?	5.5 ?
P16-D2	WEB	BQ	1.47	622 ± 5	400 ± 5	0.65	3.12 x 10 ²⁰	18.04	4.7	7.2
P16-E1	Fe-Al-Si	AE	0.72	600 ± 10 5	362 ± 5	0.33	1.59 x 10 ²⁰	18.50	1.8	5.5
P16-E2	Fe-Al-Si	BQ	0.72	600 ± 10 5	362 ± 5	0.33	1.59 x 10 ²⁰	18.52	1.8	5.5
P16-F1	WEB	AE	1.47	605 ± 10	340 ± 5	0.65	3.12 x 10 ²⁰	18.29 ^(d)	3.2	4.9
P16-F2	WEB	BQ	1.47	605 ± 10	340 ± 5	0.65	3.12 x 10 ²⁰	18.18	3.7	5.7

(a) See Table 9.2 for compositions

(b) AE: As extruded
BQ: 730 °C, 15 min, oil quench

(c) Based on reactor data

(d) Density of largest piece of sample

P11-A1 High-Purity
Uranium 2.88% ^{235}U
0.99 at.% BU, 480 °C,
 $\Delta V/V = 5.3\%$

P11-A2 High-Purity
Uranium 2.88% ^{235}U
0.99 at.% BU, 480 °C,
 $\Delta V/V = 7.6\%$

P11-B1 U+Fe-Si
0.72% ^{235}U 0.27
at.% BU, 530 °C,
 $\Delta V/V = 1.8\%$

P11-B2 U+Fe-Si
0.72% ^{235}U 0.27
at.% BU, 530 °C,
 $\Delta V/V = 2.2\%$

P11-C1 High-Purity
Uranium 0.72% ^{235}U
0.27 at.% BU, 550 °C,
 $\Delta V/V = 3.3\%$

P11-C2 High-Purity
Uranium 0.72% ^{235}U
0.27 at.% BU, 550 °C,
 $\Delta V/V = 2.8\%$

Outside View

End View

Inside View

E6199-E6215

2X

FIGURE 9.1a Macro-appearance of Specimens Irradiated
in Capsule P-11

FIGURE 9.1b Macro-appearance of Specimens Irradiated in Capsule P-11

The severe distortion of Specimen P11-A2, high-purity uranium, is attributed to the irradiation temperature, preirradiation heat treatment, and the high burnup. Irradiation growth rates in alpha uranium increase with a decrease in irradiation temperature. Consequently, Specimen P11-A2 shows greater distortion because it was irradiated at a lower temperature (480^{+20}_{-10} °C) than other specimens located in other positions within the capsule. The heat treatment--730 °C, 15 min, oil quench--produced very large grains which contributed to the surface roughening and the general distortion. In comparison, Specimen P11-A1 was in a fine-grained as-extruded condition and did not show as much surface roughening and warpage.

Although Specimen P11-C2, high-purity uranium, received the same heat treatment as P11-A2, the distortion is less severe because irradiation growth rates are less at 550 °C and the burnup is much lower than that for Specimen P11-A2. The same argument used to explain distortion in Specimen P11-C2 applies to Specimen P11-F2. The cause of the severe distortion on one end of the P11-F2 is not clear at this time. Specimen P11-E1 was severely cracked while Specimen P11-D2, which had a similar composition, contained only a few cracks. The different amount of cracking could be caused by differences in burnup, composition, or fabrication techniques. Probable contributing factors to the cracking are the large grain size and the dilute-alloy additions. Presently,

it is not known whether the cracks formed during irradiation or during the cooling of the capsule to ambient temperature prior to discharge.

Density measurements indicate that (1) 5000 psi reduces swelling in high-purity uranium by a factor of at least 30 to 40 in the 480 to 575 °C temperature range; and (2) dilute alloy additions and 5000 psi pressures reduce the swelling to the theoretical minimum established by the contribution of solid fission-product atoms. The lower "R" values for the higher burnup samples could be the result of (1) in situ alloying effects from fission products produced during the irradiation, (2) minor differences in impurity content because the low burnup material had the lowest impurity content or (3) the assumption that volume changes increase linearly with burnup could be invalid at high burnups.

Capsule P-16 contained high-purity uranium and uranium with dilute additions of iron, aluminum, silicon, and carbon. Irradiation temperatures ranged from 565 to 630 °C while estimated burnups ranged from 0.33 to 0.65 at.%. Macro-photographs of each specimen are presented in Figures 9.2a and 9.2b. Visual examination revealed that 8 of the 12 specimens were in excellent condition with respect to external appearance. The surface roughening observed on Specimen P16-A2 is probably caused by a combination of irradiation growth and the large grain size obtained as a result of the preirradiation heat treatment.

Outside View

End View

Inside View

E6276-E6296

2X

FIGURE 9.2a Macro-appearance of Specimens Irradiated
in Capsule P-16

P16-D1 U+Fe-Al-Si
1.47% ^{235}U 0.65
at.% BU, 620 °C,
 $\Delta V/V = 3.6\%$

P16-D2 U+Fe-Al-Si
1.47% ^{235}U 0.65
at.% BU, 620 °C,
 $\Delta V/V = 4.7\%$

P16-E1 U+Fe-Al-Si
0.72% ^{235}U 0.33
at.% BU, 600 °C,
 $\Delta V/V = 1.8\%$

P16-E2 U+Fe-Al-Si
0.72% ^{235}U 0.33
at.% BU, 600 °C,
 $\Delta V/V = 1.8\%$

Specimen Broke During Handling
No Photographs Available

P16-F1 U+Fe-Al-Si
1.47% ^{235}U 0.65
at.% BU, 605 °C,
 $\Delta V/V = 3.2\%$

P16-F2 U+Fe-Al-Si
1.47% ^{235}U 0.65
at.% BU, 605 °C,
 $\Delta V/V = 3.7\%$

Outside View

End View

Inside View

E6291-E6306

2X

FIGURE 9.2b Macro-appearance of Specimens Irradiated
in Capsule P-16

Specimen P16-B2 also had a large grain size, but the roughening probably was not caused by irradiation growth above 600 °C. Two specimens were broken during the macroexamination: Specimen P16-D1 was severely cracked at what appeared to be grain boundaries; Specimen P16-F1 cracked into two parts and was too brittle to handle without further damage. Both specimens were fabricated from the same materials, with densities determined on the largest portion of each specimen. The density value for P16-D1 is subject to considerable error because of the internal wetting problems associated with the cracks.

Volume changes and "R" values calculated from immersion density measurements and estimated burnups indicate that a pressure of 1000 psi reduces swelling by a factor of at least 4 to 5 in high-purity uranium irradiated at ~560 °C. Comparison of the 1000 psi data above 600 °C with data obtained from low-exposure irradiations at 30 psi indicates that "R" values are reduced by a factor of 2 for high-purity uranium. Alloy additions provide additional benefits to reduce "R" values another factor of 3 to 4 to approach the theoretical minimum. No evidence of "break away" swelling was observed.

Capsule P-18, which was controlled at 825 °C and 50 psi, contained five Th-U specimens, four high-purity uranium specimens, and three dilutely alloyed uranium specimens. The black oxide evident in the

macrophotographs (Figure 9.3a and 9.3b) developed in-cell after determination of the as-irradiated densities because the photography was delayed two days. A few light particles evident on some specimens are the result of a NaK-H₂O reaction that occurred during the initial stages of specimen recovery. The outer pressure shell of each capsule is normally removed with a large abrasive cut-off wheel partially immersed in water. The bellows on Capsule P-18 was accidentally severed by the wheel, and the water-NaK reaction resulted in a fire that was soon controlled. The NaK oxide was easily removed from most of the specimens; however, some specimens could not be completely cleaned without further damage.

The external appearances of the Th-U alloys were excellent. If the split tube geometry was to be used, the specimen wall thickness for the Th-U alloys had to be changed from that of the standard 0.030 in. thick uranium specimens to maintain compatible heat generation values. Accordingly, the wall thickness of the Th-U specimens varied from 0.023 in. for the Th-5.4 U alloy to 0.060 in. for the Th-1 U alloy. Density measurements and estimated burnups yielded "R" values of 2 to 15 with most of the values in the 10 to 15 range. The density values might be somewhat low because the samples are quite small (1 to 2 g), and the samples have a very large surface to volume

Outside View

End View

Inside View

E6357-E6372

2X

FIGURE 9.3a. Macro-appearance of Specimens Irradiated
in Capsule P-18

P18-D1 U+Fe-Si
0.72% ^{235}U 0.16
at.% BU, $\sim 835^\circ\text{C}$,
 $\Delta V/V = 10.5\%$

P18-D2 U+Fe-Al-Si
0.72% ^{235}U 0.16
at.% BU, $\sim 835^\circ\text{C}$,
 $\Delta V/V = 11.9\%$

P18-E1 High-Purity
Uranium 1.44% ^{235}U
0.31 at.% BU, $\sim 880^\circ\text{C}$,
 $\Delta V/V = 71\%$

P18-E2 U+Fe-Al-Si
1.47% ^{235}U 0.32
at.% BU, $\sim 880^\circ\text{C}$,
 $\Delta V/V = 86\%$

P18-F1 Thorium
3.85 wt% Uranium
93% ^{235}U 0.74
at.% BU, $\sim 830^\circ\text{C}$,
 $\Delta V/V = 8.2\%$

P18-F2 Thorium
3.85 wt% Uranium
93% ^{235}U 0.74
at.% BU, $\sim 830^\circ\text{C}$,
 $\Delta V/V = 8.2\%$

Outside View

End View

Inside View

E6374-E6390

2X

FIGURE 9.3b. Macro-appearance of Specimens Irradiated
in Capsule P-18

ratio. This large ratio makes the density determination quite sensitive to sample oxidation and adherent foreign material.

Four of the uranium specimens appear to be in reasonably good shape. The other three specimens operated to a higher burnup, and two of the three were irradiated at considerably high temperatures. One specimen of high-purity uranium, P18-A2, was initially operating at 795 °C until a change in reactor loading decreased the flux to a point where the specimen was operating partially in the beta phase and partially in the gamma phase. The portion that operated in the gamma phase was severely swollen while the beta phase portion appeared to be in relatively good condition. The gamma phase portion appears black in the "inside view" of Figure 9.4a. Two other specimens, P18-E1 and P18-E2, were somewhat distorted, and the surfaces appeared to be quite rough. These two specimens operated at ~880 °C to estimated burnups of

~0.3 at.%. Density measurements on P18-E1 and P18-E2 indicate that 70 to 85% volume increases with corresponding "R" values of 230 to 270. These values might be too large because of the rough sample surfaces and the attached crud from the NaK-H₂O reaction. The "R" values for the rest of the uranium specimens ranged from 55 to 90.

It would appear from these examinations that Th-U alloys offer a substantial improvement in irradiation performance over uranium at these elevated temperatures and burnups. These improvements are probably caused by the higher melting point (~1750 °C) of thorium. Optical and electron microscopy are being performed to verify volume changes and to clarify the primary modes of swelling.

REFERENCE

1. R. D. Leggett, T. K. Bierlein, B. Mastel, and H. A. Taylor. *Basic Swelling Studies*, BNWL-SA-154, Pacific Northwest Laboratory, Richland, Washington, September 1965. Also CONF-650904-Vol. 37, 1968.

ATR GAS LOOP OPERATION AND MAINTENANCE

E. A. Evans

ATR GAS LOOP SUPPORT

Problems associated with the Hastelloy-X material used in the Gas Loop regenerative heat exchangers were resolved. Discussions with consultants on the requirements of Section III of the ASME Code required replacement of some of the forgings because the direction of high

stresses coincided with the direction of the lowest tensile properties.

The analytical equipment for the Gas Loop is in the final check-out stage. PNL has had full responsibility for the inspection to determine compliance with the specifications. Shipment of the unit to the ATR site will be arranged during October.

FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS

R. E. Nightingale

EBR-II DOSIMETRYJ. L. Jackson, J. A. Ulseth,
and W. N. McElroy

A PNL-ANL cooperative dosimetry experiment has been prepared and is scheduled for irradiation in the EBR-II reactor during October 1968. The primary purposes of the experiment are to resolve the existing differences between calculated and measured fluxes and to provide accurate flux and fluence measurements in the EBR-II. The experiment has been designed to do the following:

1. Determine whether low-power flux measurements can be used to estimate full-power flux values.
 2. Measure the flux and spectral perturbations due to a structural subassembly.
 3. Derive the neutron flux at several axial and radial positions in EBR-II at full power from activation of a variety of foils (threshold and integrating).
 - Provide data from which the spectra at several axial and radial locations can be calculated and compared with physics calculations, using both transport and diffusion codes.
 - Establish a body of flux data for EBR-II that can be cross-correlated by ANL, PNL, and the major materials experimenters.
 4. Test the accuracy of different methods for measuring flux and fluence in fast reactors such as EBR-II.
- Determine the effects of self-shielding for (n, γ) monitors in a fast-reactor environment.
 - Test the stable-product fluence-monitor system based on the analysis of fission products ^{235}U , ^{238}U , and ^{237}Np , i.e. compare burnup (^{148}Nd and ^{137}Cs) results and radiochemistry (^{140}Ba and ^{99}Mo) results.
 - Test the stable-product fluence-monitor system based on the analysis for helium gas produced in a metal sample by the (n, α) reaction.

Several different types of monitor sets are required for the data needed to fulfill the objectives of the experiment. The different sets are as follows:

Spectral Sets

Spectral sets contain monitors for measuring flux, fluence, fission rate, and burnup. All of the monitors will provide data for calculating both the spectrum and the flux at the specific reactor position where the set is located. These sets will be located at several radial and axial positions. The spectral set assembly is shown in Figure 11.1, and details of materials and monitor reactions are listed in Table 11.1.

Gradient Sets

Gradient sets are placed between the spectral sets to provide axial

FIGURE 11.1. Assembled Monitor Container

TABLE 11.1. Spectral Sets

Reaction	Form	Dimensions (a)	Test (b)	Approx. Weight, mg	Number/Location	Encapsulated (c)
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	Metal-foil	5 mil	L	27	1	
	Metal-foil	0.5 mil	H	3	1	
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Metal-foil	0.5 mil	L	3	1	
	Metal-foil	0.5 mil	H	3	1	
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	Metal-wire	1 mil	L	2	1	
	Metal-wire	1 mil	H	2	1	
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	Metal-foil	0.05 mil	L	0.2	1	
	Metal-foil	0.05 mil	H	0.2	1	
$^{23}\text{Na}(n,\gamma)$	NaCl-Pellet	0.08 in. diam x 1/8 in. long	L & H	22	1	
$^{23}\text{Na}(n,2n)$						
$^{35}\text{Cl}(n,\alpha)$						
$^{54}\text{Fe}(n,p)$	Metal-wire	20 mil x 3 in. R	L	121	1	
$^{58}\text{Fe}(n,\gamma)$						
$^{54}\text{Fe}(n,p)$						
$^{58}\text{Fe}(n,\gamma)$	Metal-wire	20 mil x 1/2 in. R	H	18	1	
$^{58}\text{Ni}(n,p)$						
$^{58}\text{Ni}(n,p)$						
$^{58}\text{Ni}(n,2n)$	Metal-wire	20 mil x 1/2 in. R	H	22	1	
$^{27}\text{Al}(n,\alpha)$						
	Metal-wire	20 mil x 3 in. R	L	42	1	
	Metal-wire	20 mil x 3/16 in. S	H	3	1	X
$^{24}\text{Mg}(n,p)$	Metal-foil	5 mil	L	5	1	
	Metal-piece	5 mil x 0.080 in. x 1/8 in.	H	1.4	1	X
$^{46}\text{Ti}(n,p)$	Metal-wire	20 mil x 1/3 in. R	L	24	1	
$^{46}\text{Ti}(n,p)$	Metal-wire	20 mil x 1/2 in. R	H	12	1	
$^{47}\text{Ti}(n,p)$						
$^{48}\text{Ti}(n,p)$						
$^{127}\text{I}(n,2n)$	KI-Pellet	0.08 in. diam x 1/8 in.	L	31	1	X
	KI-Pellet	0.08 in. diam x 1/8 in.	H	31	1	X
$^{238}\text{U}(n,f)$ (d)	Metal-piece	1 mil x 1/8 in. x 5/8 in.	L	15	1	X
$^{238}\text{U}(n,\gamma)$ (g)						
$^{238}\text{U}(n,f)$	Metal-piece	1 mil x 1/8 in. x 5/8 in.	H	15	1	X
$^{238}\text{U}(n,\gamma)$ (g)						
$^{238}\text{U}(n,f)$ (e)	Metal-piece	1 mil x 1/8 in. x 3-3/4 in.	H	100	1	X
$^{235}\text{U}(n,f)$ (d)	Metal-piece	1 mil x 1/8 in. x 5/8 in.	L	15	1	X
$^{235}\text{U}(n,f)$ (d)	Metal-piece	1 mil x 1/8 in. x 5/8 in.	H	15	1	X
$^{235}\text{U}(n,f)$ (e)	Metal-piece	1 mil x 1/8 in. x 3/4 in.	H	20	1	X
$^{237}\text{Np}(n,f)$ (d)	Metal-piece	6 mil x 0.080 in. x 1/8 in.	L	20	1	X
$^{237}\text{Np}(n,f)$ (d)	Metal-piece	6 mil x 0.080 in. x 1/8 in.	H	20	1	X
$^{237}\text{Np}(n,f)$ (e)	Metal-piece	6 mil x 0.080 in. x 1/4 in.	H	40	1	X
$^{232}\text{Th}(n,f)$ (e)	Metal-piece	5 mil x 0.080 in. x 5/8 in.	L	80	1	X
$^{232}\text{Th}(n,f)$ (e)	Metal-piece	5 mil x 0.080 in. x 1/16 in.	H	9	1	X
$^{232}\text{Th}(n,f)$ (f)	Metal-piece	5 mil x 1/8 in. x 5/8 in.	H	80	1	

(a) Foils shall be a small flat washer ~1/4 in. OD x 1/8 in. ID; thickness will vary as noted. Wire shall be referenced by OD and approximate length with S (straight) and R (formed round).

(b) L refers to Low Power (50 kW) test.
H refers to High Power (50 MW) test.

(c) Check indicates material contained in stainless steel tubes ~3/4 in. long x 0.093 in. OD with ~3 mil wall, welded closed (Figure 3).

(d) Analyze for ^{140}Ba .

(e) Analyze for $^{137}\text{Cs}/^{148}\text{Nd}$. These will be included only in the spectral sets in Rows 4, 6, and 7 at midplane. One capsule containing three each of ^{235}U and ^{238}U will be in Row 4 below midplane only.

(f) Analyze for ^{137}Cs .

(g) Analyze for ^{239}Np .

flux-gradient information. Gradient monitors are 20 mil iron and nickel wires.

Experimenters' Monitors and ANL-PNL Correlation Sets

Experimenters' monitors consist of one or two fluence monitors from each experimenter's own stock of monitor material and will be limited to those types of monitors commonly used by the experimenter in his EBR-II experimental hardware. These monitors will provide an opportunity for all experimenters to correlate with the large body of data provided by the spectral and gradient sets.

ANL-PNL correlation sets consist of fission foils, iron, nickel, copper, and titanium wires. Special ANL sets for cross section studies are also included. These sets will be located close to reactor midplane in Rows 2, 4, 6, and 7 during the high-power experiment.

Self-Shielding Sets

Self-shielding sets contain different thicknesses of cobalt, copper, and scandium to determine the effect of monitor thickness on monitor activation. Sets of each type of material will be located in three axial positions in Row 2 to obtain self-shielding information in different spectral environments. The locations are approximately 3, 12, and 20 in. above midplane.

Stable-Product Sets

PNL stable-product sets contain materials such as aluminum, titanium, iron, nickel, and scandium that undergo an (n, α) reaction to produce helium. The helium produced is a measure of the integrated flux to which the monitor was exposed.

In addition, Atomics International has provided 30 pure elements and 23 crystalline compounds for analysis of helium content.

All of the stable-product materials will be analyzed for helium with a high-sensitivity mass spectrometer by Atomics International. The data obtained when correlated with the neutron flux spectral measurements will lead to estimates of the energy dependent (n, α) cross sections for the elements involved.

The complete experiment to irradiate the monitors described will consist of a low-power test (50 kW) and a high-power test (50 MW).

The low-power test monitors will be located axially only in reactor Row 2 (2B1), with one dosimetry capsule in subassembly Position 18 (Figure 11.2). Position 10 will contain a near nuclear duplicate of the capsule to be placed in position 10 during the full-power tests.

The full-power test monitors will be located in Rows 2, 4, 6, and 7 (Figure 11.2). Monitors will be located axially in the subassembly locations listed below.

FIGURE 11.2. EBR-II Loading Pattern

<u>Row</u>	<u>Reactor Location</u>	<u>Assembly Location</u>
2	2B1	18
2	2B1	10
4	4E2	1
6	6D2	8
7	7A3	8

The remainder of the subassembly will contain stainless-steel dummies.

The foil-holder assembly (Figure 11.1) contains from 15 to 20 separate monitor materials in a compact package that can be assembled easily and disassembled in a hot cell with minimum effort and time. Thin materials such as 0.00005 in. gold are cut into disks with a hole in the center and sandwiched between titanium spacers and held together for

separation outside the hot cell. To contain the fission products, fissionable monitor materials are enclosed in the small welded tubes (0.093 in. OD x 0.086 in. ID). Other materials that may melt, such as aluminum or magnesium, or may break, such as the NaCl and KI pellets, are also contained in the small welded tubes.

The disassembly procedure, including the use of a special holding jig, was checked out during a dry run in a hot cell. Remote disassembly of the monitor container required less than 5 min during the first practice run.

IRL PRESSURE-VESSEL SPECTRUM SIMULATION EXPERIMENT

J. A. Ulseth, W. N. McElroy,
and R. L. Simons

In addition to the PNL effort to determine the neutron spectrum in surveillance and pressure-vessel positions as previously reported,⁽¹⁾ Westinghouse will determine the neutron spectra at 18, 23, 34, 45, and 76 cm from the core edge. In effect, this will show how the neutron flux incident upon the pressure vessel is moderated by various thicknesses of water.

PNL will provide the foils, but Westinghouse will count the foils and analyze the subsequent data. Therefore, an intercalibration of PNL and Westinghouse counting facilities will be necessary.

Scheduling of the IRL test has been revised since the last quarterly progress report.⁽¹⁾ The 1 yr irradiation of fluence monitors, damage

samples, and Charpy-V notch specimens in the simulated pressure vessel will start in the latter part of September. The irradiations to define spectra and for intercalibration purposes will be performed at a later date.

DEVELOPMENT OF DAMAGE-CORRELATION PROCEDURES

W. N. McElroy, R. E. Dahl,
and J. L. Jackson

To assure satisfactory service of cladding and structural materials in the FTR, some method is needed for estimating the damage expected in FTR components during their service life. To do this requires the definition of the relationship in damaging potential between the fluence received during a test irradiation and the fluence expected at the service position. An effort is currently underway to empirically determine functions for relating fluence on an equivalent damage basis at different facilities and to experimentally check the validity of these functions.

The analytical work involves the use of damage unfolding techniques developed for the definition of cross sections (of fluence monitors) to determine a damage function. The damage function is the relationship between the energy of a neutron and its effectiveness in causing the property change of interest. By defining this damage function, one can perform the damage equivalent assessment for various spectra at constant temperatures. The following is a brief review of the plan to use the SAND-II code's iterative procedure to define damage functions.

The following equation can be used to represent a specified property change $S_{ij}(T,H)$ of the i^{th} material:

$$S_{ij}(T,H) = \iiint G_i(E,T,H,t) \times \phi_j(E,t) dE dt \quad (1)$$

where T is the temperature, H is a material specimen history parameter,* E is the neutron energy, t is the time from the start of the neutron irradiation, $\phi_j(E,t)$ is the neutron differential flux for the " j " environment, and $G_i(E,T,H,t)$ is the damage function that is to be determined by the SAND-II iterative procedure.

In the planned study, the following assumptions will be made:

- (1) G_i and ϕ_j can be represented by time-averaged values $\bar{G}_i$ and $\bar{\phi}_j$.
- (2) H can be absorbed into " i " for brevity of notation.

By using these assumptions and notation, Equation (1) is rewritten as

$$S_{ij}(T) = t \cdot \int \bar{G}_i(E,T) \bar{\phi}_j(E) dE. \quad (2)$$

For a set of material specimens irradiated within a specified temperature band about T , the calculated-measured** spectra will be used together with the measured values of $S_{ij}(T)$ as input to the SAND-II code

* Related to impurities, stress conditions, heat treatment before irradiation, testing procedure used to measure property change, etc.

** Multigroup results adjusted in magnitude by foil-monitor measurements.

to obtain $G_i(E,T)$ solutions [different input approximations, $G_i^{(0)}(E,T)$, will be used to define an envelope of solutions]. The width of this envelope for a given neutron energy region will define the degree to which the $\bar{\phi}_j(E)$'s were sufficiently different to define a reasonable solution for $G_i(E,T)$.

The above basic procedure has been used successfully in the evaluation of cross sections and the results are discussed in Reference 2. Whether the procedure will be successful for defining $G_i(E,T)$ will depend mainly on the degree of difference among the $\bar{\phi}_j(E)$'s.

Computer simulation work will provide initial estimates for the damage function. The computer simulation codes provide an estimate of the efficiency for damage production of neutrons of various energies. One can utilize the energy dependence of either the total number of defects or defects of a particular kind. Such initial estimates are necessary for the more efficient operation of the SAND-II code.

The K^{th} solution $G_i^{(K)}(E,T)$ from the SAND code, when compared with the initial $G_i^{(0)}(E,T)$, should guide further modification both of the computer simulation codes and of the SAND-II procedure. The final damage function will be defined by an iterative process involving modification of the initial damage function and the SAND-II procedure on the basis of the solution $G_i^{(K)}(E,T)$.

One set of suitable data for assessing the feasibility of the method has been received from the U. S.

Naval Research Laboratory,⁽³⁾ and another set of data are available in the literature.⁽⁴⁾ These two sets of data provide the irradiation-induced shifts in nil-ductility temperature for pressure-vessel steels. Data are from tests conducted in light- and heavy-water-moderated thermal test reactors.

These data are being used to test the concept of damage function unfolding. Preliminary results have been obtained by using an initial damage function based on a Kinchin and Pease relation for the number of direct neutron-induced displacements⁽⁵⁾ and a superimposed low-energy contribution based on the total number of recoil atoms from the (n, γ) reaction. For the low-energy region from 10^{-10} to $\sim 10^{-2}$ MeV, the damage function was assumed to be proportional to the total (n, γ) cross section for iron. The proportionality constant was eight displacements per (n, γ) reaction.^(4,6)

This initial damage model together with the experimentally determined values of fluence for a number of different spectra (that were used to produce a specified change in the nil-ductility temperature for A212 steel of $T = 250$ °F for the NRL's data and $T = 75$ °C for Rossin's) were used as input to the SAND-II code. Solutions were obtained that represent the neutron energy dependence of the property change, $S_{ij}(T)$, of shift in nil-ductility temperature. The dependence of the solutions on the initial approximation was tested by inputting such forms as a constant (between 10^{-2} and 18 MeV)

with an E power below 10^{-2} . For the cases studied, the solutions lead to increased damage effectiveness for thermal neutrons.

The results of these studies establish the feasibility of damage-function unfolding, and the preliminary results suggest that low-energy neutrons are more important in their contribution to damage than predicted by current damage models. Further analysis of these and other data will be required to definitely prove or disprove the validity of this latter conclusion. It is expected that the current studies will generate damage functions that can be used in assessing the problem of damage posed by the extremely soft spectrum present at the FTR pressure vessel.

COMPUTER SIMULATION OF CRYSTALLINE DEFECTS

D. G. Doran

The simulation of short-term annealing of displacement spikes in α -iron at temperatures near $0.5 T_m$ is being carried out with the computer code described previously;⁽¹⁾ however, the technique has been modified.

In the sample problem reported earlier⁽¹⁾ the interstitials, being the more mobile defect, were allowed to move for 100 time steps while the vacancies were held fixed. The vacancies were then allowed to migrate for up to 300 time steps while the interstitials were held fixed. The numbers of time steps in each mode of operation were based on the

attainment of a relatively stable cluster size distribution. Although admittedly crude, this procedure was tried in order to conserve computer time. The results were unsatisfactory, however, because the partial annihilation by the mobile vacancies of small interstitial clusters left more mobile interstitials, i.e., singles and pairs, than had been anticipated. These newly mobile defects were produced in the vicinity of vacancy clusters and, hence, should have been given the opportunity to either annihilate vacancies, cluster, or migrate unscathed away from the action.

The procedure has been modified so that the separate operating modes for vacancies and interstitials have been retained, but the modes have been operated alternately keeping the ratio of interstitial time steps to vacancy time steps at 100 to 1. The initial sequence is in fact 100 interstitial time steps alternated with a single vacancy time step. Eventually, the interstitials become either clustered (hence, immobile) or widely dispersed, and the frequency of interactions decreases drastically so the numbers are raised to, say, 1000 and 10. When all the interstitials are either clustered or essentially removed to infinity, the vacancies are allowed to migrate for many time steps (perhaps an order of magnitude increase in the number of vacancy time steps) which lets them assume an "ultimate" cluster size distribution. It is significant that the spatial distributions of interstitials and vacancies become

sufficiently separated to make this a reasonable procedure.

The 20 keV, α -iron spike annealing simulation reported previously⁽¹⁾ has been repeated by using the new procedure (ANNEAL Problem 17A); two other 20 keV spikes have also been "annealed." The initial distributions of defects for the latter pair were taken from individual CASCADE-CLUSTER runs performed several years ago at GE-NMPO.⁽⁷⁾ The takeoff directions of the primary knock-on atoms for the three cases are shown in Figure 11.3; the resulting cluster size distributions are in Tables 11.2a, b and c.

The numbers of time steps in each mode are included in the tables. Although several thousand steps were used, it should be emphasized that the real-time equivalent is only of the order of 10^{-6} sec. Also, the jump probability of an isolated defect is taken to be 0.1, so several thousand time steps involve only several hundred jumps for most defects.

The "interim distribution" of Table 11.2a corresponds to about the same number of vacancy migration time steps as used with the earlier method, so it is of interest to compare* the present (interim) results with those reported earlier. The principal

* No absolute comparison can be made, of course, because the order of movement, jump direction, and probability of a jump at a given time step are determined by random number selection and the sequence of random numbers is not repeated.

FIGURE 11.3. PKA Take-Off Directions Shown in Stereographic Projection

differences are that with the new procedure the total number of defects is decreased by ~25%, the numbers of large and small vacancy clusters are decreased, and interstitial clustering is increased with a concomitant decrease in the number of single interstitials. Not obvious from the table is the much more pronounced spatial separation of vacancies and interstitials due to the much larger number of interstitial time steps.

The "final distribution" for Problem 17A, after 1000 additional vacancy time steps, exhibits growth of the large vacancy clusters and elimination of intermediate sizes. The clusters are well separated, and only 3 or 4 vacancies or mobile interstitials would have any chance of interacting with them even if several thousand more time steps were used. It is interesting to note that, of the five clusters of Size 7 or greater initially present, only the V_7^* and V_{16} survived to become

* V_n signifies a vacancy cluster of size n .

the V_{10} and a V_8 , respectively. During the early stages of the annealing process, there is drastic attrition of vacancy clusters by migrating single- and di-interstitials; at one time the largest vacancy cluster was a V_5 .

Problem 231 ended with no vacancy cluster larger than a V_5 , even though it started with a V_8 and a V_9 . Problem 241, more abundantly endowed with large clusters initially, finished with four clusters of Size 7 or greater. The clusters V_9 and V_{11} are descendants of V_{11} and V_{10} of the original distribution.

A comparison of the spatial arrangements of the original distributions (from CASCADE-CLUSTER runs) for the three spikes shows that Problem 231 is considerably less concentrated than the others. For example, the average distance between single vacancies was 21.6 lattice constants for Problem 231, 13.5 for 241, and 14.8 for 17A. The absence of large clusters after annealing in Problem 231 reflects the diffuse nature of this displacement spike.

Some general observations can be made based on the simulated annealing of the three 20 keV spikes:

- About 80% of the defects are annihilated during the simulation (a time interval on the order of 10^{-6} sec in real time).
- Perhaps half of the vacancy clusters of Size 7 or larger survive and, subsequently, grow after the severe attrition during the early stages of interstitial migration.
- Some large vacancy clusters grow during annealing in regions of

TABLE 11.2. Size Distribution of Clusters for 20 KeV Spikes
(entries are numbers of clusters of indicated size)

Annealing Stage	Cluster Size											Total Defects
	<u>1</u>	<u>2</u>	<u>3</u>	<u>4</u>	<u>5</u>	<u>6</u>	<u>7</u>	<u>8</u>	<u>9</u>	<u>10</u>	<u>>10</u>	
<u>a) ANNEAL Prob. 17-A</u>												
	<u>Vacancies</u>											
Initial Distribution ^(a)	58	21	10	3	1	1	1	3	-	-	1(8)	200
Interim Distribution ^(b)	15	3	-	-	-	1	1	1	-	-	-	42
Final Distribution ^(c)	13	-	-	-	-	-	-	2	-	1	-	39
	<u>Interstitials</u>											
Initial Distribution	182	7	-	-	1							201
Interim Distribution	15	3	3	2	1							43
Final Distribution	15	3	2	2	1							40
<u>b) ANNEAL Prob. 231</u>												
	<u>Vacancies</u>											
Initial Distribution ^(d)	86	25	10	3	3	3	-	1	1	-	-	228
Final Distribution ^(e)	23	-	-	-	3	-	-	-	-	-	-	38
	<u>Interstitials</u>											
Initial Distribution ^(d)	220	4	-	-	-	-	-	-	-	-	-	228
Final Distribution ^(e)	23	-	5	-	-	-	-	-	-	-	-	38
<u>c) ANNEAL Prob. 241</u>												
	<u>Vacancies</u>											
Initial Distribution ^(f)	79	15	12	6	1	1	1	1	-	1	1(9)	216
Final Distribution ^(g)	9	-	-	1	-	-	1	1	1	-	1(9)	48
	<u>Interstitials</u>											
Initial Distribution ^(f)	201	8	-	-	-	-	-	-	-	-	-	217
Final Distribution ^(g)	13	3	4	3	-	1	-	-	-	-	-	49

(a) CASCADE-CLUSTER Run 6A; PKA direction [531].

(b) 6200 interstitial and 260 vacancy time steps.

(c) 6200 interstitial and 1260 vacancy time steps.

(d) CASCADE-CLUSTER Run 20C1; PKA direction near [101].

(e) 6000 interstitial and 1450 vacancy time steps.

(f) CASCADE-CLUSTER Run 20D1; PKA direction near [111].

(g) 5000 interstitial and 2050 vacancy time steps.

the crystal that contain only small clusters in the initial distribution.

- The net effect of short-term annealing on vacancy clustering is a decrease in clusters of Size 7 or greater.
- The majority of surviving interstitials become members of small clusters.

The interpretation and the implications of these data will be examined further. Meanwhile, several 5 keV spikes are undergoing simulated annealing for comparison with the higher energy spikes.

POINT-DEFECT CONFIGURATIONS IN METALS: EFFECTS OF INTERATOMIC POTENTIAL TYPE AND RANGE

A. DePino, Jr.

This work sought to determine the generality of some point-defect calculations in metals to see if calculated characteristics of vacancy and interstitial defects depend primarily on the potential or on the lattice structure.

The study involved determining the behavior of defects in α -iron (bcc) and copper (fcc), by use of interatomic potentials with ranges extending through 3rd and 4th neighbor distances, for comparison with published calculations made with shorter and longer range potentials. Specific characteristics calculated were:

- Vacancy relaxations
- Vacancy relaxation and migration energies
- Divacancy binding energies
- Interstitial configuration energies.

A basic assumption is that a pairwise-acting, central-force potential approximates the interaction between lattice atoms. Common potential forms used in previous computer simulation calculations are shown in Figures 11.4 and 11.5. The modified Morse potential representation by Sohm and Domingos⁽¹⁰⁾ (Figures 11.6 and 11.7) was adopted for these calculations. This potential has the form

$$\phi(r_{ij}) = \frac{A(Be^{-2\alpha r_{ij}} - e^{\alpha r_{ij}})}{e^{\beta(r_{ij} - \rho)} + 1}$$

where $\phi(r_{ij})$ is the potential energy of the two atoms i and j separated by a distance, r_{ij} ; and A , B , α , β , and ρ are constants. The numerator has the form of a Morse potential, and the denominator adjusts the range of the potential. Detailed description of the modified Morse potential can be found in Reference 11. Briefly, the Morse potentials for iron and copper were constructed so that they decreased to approximately zero at 4th and 5th neighbor distances, Figures 11.6 and 11.7. They were derived to satisfy the experimental values of heat of sublimation, lattice constant, compressibility at the equilibrium atomic spacing.

Method of Calculation

The operation of DEFECT⁽¹²⁾ computer program⁽¹¹⁾ approximates an infinite bcc or fcc lattice by a finite lattice. This finite lattice consists of two regions referred to

Neg. 0683142-3

FIGURE 11.4. Comparison of Interatomic Potentials for Copper

as the "crystallite" and the "mantle". The crystallite contains movable atoms that interact with one another and the boundary region. Surrounding the crystallite is the mantle of immovable atoms representing the remainder of the crystal lattice. In the present study, the crystallite contains 559 and 665 atoms for bcc and fcc lattices, respectively. Defects can be introduced by removing, displacing, or adding atoms in the crystallite.

DEFECT operates by calculating the force on each atom at its initial position and after a small virtual displacement; a linear extrapolation is then made to estimate where the

force components vanish, and the atom is relocated at this position. After each atom in the crystallite is moved, the forces on the atoms no longer vanish, so the process must be repeated. As many as 25 iterations are required for the forces to vanish and the energy to converge or stabilize.

Because of symmetry, there are six interstitial equilibrium configurations for the bcc and fcc lattices, as shown in Figures 11.8 through 11.11. For these interstitials the configuration energy was calculated and the order of stability determined.

Single vacancies and two vacancies separated by 1st, 2nd, 3rd, and 4th

Neg. 0682564-5

FIGURE 11.5. Comparison of Interatomic Potentials for Iron.

Neg. 0683142-1

FIGURE 11.6. Modified Morse Potentials for Copper

Neg. 0683142-2

FIGURE 11.7. Modified Morse Potentials for Iron

- I_1 - $\langle 100 \rangle$ SPLIT INTERSTITIAL (TWO ATOMS ARE SYMMETRICALLY SPLIT IN A $\langle 100 \rangle$ DIRECTION ABOUT A VACANT NORMAL LATTICE SITE).

- I_2 - $\langle 110 \rangle$ SPLIT INTERSTITIAL (TWO ATOMS ARE SYMMETRICALLY SPLIT IN A $\langle 110 \rangle$ DIRECTION ABOUT A VACANT NORMAL LATTICE SITE).

- I_3 - $\langle 111 \rangle$ SPLIT INTERSTITIAL OR CROWDION (TWO ATOMS ARE SYMMETRICALLY SPLIT IN A $\langle 111 \rangle$ DIRECTION ABOUT A VACANT NORMAL LATTICE SITE)

Neg. 0682564-11

FIGURE 11.8. Six Interstitial Equilibrium Configurations (bcc)

- I_4 - ACTIVATED CROWDION (ATOM LOCATED MIDWAY BETWEEN TWO NORMAL NEAREST NEIGHBOR LATTICE SITES).

- I_5 - OCTAHEDRAL INTERSTITIAL (ATOM LOCATED MIDWAY BETWEEN TWO NORMAL SECOND NEAREST NEIGHBOR LATTICE SITES).

- I_6 - TETRAHEDRAL INTERSTITIAL (ATOM LOCATED MIDWAY BETWEEN TWO NEAREST OCTAHEDRAL INTERSTITIAL SITES).

Neg. 0682564-10

FIGURE 11.9. Six Interstitial Equilibrium Configurations (bcc) (cont'd)

O - BODY CENTERED INTERSTITIAL (OCTAHEDRAL SYMMETRY)

H_0 - $\langle 100 \rangle$ SPLIT INTERSTITIAL

H_t - $\langle 111 \rangle$ SPLIT INTERSTITIAL

Neg. 0682509-1

FIGURE 11.10. Six Interstitial Equilibrium Configurations (fcc)

H_c - $\langle 110 \rangle$ SPLIT INTERSTITIAL

C - ACTIVATED CROWDION

T - TETRAHEDRAL INTERSTITIAL

Neg. 0682909-2

FIGURE 11.11. Six Interstitial Equilibrium Configurations (fcc) (cont'd)

neighbor distances were included in the computations. Quantities determined were the relaxations around the vacancies, vacancy relaxation migration energies, and binding energies of the divacancies.

Results

Determination of vacancy relaxations, relaxation energies, and divacancy binding energies for iron and copper were straightforward. As long as the cutoff distance in iron was approximately midway between 3rd and 4th neighbor the results were the same. Using both the polynomial and exponential forms for iron gave results differing by less than 5% for relaxations, relaxation energies, and binding energies (except for divacancies separated by a 4th neighbor distance where there was a 0.1 eV difference). The results for vacancies in copper and iron are compared

with other published data in Tables 11.3 through 11.10. Tables 11.3 through 11.6 show results obtained with 3rd and 4th neighbor polynomial forms of the copper potential. Tables 11.7 through 11.10 show results obtained with 3rd and 4th neighbor exponential forms of the iron potential.

Interstitial calculations, however, were more complex. Calculations were made at a number of cutoff distances for each configuration in iron. Although different cutoff distances gave configuration energies varying by as much as 5 eV, the energy differences among configurations remained approximately the same and the order of stability was unchanged.

The most significant result for iron was that the configuration energies obtained for the $\langle 111 \rangle$ split interstitial (I_3) and the activated crowdion (I_4) were consistently lower

TABLE 11.3. Comparison of Relaxations About a Vacancy in Copper (in % of normal distance from vacancy)

Neighbor	Cotterill and Doyama (Morse - 10th)	Girifalco and Weizer (∞ Morse)	Anderman (Morse - 4th)	Present Work	
				3rd Neighbor Cutoff	4th Neighbor Cutoff
1	3.0	2.2	1.83	2.5	2.4
2	-0.75 ^(a)	-0.4	-0.48	-0.90	-0.75
3	0.35	-	0.01	0.28	0.14
4	0.45	-	0.17	0.50	0.35
5	-	-	-0.07	-0.11	-0.11

(a) Negative sign denotes movement away from vacancy.

TABLE 11.4. Comparison of Vacancy Relaxation Energies in Copper (eV)

	Single Vacancy	Divacancy (1st Neighbor Separation)
Johnson ^(a)	0.104	0.308
Girifalco and Weizer	0.56	1.52
Wynblatt and Gjostein ^(b)	0.28 ^(c)	0.74
Cotterill and Doyama	~0.34	--
Anderman	0.22	--
Present Work		
3rd Neighbor Cutoff	0.25	0.70
4th Neighbor Cutoff	0.30	0.85

(a) Born-Mayer

(b) Infinite Morse

(c) Wynblatt later obtained 0.31 eV by increasing the number of atoms relaxed.⁽¹³⁾

than for the $\langle 110 \rangle$ split (I_2). Energies for I_3 and I_4 were at least 0.25 eV lower than the energy for I_2 for either the polynomial form (3rd or 4th neighbor), or exponential form (3rd neighbor) with variations in cutoff.

Johnson⁽¹⁸⁾ and Erginsoy et al.⁽¹⁹⁾ found the $\langle 110 \rangle$ split interstitial to

TABLE 11.5. Comparison of Vacancy Migration Energies for Copper

	eV
Wynblatt ⁽¹⁴⁾	0.90
Damask, Dienes and Weizer ⁽¹⁵⁾	1.3
Johnson and Brown ⁽¹⁶⁾	0.43
Cotterill and Doyama	0.69
Anderman	0.95
Present Work	
3rd Neighbor Cutoff	0.83
4th Neighbor Cutoff	0.73

be the stable configuration. Figure 11.12 shows the results (relative to I_2) of Johnson, the 3rd neighbor exponential form cutoff at 3.05 half lattice constants (nearly midway between 3rd and 4th neighbors), and the 3rd neighbor polynomial form (value and slope = 0 at 4th neighbor).

Interstitial configuration energies in copper were much less sensitive to cutoff. Approximately the same order of stability was found by using 3rd and 4th neighbor polynomial potentials, with the $\langle 100 \rangle$ split (H_0)

TABLE 11.6. Binding Energy of Divacancies in Copper (eV)

Neighbor Separation	Bartlett ^(a) and Dienes	Johnson	Girifalco and Weizer	Wynblatt and Gjostein	Cotterill and Doyama	Present Work	
						3rd	4th
1st	0.23-0.59	0.05	0.64	0.36	0.53	0.63	0.55
2nd	-	-	0.32	-	0.19	0.16	0.21
3rd	-	-	0.18	-	-	-0.01	0.03
4th	-	-	0.08	-	-	-0.02	-0.02

(a) Theoretical Values (Morse)⁽¹⁷⁾

TABLE 11.7. Comparisons of Relaxations About a Vacancy in Iron
(in % of normal distance from vacancy)

Neighbor	Johnson (a)	Girifalco and Weizer	Wynblatt and Gjostein	Present Work	
				3rd	4th
1	2.9	6.07	6.7	4.8	6.3
2	-2.6 (b)	-2.12	-2.3	-3.2	-3.5
3	0.30	-0.25	0.70	0.40	0.60
4	-0.16	-	-	-0.19	-0.34

(a) 3rd order polynomial fit to elastic constants

(b) Negative sign denotes movement away from vacancy

TABLE 11.8. Comparison of Vacancy
Relaxation Energies in Iron

	eV
Johnson	0.17
Girifalco and Weizer	0.41
Wynblatt and Gjostein	0.85 (a)
Present Work	
3rd Neighbor Cutoff	0.43
4th Neighbor Cutoff	0.58

(a) In a later work Wynblatt⁽¹⁴⁾
revised this value to 1.16 eV.

TABLE 11.9. Comparison of Vacancy
Migration Energies for Iron

	eV
Johnson	0.68
Wynblatt	0.88
Present Work	
3rd Neighbor Cutoff	0.73
4th Neighbor Cutoff	0.63

TABLE 11.10. Binding Energies for
Divacancies in Iron (eV)

Neighbor Separation	Johnson	Present Work 3rd
1st	0.131	0.27
2nd	0.200	0.64
3rd	-0.027	-0.08
4th	0.051	0.15

being the most stable. The energy levels decreased from 3rd to 4th neighbor potentials. Figure 11.13 illustrates the results obtained with two Born-Mayer potentials, a Morse potential extending to 10th neighbors, and the 3rd and 4th neighbor polynomial form.

The calculations using DEFECT required at least 10 iterations per vacancy configuration and 15 iterations per interstitial configuration to achieve a convergence to within 0.01 eV for the configuration energies.

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FIGURE 11.12. Comparison of Interstitial Results for Iron

Neg. 0683142-5

FIGURE 11.13. Comparison of Interstitial Results for Copper

Discussion

Vacancy relaxations, relaxation energies, and divacancy binding energies for copper are in general agreement with other calculations using Morse type potentials (Tables 11.3 through 11.6). The direction of atom movement is the same in the work shown in Table 11.3, but the magnitudes are different. Differences in the potential depth and cutoff probably account for the differences between vacancy results from Anderman's 4th neighbor potential⁽²⁰⁾ and the present 4th neighbor potential. Anderman's procedure for calculating the 4th neighbor Morse potential constants was essentially the same as that of Girifalco and Weizer.⁽²¹⁾ However, he modified the potential to fit a Born-Mayer form at 2 Å and added a smoothing function to force the potential to zero approximately halfway between 4th and 5th neighbor distances (Figure 11.4).

Vacancy migration energies for copper, Table 11.5, show good to fair agreement with the experimental value of 1.08 eV.⁽²²⁾

Two vacancies separated by a 1st neighbor distance was the most stable divacancy in agreement with previous investigations. The larger binding energies obtained by Girifalco and Weizer for divacancies separated by 2nd, 3rd, and 4th neighbor distances (Table 11.6) are attributed to more bond-sharing than exists with the present shorter range potentials. (The binding energy is determined by the number of shared bonds modified by the relaxations.) The small

(negative) binding energies in the present calculations are caused by relaxation energies overriding the small bond contribution.

The results for iron vacancy relaxation⁽²³⁾ fall within the range of other calculations (Tables 11.7 through 11.10).

Vacancy migration energies in iron are listed in Table 11.9. The energy barrier for single vacancy motion of the present calculation consisted of a single peak in agreement with Wynblatt's result, but in contrast with Johnson's calculation. The divacancy binding energies listed in Table 11.10 are consistent with Johnson's shorter range results. Two vacancies separated by a 2nd neighbor distance were the most stable divacancy, in agreement with Johnson's results. The greater well depth of the present potential is reflected in the higher binding energies.

The <100> split interstitial was found to be the lowest energy configuration in copper, in agreement with previous calculations (Figure 11.13). The 3rd and 4th neighbor polynomial potentials gave approximately the same order of stability for the six equilibrium interstitial configurations. There was, however, a decrease in magnitude of the energies above the <100> split. Upon increasing the potential range from 3rd to 4th neighbor, the order of stability becomes the same as found by Cotterill and Doyama.⁽⁹⁾

The repulsive Born-Mayer potentials favor open configurations. Thus, the octahedral is second in stability according to the results of

Johnson and Brown,⁽¹⁶⁾ and Domingos and Carbone,^(13,24) in contrast to the Morse potential results. Other fcc interstitial calculations,^(25,8) although all in agreement on the stability of the $\langle 100 \rangle$ split, differ in magnitude and order for the remaining configurations.

The $\langle 111 \rangle$ split (I_2) and the activated crowdion (I_4) were the most stable interstitial configurations in iron. This differs from the result of Johnson and Erginsoy et al. that I_2 is the stable configuration. Several variations in cutoff of the 3rd neighbor exponential form (shown in Figure 11.12) failed to change the order of stability. There was no distance at which the potential could be terminated without significantly affecting the magnitude, though not the order, of the configuration energies.

The change in energies is attributed to the large movement of the atoms in the bcc lattice. The strains produced by the interstitial atoms in the fcc lattice were smaller than in the bcc lattice, and rapidly diminished with increasing distance away from the defect. For many of the iron configurations, atoms originally at 5th neighbor distances moved well inside original 4th neighbor positions. Some 3rd neighbor atoms moved out close to 4th neighbor distances. The overlap in atom movement made it impossible to define a cutoff distance which no atoms would cross.

The relative stability of the I_3 and I_4 configurations (both along close packed directions) is, perhaps,

caused by the large strains they produce which cause many atoms to move in closer and into the deep potential well - much deeper than that used by Johnson and Erginsoy et al.

Conclusions

As a result of this study, it is concluded that the type of potential and range (cutoff) affect the results of interstitial point-defect calculations for iron. Interstitials are more sensitive because of the large strains induced. With the modified Morse potential used in this study, the stable interstitial configuration in iron could be either the $\langle 111 \rangle$ split or the activated crowdion since their configuration energies are quite close. The stability of these configurations over the $\langle 110 \rangle$ split, as found by Johnson and Erginsoy et al., is believed to be due to the greater depth of the potential well in the present calculations.

The $\langle 100 \rangle$ split interstitial was found to be the stable configuration in copper, in agreement with previous calculations using different potentials. The stable configuration appears to be a function of the lattice only, and independent of the interatomic potentials employed thus far.

The vacancy results are in general agreement with calculations published in the literature. Divacancies separated by 1st and 2nd neighbor distances were found to be the most stable for copper and iron, respectively.

COMPUTER SIMULATION OF GRAIN BOUNDARIES

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

A better understanding of mechanisms that cause changes in the properties of materials under irradiation should lead to a better basis for predicting the behavior of materials and for developing improved materials. Computer studies of radiation effects are concerned with the production and annealing of defects under the environmental conditions expected in advanced reactors. These calculations provide information on the stability of various types of atomic-scale defects and centers. Such defects nucleate larger defects which are visible under an electron microscope and which can affect the mechanical properties of materials.

Studies on grain boundaries were initiated because of their influence on radiation damage. The overall objective was to determine the influence of grain boundaries on the formation and migration energy of defects. Such defects include vacancy, vacancy clusters, impurity atoms, interstitials, and interstitial clusters.

After the study began, a need for a better definition of the structure of grain boundaries soon became apparent. Therefore, a preliminary objective of the study was to determine the geometry of atom arrangements near the grain boundary. This should disclose voids, regions of poor fit, the extent of disorder on either side of the boundary, and the extent of stress and strain fields.

Method

The GRAINS code simulates grain boundaries and defines equilibrium positions. If desired, the code will permit the introduction of various defects including vacancies, solute or solvent interstitials, or complex combinations of these, and will determine their equilibrium configurations also. However, GRAINS is not limited to grain-boundary studies. Concurrent studies have been conducted on perfect crystals, on free surfaces, and extended defect problems in fcc or bcc crystal systems.

This pseudo-dynamic program uses a central-force interatomic potential,⁽⁸⁾ and calculates the force and velocity vectors for each atom from its interaction with all other atoms within the range of the potential function. All atoms relax simultaneously a distance determined by the time interval and the velocity vector.

In a fully dynamic program, the kinetic energy of the computation cell would oscillate through maxima and minima. Many oscillations would occur before the equilibrium value of potential energy could be calculated (Figure 11.14).

A pseudo-dynamic program retains the accuracy of the dynamic program in avoiding metastable states but accelerates convergence by setting the kinetic energy of the system equal to zero when a maximum in kinetic energy is calculated. At the true maximum of kinetic energy, the potential energy would be a minimum which would indicate an equilibrium state. However, because of the

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FIGURE 11.14. Computer Experiment Methods

finite time intervals in the numerical analysis, the calculated maximum may not be the true maximum. Typically, the kinetic energy again rises and passes through another maximum as the computation continues (Figure 11.14).

After a few iterations the potential energy changes little between iterations, and convergence is reached. In the computations thus far, a 1000-atom cell was used. About 15 to 20 iterations requiring 6 to 8 min of UNIVAC 1108 computer time were needed for convergence. If the potential energy changes $< 0.0005\%$ between iterations, convergence is assumed.

The equations of motion used in this program are those developed by

Vineyard⁽²⁶⁾ based on classical Newtonian mechanics. The equations of motion and the iterations scheme are as follows:

$$F = ma$$

$$a = m^{-1} F [x_1(t), \dots, x_N(t)]$$

$$\dot{x}_i(t) = v_i(t) \quad i = 1, 2, \dots, N$$

$$N = 3n$$

$$n = \text{Number of Atoms.}$$

Replacing time derivatives by finite difference, Δt , coordinates are defined on integer steps and velocities on half-integer steps:

$$\dot{v}_i(t) \approx [v_i(t + \Delta t/2) - v_i(t - \Delta t/2)]/\Delta t$$

$$\dot{x}_i(t + \Delta t/2) \approx [x_i(t + \Delta t) - x_i(t)]/\Delta t.$$

Rearrangement gives the equations of motion:

$$v_i(t + \Delta t/2) \approx v_i(t - \Delta t/2) + \Delta t m^{-1} F_i$$

$$[x_1(t), \dots, x_N(t)]$$

$$x_i(t + \Delta t) \approx x_i(t) + \Delta t v_i(t + \Delta t/2)$$

$$i = 1, 2, \dots, N.$$

The computational cell is bounded on the exterior xz and yz planes by a mantle of fixed atoms. This mantle extends a distance that slightly exceeds the range of the potential function. Although immovable, these mantle atoms exert forces on the movable atoms in the cell interior. The top and bottom xy planes of the computational cell simulate a periodic boundary condition. Atoms in these planes are free to move within the plane, but are not allowed to move in the z direction, which imposes an image plane at the top and the bottom of the cell. Defects, such as vacancies or impurity atoms, are introduced in the center of the cell, with movement of the atoms in the z direction calculated near the defects.

The effect of boundary conditions on the computations is a definite cause for concern. To assess the magnitude of this effect, the potential energy of all the surface atoms is summed after each iteration and tabulated as the surface energy. If the change in surface energy is more than 1% of the total energy change, the computational cell is considered too small.

The construction of a grain boundary is seen in Figures 11.15, 11.16,

11.17, and 11.18 for a 6° and a 50° boundary. Octagons and squares in these figures represent movable and fixed atoms, respectively. The numbers identify each atom, and the position of the atom is at the end of the line in the center of each figure. Figures 11.15 and 11.17 show the grain boundaries before relaxation, and Figures 11.16 and 11.18 after relaxation.

Before relaxation, a gap exists between atoms on either side of the grain boundary. The nearest approach distance between atoms in grain A and atoms in grain B before relaxation is a specified quantity. Several nearest approach values were chosen.

(Atoms that would be closer are rejected from the problem.) As seen in Figure 11.19, the minimum grain-boundary energy occurred for a value equal to $\sqrt{2} \underline{a}$, where $\underline{a}$ is the lattice spacing. This is the first neighbor distance in the fcc lattice. Boundaries generated with reject values less than $\sqrt{2} \underline{a}$ can be considered in compression; those with a value greater than $\sqrt{2} \underline{a}$, in tension. This study was made on cells that were identical in size and in surface area with the reject parameter the only variable.

Results

The structure of the 6° grain boundaries, seen in Figure 11.16, shows several distinct regions. Starting from the top, there is a void near Atom 325 which will be referred to as a "poor-fit" region. Proceeding down the boundary, there

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FIGURE 11.15. This Plot Is of an fcc Lattice Viewed in the (100) Plane Grain Boundary at $X = 11$. (For a rotation of 6.00 degrees.) This Plane Is: $Z = 3$.

Neg 0682827-2CN

FIGURE 11.16. This Plot Is of an fcc Lattice Viewed in the (100) Plane Grain Boundary at $X = 11$. (For a rotation of 6.00 degrees.) This Plane Is: $Z = 3$.

Neg 0682827-3CN

FIGURE 11.17. This Plot Is of an fcc Lattice Viewed in the (100) Plane Grain Boundary at $X = 11$. (For a rotation of 50.00 degrees.) This Plane Is: $Z = 3$.

FIGURE 11.18. This Plot Is of an fcc Lattice Viewed in the (100) Plane Grain Boundary at $X = 11$. (For a rotation of 50.00 degrees.) This Plane Is: $Z = 3$.

Neg 0683199-1

FIGURE 11.19. "REJ" Optimization Study

is a high-energy region near Atom 281. For example, Atom 281 has an energy of -0.97 eV; whereas, a typical atom in the bulk would have an energy of -1.45 eV. This high-energy region will be referred to as a "tight-fit" region. The area of Atom 264 is a "good-fit" region. Atoms have approximately the same energy as atoms in the bulk and the grain boundary is quite indistinct.

The extent of the grain boundary, or of the strain field around it, can be determined by the displacement of the atoms during relaxation to the equilibrium configuration. As seen for the 6° and 50° boundaries (Fig-

ures 11.20 and 11.21), the grain-boundary region is quite narrow. Displacements of atoms are less than $1/10$ of the lattice constant at approximately 2 unit cells removed from the boundary, a distance of about $7 \text{ \AA}$. The width of the boundary as seen by electron microscopy, 20 or $30 \text{ \AA}$, depends on the strain necessary to affect diffraction. This would be consistent with the calculations since 5% strain is observed at 12 to $15 \text{ \AA}$ from the boundary. Energy contours are complex, with high-energy regions (i.e., low negative energy) found adjacent to very low energy regions. Atoms differing in energy from the

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FIGURE 11.20. Strain-Field 6° Tilt Boundary fcc Nickel

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FIGURE 11.21. Strain-Field 50° Tilt Boundary fcc Nickel

bulk value extend about the same distance from the grain boundary as atoms with significant displacements.

Vacancy formation energy is an important parameter in the production of radiation damage and in annealing. The formation energy of a vacancy is defined by the equation

$$E_f^{lv} = E_e^{lv} - E_s^{lv}$$

where E_e^{lv} is the configuration energy and E_s^{lv} is the surface energy. To calculate this quantity, the computational cell with the grain boundary is relaxed, an atom is removed, and the cell is again relaxed. The difference in potential energy of the

cell with and without the vacancy is the configuration energy. Subtracting the energy recovered when the atom is placed on the surface from this value yields the formation energy.

Formation energy calculations were made by removing atoms from a number of different positions on close packed $\langle 110 \rangle$ lines from the grain boundary. The energy recovered by putting an atom on the free surface and on a step in a free surface was calculated for self-consistency. A number of configuration energies of

atoms along close-packed lines extending from the tight-fit and bad-fit regions of the 6° grain boundary are plotted in Figure 11.22. From values for Atoms 325, 319, and 313, about 4 unit cells are required to approach the formation energy for a perfect crystal. Atom 301 is more tightly bound than an atom in a perfect crystal, and a slightly higher formation energy for a vacancy is required.

Conversely, Atoms 281 and 286, which are in a high-energy region, are found to have negative formation energies. This implies these atoms would be ejected from their sites to form vacancies spontaneously if this

could be accommodated geometrically. However, this might also mean that the reject criterion was in error and that atoms should not be in these positions. Formation energies vary along the close-packed line extending from the tight-fit region into the crystal. Obviously, the grain-boundary region is unique and perfect-crystal values are inappropriate for atoms in this region.

Migration Energies

Migration energies for defects are essential for calculations of annealing and diffusion. The migration energy of a vacancy is calculated by incrementally moving an atom in the first-neighbor position toward the vacancy. This movement is accomplished by changing the value of the coordinates by the incremental distance for the atom to be moved, allowing the atom 2 degrees of freedom during relaxation, and permitting the atom to find its equilibrium position. In this manner the atom chooses its migration path. The energy required to move the atom and, conversely, the vacancy, is the relative change in the potential energy calculated after each move. The path of the atom in moving from Position 319 to a vacancy at the side of 325 is seen in Figure 11.23. This path involves little movement in the y direction until the Peierl's barrier has been overcome. The atom then moves rapidly in the y direction and finally hooks into the vacant site.

The migration energy calculated for this path (Figure 11.24) is

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FIGURE 11.22. Vacancy Formation Energy Near 6° Grain Boundary

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FIGURE 11.23. Vacancy-Migration Path
Bad-Fit Region 6° Boundary

0.6 eV for movement of the vacancy toward the grain boundary. However, 1.04 eV is required to move in the opposite direction, that is, from the grain boundary into the bulk. In this case the grain boundary would be a sink for vacancies, and the void around Atom 325 might enlarge.

Vacancy migration toward the good-fit region displays the opposite behavior (Figure 11.25); about 0.6 eV is required for the vacancy to migrate into the bulk. However, 1.32 eV would be required for a vacancy to diffuse to the grain boundary, with the grain boundary serving as a source of vacancies.

The path of an atom moving toward the grain boundary, as seen in Figure 11.26, is a tortuous path.

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FIGURE 11.24. Vacancy-Migration Region of Bad-Fit 6° Boundary

VACANCY MOVEMENT
UNITS: NEAREST NEIGHBOR DISTANCE $2.49\text{\AA}$

Neg. 0683033-5

FIGURE 11.25. Vacancy-Migration Region of Good-Fit 6° Boundary

Grain-boundary diffusion is a common and often dominant mechanism. The diffusion of a vacancy on the grain boundary was calculated by starting with Atom 325 removed and moving Atom 440 (a first neighbor in the plane immediately above) in the z direction until it came to rest near the original site of Atom 325. An energy of less than 0.6 eV was calculated for this movement, demonstrating that this is a favored diffusion mechanism.

Calculations were also performed to determine the value for vacancy migration in a perfect crystal. The value of 1.07 eV was calculated, in agreement with Doran's calculations,⁽²⁷⁾ by using the DEFECT program for fcc nickel.

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FIGURE 11.26. Vacancy-Migration Path Good-Fit Region 6° Boundary

Impurities

The energy required to insert impurity atoms at various locations near a grain boundary is required to determine the stability of impurity complexes or impurity-vacancy complexes. Only carbon impurity atoms have been considered thus far, since they significantly affect grain-boundary sliding and migration in fcc metals. Of primary interest is the energy required to form clusters of impurity atoms at grain boundaries. These clusters could impede grain-boundary movement and thus affect plastic deformation at elevated temperatures.

The energy required to insert carbon atoms at good-fit regions and at different distances from bad-fit regions are tabulated in Table 11.11. To insert a carbon atom in a bad-fit region requires only 1/4 of the energy required in a perfect region and less than 1/3 of that required at the good-fit region. The energy required to insert the carbon atom rises rapidly along the 100 direction from the bad-fit region and is nearly equal to the perfect-crystal value approximately 3 planes in. To insert a carbon atom in a good-fit region requires 71% of the energy required for the perfect crystal. To make these calculations, the carbon atom is inserted in a somewhat arbitrary position and is given 3 degrees of freedom to move to its equilibrium position. In all cases the favored position for the carbon atom was a tetragonal impurity position.

Johnson's potential function was used for the nickel-carbon interac-

tion,⁽²⁸⁾ a short range potential extending over 2.4 Å.

Free Surfaces

Calculations of a free surface with this program yielded a value of 1125 ergs/cm², in agreement with other investigators.

Calculations are in progress on the energy of vacancy migration toward free surfaces and the energy required to insert carbon atoms near a free surface.

Conclusions

The GRAINS code can apparently be used to determine detailed structures and properties of grain boundaries. The code can also be used to study other types of extended defects with or without the presence of a grain boundary. Apparently, grain boundaries significantly change formation and migration energies for defects involved in the production or annealing of radiation damage.

TABLE 11.11. Energy Required to Insert a Carbon Atom into Various Positions near a 6° Tilt Boundary in fcc Ni

Position	E, eV	E/E _b
At Good-Fit Region	1.2	0.71
At Bad-Fit Region	0.40	0.23
One Plane in from Bad-Fit Region	1.24	0.73
Two Planes in from Bad-Fit Region	1.26	0.75
Three Planes in from Bad-Fit Region	1.64	0.97

E_b = The bulk value (i.e., the energy required to insert an atom into a perfect crystal).

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IRRADIATION DAMAGE TO REACTOR METALS

A. L. Bement

REACTOR METALS RESEARCHIRRADIATION FACILITIES OPERATION

A. L. Ward

The purpose of this phase of the program is to provide for the timely processing of data obtained from the radiation and testing of a variety of reactor structural materials. Irradiations will be performed in aqueous, gaseous, and molten metal environments. Elevated temperature irradiations and tests will be emphasized. Special testing techniques, applicable to either irradiated or un-irradiated specimens, will be developed.

Facilities Operation

The experiments discharged from the various Engineering Test Reactor

(ETR) Core positions for Cycle 96 are summarized in Table 12.1. Estimates of neutron fluxes and exposures are also given.

The operating history of the ETR and of the G-7 hot water loop for Cycles 95 and 96 are given in Table 12.2. Loop operation was normal for these cycles. No data is yet available for control materials inasmuch as out-of-reactor loop Cycle 95 has not been completed.

The present schedule for the ATR Pressurized Hot Water Loop calls for Cycle 1 start-up on January 6, 1969. A total of 168 specimens of 304 and 316 stainless steels are being machined for the initial loading and the support hardware designs are complete.

TABLE 12.1. Summary of Experiments Discharged from ETR Core Positions, Cycle 96

Experiment Designation	GEH-20-294
No. of Specimens	36
Type of Specimens	Zr-2 corrosion blanks
Estimated Flux, $n/cm^2/sec$ (a)	10.2×10^{13}
Estimated Fluence, n/cm^2 (a)	5.75×10^{20}

(a) Neutron with energies greater than 1 MeV.

Note: Total specimens discharged, ETR G-7 Loop, Cycle 96 = 36
Total specimens discharged, ETR G-7 Loop, to date: 3560

TABLE 12.2. Summary of Operating History for ETR and G-7 Hot Water Loop--ETR Cycle 94

<u>Operating History</u>	<u>ETR Cycle Number</u>	
1. <u>Reactor History</u>	<u>Cycle 95</u>	<u>Cycle 96</u>
Start up, ^(a) date	4-17-68	6-5-68
End of Cycle, date	5-26-68	7-17-68
Megawatt Days	5521	5891
Effective Days at 175 MW	31.6	33.66
Number of Scrams ^(b)	15	14
Number of Shutdowns	3	4
2. <u>G-7 Hot Water Loop History</u>		
Maximum Temperature	530 °F	530 °F
Effective Days Above 200 °F	33.0	34.66
Effective Days at Operating Temp.	30.4	30.74
Operating Efficiency ^(c)	104.5%	97.4%
Temperature Efficiency ^(d)	92.2%	88.7%

(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).*

MICROSTRUCTURAL MODIFICATIONS
TO AUSTENITIC STAINLESS STEELS
AND NICKEL-BASE ALLOYS
 I. S. Levy

The purpose of this program is to determine the effects of modified microstructures upon the irradiation-stability of austenitic stainless steels and nickel-base alloys. These structural modifications are evaluated by tensile and stress-rupture tests and by microstructural examinations.

Austenitic Stainless Steels

As reported last quarter, ⁽¹⁾ thermomechanical treatments capable of producing thermal- and radiation-resistant microstructures while retaining significant as-treated ductility were investigated. These treatments involved the application of a constant tensile stress at temperature ("stress-aging").

Tensile tests at 1200 °F (649 °C) were conducted during the past quarter on several treatments. Compared

to the annealed condition, a treatment (30,000 psi at 1200 °F for 1/2 hr) increased the proportional elastic limit (PEL) by about 350% (21,400 versus 6,300 psi), the 0.2% yield strength (YS) by over 100% (30,450 versus 14,310 psi), and the ultimate tensile strength (UTS) by about 20% (51,450 versus 42,100 psi) with no change in ductility from that of the annealed condition [28.7% uniform elongation (UE) and 43.7% total elongation (TE)].

Another treatment (38,000 psi at 1200 °F for 1/2 hr) produced essentially the same yield strength as does 20% cold-rolling (42,300 versus 43,800 psi) while retaining much greater ductility. The uniform and total elongations for this treatment were 13.5 and 24.5%, respectively, compared with 2.5 and 7.1% for the cold-rolling process.

A comparison to determine the similarity, if any, between effects of the previously noted stress-aging treatments and 20% rolling at 1100 °F (593 °C) was made. A 10% advantage in yield strength for the stress-aged condition was found, with both treatments producing the same elongation values. Moreover, while a 20% reduction in thickness was required to achieve this strength level with the warm-working treatment, the same results were obtained by only an 8% reduction in specimen diameter with the stress-aging treatment.

These results suggest that the continuous application of stress for a period of time may result in strengthening by the decoration of

relatively few dislocations, rather than by the generation of massive tangles as in normal working techniques. Such results may be advantageous, not only from the point of view of ductility retention, but because prior precipitation of phases might tend to precipitate during irradiation. This effect could lead to improved irradiation stability, as observed in nickel-base alloys.⁽²⁾

Alternatively, a cell structure developed because of recovery tendencies at the stress-aging temperature could produce these results and, thereby, a given strength level faster (i.e., with less deformation being required) than with the other techniques. In this case, then, improved radiation stability due to prior precipitation might not be observed.

IN-REACTOR MEASUREMENTS OF MECHANICAL PROPERTIES

E. R. Gilbert and N. E. Harding

The in-reactor measurements program is intended to determine the effects of irradiation on the mechanical properties of reactor structural materials, to establish property data suitable for design purposes, and to consider mechanisms through irradiation effects on macroscopic deformation. The program currently involves conducting in-reactor creep measurements on annealed 304 SS, Hastelloy X, heat-treated Zr-2.5 Nb, and Zircaloy-2. Capsule development is being pursued for the purpose of extending program capabilities.

304 Stainless Steel

One of the pertinent questions to the problem of in-reactor creep is the applicability of postirradiation creep testing to in-reactor creep behavior. In consideration of the stress-rupture life of Type 304 SS as affected by neutron irradiation, a creep stress versus temperature-compensated rupture-life (theta parameter) plot (Figure 12.1) was made for in-reactor, postirradiation, and unirradiated control creep tests using data by Bloom.⁽³⁾ The conclusion to be drawn from this plot is that the rupture-life of 304 SS tested in-reactor during neutron irradiation or postirradiation is essentially the same for comparable

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FIGURE 12.1. Creep Stress Versus Temperature-Compensated Rupture Life (Theta Parameter) for Type 304 SS

conditions of stress and temperature. The effect of neutron irradiation, as reflected both in in-reactor and postirradiation tests, is to reduce the rupture-life or the rupture-strength as compared to the unirradiated control values.

Zr-2.5 Nb

An analysis using the Blackburn yielding creep model⁽⁴⁾ was made of PNL in-reactor and unirradiated control creep data generated at 300 °C. The results of this analysis are shown in Figure 12.2 as a plot of the acceleration factor, f , versus

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FIGURE 12.2. The Creep Rate Acceleration Factor, f , as a Function of the Applied Stress, w , and the Internal Stress, I , for Zr-2.5 Nb at 300 °C

the applied stress, w , and the internal stress, I . In making this correlation, a value of 4 for the stress power was used as the most appropriate for the data. The experimental data points appear to fit the theoretical curve fairly well.

Another gratifying feature of this analysis was that the internal stress was found to be 30 kg/mm^2 for Zr-2.5 Nb, while Hesketh⁽⁵⁾ found a value of 17 kg/mm^2 for Zircaloy-2. Since the ratio of the internal stress for Zr-2.5 Nb to that of Zircaloy-2 is similar to the ratio of the yield strengths of the two alloys, the greater calculated internal stress for Zr-2.5 Nb is consistent with its higher yield strength and its ability to support a greater internal stress.

GRAIN BOUNDARY STABILIZATION

E. R. Bradley

The purpose of this program is to study the influence of inert gases on the irradiated and unirradiated properties of metals. Particular emphasis is placed on the interaction of grain boundaries with inert gas, with gas filled bubbles, and with voids.

During this period, thin nickel foils (~ 3 mils) have been produced by sputtering pure nickel onto an aluminum substrate. The aluminum substrate was removed by dissolution in boiling caustic, leaving a circular 1 in. diam foil.

The principal reason for using sputtered foils was to obtain extremely fine grain size material

containing small concentrations of inert gas. Grain sizes of approximately 100, 200, and 500 Å (as measured by X-ray line broadening) were obtained by variation of substrate temperature during deposition. The concentration of argon gas entrapped in the foil was varied by changing the substrate potential used during deposition. Gas analyses of the foils are not available, so neither the argon concentration nor the effectiveness of changing the substrate potential on the argon concentration is known.

Isothermal annealing at 450 °C in 5-hr intervals up to a maximum of 30 hr has been investigated. The temperature was picked to correspond with the highest temperature of a three-zone capsule in which the foils will be irradiated to determine the effect of small grains and inert gas on the formation of voids in nickel.

The apparent grain size was found to increase from 100 to 200 Å during the first 10 hr, remain constant for 5 hr, and then decrease to 150 Å for the remaining 15 hr. This increase and decrease in grain size was accompanied by an increase and decrease in the intensity of the diffracted X-ray beam. The initial increase was probably due to recovery of strain in the foil rather than an actual increase in grain size. The reason for the subsequent decrease is not known at this time. Possible explanations include (1) substructure formation, (2) precipitation reaction, or (3) a precrystallization structure. Further experiments are underway to determine the process involved.

A foil in the as-sputtered condition has been electrolytically thinned and examined by electron microscopy, Figure 12.3. The material appears to have a cellular type structure with a high dislocation density closely approximating a heavily deformed material, but on a

much finer scale. Electron diffraction patterns of the material indicate only slight misorientation between the cells. A foil annealed at 700 °C will be examined to determine whether this cell structure will change into a regular grain type structure.

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120,000 X

FIGURE 12.3. Microstructure of Sputtered Nickel Foil
(Grain size as measured by X-ray line broadening was
approximately 100 Å)

IRRADIATION DAMAGE TO HEAVY SECTION VESSEL STEELS

J. A. Williams and C. W. Hunter

The objective of this program is to obtain a quantitative engineering evaluation of the effects of irradiation on heavy-section, pressure vessel steels. Included in this broad objective are:

- The establishment of a suitable specimen and analytical approach for investigating the effects of irradiation on plane strain fracture toughness.
- The development of valid correlations and surveillance methods for analyzing the service integrity of heavy-section vessels from fracture-mechanics specimen data.
- The determination of the effects of typical nuclear environments on possible microstructural embrittling reactions in the base metal, weld metal, and heat affected zones of candidate pressure vessel steels.

Investigations of Fracture Toughness of ASTM A533B Steel with the DCB Specimen

The size of specimens is recognized to be of major importance in irradiation experiments in terms of nuclear heating, neutron economy, and test material consumption. A major task of the irradiation damage program was to compare the size effects of various fracture mechanics specimens to determine the optimum specimen for an irradiation program. The experimental work of this task

at PNL has been primarily oriented towards the characterization of the Double Cantilever Beam (DCB) specimen. The DCB type specimen offers some advantage over other types of specimens in that multiple fracture toughness measurements can be made, fracture arrest values can be determined, and fatigue crack preparation is unnecessary. Its major disadvantages are related primarily to the limited degree of characterization of the specimen and, more specifically, to the limited available knowledge regarding the effect of side grooves on plane strain fracture toughness.

The initial material for the PNL Irradiation Effects to Heavy Section Pressure Vessel Steels program was obtained from Plate 1 of A533, Grade B, Class 1 steel procured for the HSST program. Heat treatment and characterization data of the plate are reported in Reference 6. Fracture initiation and fracture arrest measurements were made using the Double Cantilever Beam (DCB) specimen. A typical DCB specimen is shown in Figure 12.4. The specimen geometry used for this investigation for each size specimen was $B = h/2 = w/0.35$ where B is the gross thickness, w is the net thickness, and $h/2$ is the beam depth. Root radii were 0.001 in. maximum per inch of gross thickness. The crack starter saw cut, C_0 , is approximately $3B$ and the length L is approximately $13B$. The crack plane of the specimen was normal to the plate surface and parallel to the rolling direction (WR) for all specimens.

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FIGURE 12.4. Double Cantilever Beam Specimen

Test temperatures were maintained on the specimen using two types of cryostats, one combining liquid nitrogen flow control and heaters, and the other using flow control of liquid nitrogen sprayed on the specimen. Thermocouples along the length of the specimens were used to control and monitor temperatures at various crack lengths.

An Instron tensile machine was used to load the specimens. Load and deflection measurements on the specimen were determined from load cell and crosshead displacement measurements of the tensile machine. Crosshead speeds were varied with crack length so that stress intensity rates were held within approximately 10 to 20 ksi/ $\sqrt{\text{in.}}$ /min. A specimen balancing mechanism was used to minimize the nonlinearity in the lower portion of the load extension curve.

The results of the work to date are summarized in Table 12.3 and Figure 12.5. The value $B/(K_{I_C}/\sigma_{ys})^2$ in Figure 12.5 is indicative of the

ratio between the test specimen thickness and plastic zone size at the crack tip. A significant increase in fracture toughness, K_{I_C} , as a function of this ratio can be interpreted as representing a significant decrease in plane strain constraint. The value of $B/(K_{I_C}/\sigma_{ys})^2$ appears to be about 4.0 for the lower limit of valid plane strain fracture toughness for A533B using the DCB specimen. Since the actual crack runs in the web area of the specimen, a more realistic validity criteria for the DCB specimen thickness, $w/(K_{I_C}/\sigma_{ys})^2$, would yield a value of 1.4 as a lower limit for $w = 0.35B$, the specimen geometry used in these investigations.

The temperature dependency of fracture toughness and arrest are shown in Figure 12.6. The fracture toughness of A533B appears to be adequately defined between -160°C and -80°C using the DCB specimen geometry of this study. All data plotted are taken from Table 12.3 and are valid within the above established criteria. It is observed that the difference between K_{I_C} and K_{I_a} is generally temperature insensitive and not large.

Fracture toughness profile data (Figure 12.7) indicate an increase of K_{I_C} at the surface of the plate. Data for plate center (1/2 T position) are the average of a number of points as shown in Figure 12.6. The invalid point, $B/(K_{I_C}/\sigma_{ys})^2 < 4$, at -140°C was included in the plot because of the significance that data at -140°C for the 1/4 T and 1/2 T positions from the 1 in. specimen

TABLE 12.3. Fracture Toughness Values of ASTM A533, Grade B, Class 1 Steel

Specimen Number	Thickness, in.	Temp, °C	Yield Strength, (a) Ksi	K_{Ic} , Ksi√in.	K_{Ia} , Ksi√in.	$\frac{B}{(K_{Ic}/\sigma_{ys})^2}$
01KA9 (b)	0.5	-160	117	32.1	29.9	6.64
01KA9 (b)	0.5	-160	117	32.8	30.8	6.36
01KA9 (b)	0.5	-160	117	38.6	29.8	4.59
01KA9 (b)	0.5	-160	117	35.8	32.8	5.34
01KA9 (b)	0.5	-140	105	58.5	31.0	1.60
01KA9 (b)	0.5	-140	105	68.0	(c)	1.19
01KA32 (b)	0.75	-160	117	39.3	28.1	6.64
01KA32 (b)	0.75	-160	117	41.0	33.8	6.14
01KA32 (b)	0.75	-160	117	41.7	30.1	5.90
01KA32 (b)	0.75	-160	117	47.8	28.6	4.48
01KA32 (b)	0.75	-140	105	45.4	31.9	4.18
01KA32 (b)	0.75	-140	105	35.9	32.8	6.40
01KA32 (b)	0.75	-140	105	38.1	55.8	6.00
01KA32 (b)	0.75	-140	105	46.4	(c)	3.84
01KA22 (b)	1.0	-160	117	30.4	29.9	14.84
01KA22 (b)	1.0	-160	117	33.7	31.1	12.02
01KA22 (b)	1.0	-160	117	40.5	35.7	8.36
01KA22 (b)	1.0	-160	117	40.5	37.8	8.36
01KA22 (b)	1.0	-160	117	42.8	36.3	7.46
01KA22 (b)	1.0	-140	105	48.1	37.8	4.75
01KA22 (b)	1.0	-120	97	61.1	56.0	2.52
01KA22 (b)	1.0	-120	97	77.5	(c)	1.57
01KA22 (b) (d)	1.0	-160	117	42.2	33.6	7.70
01KA22 (b) (d)	1.0	-160	117	41.0	39.0	8.16
01KA22 (b) (d)	1.0	-160	117	43.9		8.90
01KA60 (b)	2.0	-160	117	44.0	41.0	14.13
01KA60 (b)	2.0	-160	117	43.6	42.5	14.40
01KA60 (b)	2.0	-140	105	51.3	46.7	8.36
01KA60 (b)	2.0	-140	105	53.7	44.5	7.67
01KA60 (b)	2.0	-120	97	57.3	46.7	5.85
01KA60 (b)	2.0	-100	91	59.8	53.3	4.63
01KA60 (b)	2.0	-80	86	58.5	58.2	4.31
01KA60 (b)	2.0	-80	86	85.7	61.8	2.00
01KA60 (b)	2.0	-60	83	71.5	71.1	2.70
01KA60 (b)	2.0	-60	83	92.8	78.6	1.63
01KA60 (b)	2.0	-40	79	167.8		.45
01KA20 (e)	1.0	-160	117	37.5	35.1	9.91
01KA20 (e)	1.0	-160	117	44.8	29.5	6.83
01KA20 (e)	1.0	-140	105	41.5	30.8	6.39
01KA20 (e)	1.0	-140	105	41.8	30.4	6.30
01KA20 (e)	1.0	-120	97	39.7	37.5	6.00
01KA20 (e)	1.0	-120	97	55.2	43.0	3.14
01KA18 (e)	1.0	-160	133	43.2	39.2	9.47
01KA18 (e)	1.0	-160	133	59.5	34.3	5.00
01KA18 (e)	1.0	-140	119	57.8	44.4	4.37
01KA18 (e)	1.0	-140	119	66.9	45.7	3.18

(a) Yield Strength values for Plate #2 of ASTM A533B steel from Reference 2.

(b) Specimen from plate center position in 12" A533B plate.

(c) Arms of specimen broke.

(d) Specimen was remachined with pin holes and crack starter slot after arm breakage.

(e) Specimen 01KA20 from 1/4 T position and 01KA18 from surface of plate.

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FIGURE 12.5. K_{IC} Plotted as a Function of $B/(K_{IC}/\sigma_{YS})^2$ at -160, -140, and -120 °C for ASTM A533B

exhibited plane strain validity. The fact that the toughness value was forced to be invalid, even with a higher yield strength at the plate surface, indicates a higher toughness at the plate surface.

On the basis of validity criteria established, the DCB specimen offers little size advantage over other types of specimens for use with low yield strength steels. Some qualification is due since the total number of tests obtained from the DCB specimen would probably be greater for an

equivalent volume of material used for other types of specimens. A substantial cost savings could, therefore, be realized using the DCB specimen. Large DCB specimens would probably encounter large fluence gradients over their length during irradiation in most reactors. This fluence gradient, then, might be used to some advantage with DCB specimens since they provide a simple means of measuring the effect of fluence gradients on material properties.

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FIGURE 12.6. Temperature Dependency of Plane Strain Fracture Toughness and Fracture Arrest Values for ASTM A533B Steel (The data are based on the plane strain fracture toughness validity criteria for the DCB specimen as shown.)

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FIGURE 12.7. Fracture Toughness Profile Through 1/2 Thickness of A533B Steel Plate (T denotes plate thickness.)

Some additional studies are yet required if the DCB specimen is to be extensively applied to the measurement of fracture toughness of irradiated steels. The specimen might be optimized by increasing the beam height and increasing the net thickness. Increasing the beam height would decrease the maximum fiber stress at the crack plane. Increasing the net thickness would probably increase the temperature range over which valid fracture toughness measurements could be made.

Additional work in progress includes an investigation of the effect of round side grooves versus sharp side grooves and the effect fatigue precracking on fracture toughness for DCB specimens.

Irradiation Program

The compact K_{I_C} specimen will be used for the determination of irradiation effects on fracture toughness of A533B base material during program phase one. The objective of this phase is to determine effects of irradiation at fast fluences of 1×10^{19} n/cm² at 450 °F and 5×10^{19} n/cm² at 450 and 550 °F. Compact K_{I_C} specimens of the 1 T geometry will be used to determine whether there are any significant shifts in toughness due to irradiation in temperature ranges where valid measurements can be made.

Initial irradiation capsule designs have been completed for irradiations at both temperatures. The 550 °F irradiations will be conducted in the M-3 hot water loop of the

Idaho Nuclear Experimental Test Reactor (ETR). Temperature control will be maintained by gamma heating and control of coolant flow through the capsule. Each specimen temperature will be monitored with thermocouples. Flux monitors will be used to determine the fluence of each specimen and a dummy specimen will be monitored to determine neutron attenuation through the specimen. The 1T compact K_{I_C} specimens are fitted in the capsule with their thickness direction parallel to the axis of the pile and the machined crack opening pointing radially toward the center of the pile. This irradiation position, by orienting temperature and fluence gradients radially from the subsequent crack line of the specimen, will reduce the influence of such gradients along the crack front. The capsule accommodates 30 specimens divided into three subsections of 10 specimens each. Each subsection can be translated or rotated end to end so that flux gradients from specimen to specimen can be minimized. Tubes will hold 30 tensile specimens for use in evaluating mechanical properties for the same conditions of fluence, spectrum, and temperature to which the fracture specimens are subjected.

The 450 °F irradiation will be conducted in a reflector position of the ETR since temperatures this low cannot be obtained in the M-3 loop facility. The irradiation capsules will maintain specimen temperature close to 450 °F through the use of a gas annulus, gamma heating, and heaters for final temperature control.

The compact K_{I_C} specimens, in pairs, will be oriented with their tension testing axis parallel to the axis of the reactor and with the machined crack opening facing radially toward the center of the pile. The capsule will accommodate a total of 10 specimens. The temperature monitoring and control of all specimens will be accomplished using one thermocouple. Flux monitoring will be similar to that described for the 550 °F irradiation capsule. No fluence gradient at a given crack line will be encountered with such an arrangement of specimens, but temperature gradients of about 20 °F maximum are expected.

FRACTURE STUDIES

R. G. Hoagland

The report for the previous period⁽¹⁾ outlined a method for calculating the orientation factors about a crack tip for any shear plane and shear direction in the hcp crystal system. It was noted that the orientation factors are derived from the transformation matrix, $\tilde{T}$, whose terms are the direction cosines between the axes of the orthogonal $x y z$ coordinate system and the $a_1 a_2 a_3 C$ axes in the hexagonal Miller system. We examine the role of twinning in influencing the anisotropy in toughness on the premise that the orientation having the greatest deformation will also have the greatest toughness, other things being equal. The task, then, becomes one of determining the orientation factors for the crack-tip shear stresses resolved onto the twinning planes for the two

cracking directions. The orientations of the unit cell with respect to the crack plane that need to be considered are shown in Figure 12.8. The analysis has been simplified by considering only basal poles contained within the ND-TD plane, this being justifiable since this plane has by far the largest density of basal poles in rolled Zircaloy-2 plate. There are three angles defining the unit cell orientation:

- θ , the angle made between a radius vector from the crack tip to the unit cell and the crack plane.
- ϕ , the angle between the basal pole and the crack front measured from the ND direction.
- λ , the angle of rotation of the unit cell about the basal pole measured between the $[11\bar{2}0]$ direction and the rolling direction.

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FIGURE 12.8. Schematic Showing the Manner in which the Orientation of the hcp Unit Cell with Respect to the Crack Plane is Defined. The Angle θ is Measured in the RD-TD Plane from the Plane of the Crack.

If we consider the $\vec{a}$ vectors to be unit vectors (for which $\vec{c}$ has length c/a), then the $\tilde{T}$ matrix is given for a RD crack as

$$\tilde{T} = \begin{bmatrix} \cos\lambda & \cos\phi\sin\lambda & \sin\phi\sin\lambda \\ -\sin(\lambda + \pi/6) & \cos(\lambda + \pi/6) & \cos\phi\sin\phi\cos(\lambda + \pi/6) \\ -\cos(\lambda + \pi/3) & -\cos(\lambda - \pi/6) & \cos\phi\cos(\lambda - \pi/6)\sin\phi \\ 0 & -(c/a)\sin\phi & (c/a)\cos\phi \end{bmatrix}$$

In the case of a TD crack, it can be shown that the $\tilde{T}$ matrix can be obtained from the one given above by exchanging the first and second columns and changing the signs of the terms in the first column. Details of the analysis for determining the orientation factors for the crack-tip shear stress resolved on a particular plane and direction for the hcp system have been described in the previous report. The orientation factors were determined for the three most important deformation systems, the two twin systems, $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ and $\{11\bar{2}1\} \langle 11\bar{2}\bar{6} \rangle$, and the slip system, $\{10\bar{1}0\} \langle \bar{1}2\bar{1}0 \rangle$. For each of these deformation systems, the orientation factors were calculated for both the RD and TD crack orientation at 10° intervals for each of the three angles θ , ϕ , and λ over a range of orientations sufficient to completely describe the orientation factors for each of the deformation systems.

As shown previously, the calculations are based on linear elastic crack-tip stress field equations neglecting anisotropy. Because an elastic solution is being applied in a region that would be occupied by a plastic zone, not much faith can be placed in the accuracy of the

absolute values of the orientation factors although the calculations should indicate the probability for the occurrence of deformation.

The results, while rather complex, are not too dissimilar for the various deformation systems; so only the results for the $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ twin system are shown here. To afford a means of presenting the results, it is necessary to eliminate one of the dimensions. This is accomplished by further idealizing the texture by setting the $(10\bar{1}0)$ pole parallel to the rolling direction. This idealization is not unrealistic in view of the textures. Therefore, for each orientation of the unit cell defined by some θ and ϕ , there are six orientation factors for each of the six possible λ values. These orientation factors can be summed up to give a relative value of the orientation factor that reflects the probability for deformation for the unit cell orientation. For the twin systems, only positive orientation factors can be included in this sum because twinning can occur in only one direction. A negative orientation factor simply indicates that a twin cannot be propagated under the action of the resolved shear stress no matter how large. In the case of slip, the sign of the orientation factor is disregarded in the sum.

For the $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ twin system, the results of these computations, expressed in terms of the sum of orientation factors for the idealized texture, are shown in Figures 12.9 and 12.10 for the two crack orientations. For the RD crack orientation, it can be seen that the orientation factors for $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ twinning decreases rapidly as the basal pole is rotated away from the ND direction (i.e., as ϕ increases). While this is also true for small values of θ

for a TD crack, the orientation factors increase as ϕ increases for θ between 50° and 90° , giving much larger values for the TD crack orientation factors than for the RD crack over a large range of ϕ . These results indicate that $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ twinning should be more predominant in the vicinity of a crack oriented perpendicular to the rolling direction than for a crack parallel to the rolling direction; hence, $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ twinning should contribute

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FIGURE 12.9. A Plot of the Crack-Tip Orientation Factors for the $\{10\bar{1}2\} \langle 10\bar{1}\bar{1} \rangle$ Twin System as a Function of θ and ϕ . [These orientation factors have been summed from the six orientation factors resulting from the six possible orientations of $\{10\bar{1}0\}$ poles aligned parallel to the rolling direction (negative orientation factors not included in the sum).]

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FIGURE 12.10. Same as Figure 12.9 (Except these are for TD cracking)

more to the TD toughness. The other twin system, $\{11\bar{2}1\} \langle 11\bar{2}\bar{6} \rangle$, shows similar results with a similar conclusion. The orientation factors for the slip system, $\{10\bar{1}0\} \langle \bar{1}2\bar{1}0 \rangle$, are somewhat different because the magnitudes of the negative orientation factors are included in the sum. The differences in the orientation factor sum between the two cracking directions are not nearly as great, which suggests, also, a more isotropic contribution from slip to the toughness of the material. It is believed that these calculations establish the ex-

planation for the toughness difference between the RD and TD orientations, particularly since no other reasonable explanation for this anisotropy has yet been found.

Further refinement needs to be conducted on several fronts concerning the analysis of the dependence of the toughness of α -zirconium alloys on twinning deformation. Of critical importance, a comparison of the relative contribution to the plastic strain caused by slip and twinning should be made as a function of irradiation-produced defects. The effect

of lattice defects in zirconium alloys in hindering the propagation of twins is not yet known although in the case of bcc metals, it has been shown that impurities can inhibit twinning. The possibility also exists that slip within twins in irradiated grains can occur more easily than in the untwinned lattice because the twinning dislocations--in sweeping through the lattice as the twin grows--may alleviate the strengthening caused by the defects and, hence, contain a softer lattice. Fortunately, twins in α -zirconium do not appear to cause significant amounts of microcrack formation in the matrix (although microcracks are formed in hydrides by twin intersection); but, unless the stresses around a twin are completely relaxed by kink bands or by slip, a twin is always a potential microcrack generator. If they exist, conditions where twinning would cause a reduction in toughness by microcrack formation need to be studied further.

DAMAGE MECHANISMS

F. A. Smidt

The objective of this study is to determine the dislocation-defect interactions that give rise to the changes in mechanical properties produced by neutron bombardment. Past work on this program has consisted of a study of the thermally activated flow process in high-purity iron to determine if the basic mechanism of thermally activated flow was changed by the formation of defect aggregates during irradiation. It was concluded

from experimental studies of the temperature dependence of the flow stress and strain rate sensitivity that the mechanism of thermally activated flow is unchanged but that the athermal component of the flow stress is increased. This increase in the athermal component is basically a consequence of the long range elastic interactions between dislocations and the defect aggregates.

This report describes some additional evidence that the defect aggregates introduced by the irradiation do not change the thermally activated mechanism, although strain rate change tests near room temperature do show a slight change in activation volume. An oversaturation effect at fast fluences above 8×10^{19} n/cm² is also reported and discussed in terms of possible defect configurations. Among other activities during this period, a paper, "Dislocation-Substructure Interactions in Iron and Steel", was presented* at San Francisco in June. Future plans include an extension of these studies to damage mechanisms in pressure vessel steels. Specimens of A533B are now being machined for these studies.

Trapping of Carbon Atoms

One of the more perplexing contradictions of studies of thermally activated flow in irradiated iron is

* U.S.-Japan Seminar on Radiation Damage in Iron and Steel, sponsored by the National Science Foundation. June 23-25, 1968

that temperature change tests indicate no major change in mechanism nor, therefore, in the temperature dependence of the flow stress. However, a rather large change in activation volume is observed at room temperature. This change plus the trend for this activation volume to approach the unirradiated value as deformation progressed and "channels" cleared of defects developed led Laidler and Smidt⁽⁷⁾ to postulate a change in deformation mechanism. Subsequent experiments on the temperature dependence of the flow stress, however, showed that no major change in mechanism occurred⁽⁸⁾, thus leaving a contradiction between the two sets of experiments.

This contradiction has been rationalized in a number of ways. Smidt⁽⁸⁾ has pointed out that the activation volume is an extremely sensitive function of stress in this temperature range, so that a change in the density of mobile dislocations might influence the activation volume. Smidt also pointed out that the changes caused by irradiation were larger than the range normally observed between quenched and aged specimens from the same lot and that the activation volume recovered more rapidly than the hardening. Both observations indicate an interaction with a defect produced by the radiation. Arsenault⁽⁹⁾ examined the possibility of observing a thermally activated deformation controlled by the Fleischer mechanism in a material where the Peierls mechanism was ordinarily controlling; he concluded it would only be observed near T_0 (the temperature where τ^* approaches

zero). Since T_0 in iron occurs around room temperature, it seemed possible that a new rate-controlling mechanism might be observed at room temperature but would not occur as a change in temperature dependence at lower temperatures. However, Smidt pointed out that the change in activation volume was not a function of fluence as might be expected were the Fleischer mechanism actually being observed. Arsenault⁽⁹⁾ also suggested that the changes of V^* with strain may be a consequence of the highly localized deformation in irradiated materials, similar to the changes in true strain rate observed by Schwink and Neuhauser⁽¹⁰⁾ in irradiated copper. This should not influence the results, however, since the activation volume is determined from a ratio of strain rates. If this ratio is proportional to the ratio of crosshead velocities, the results should be valid.

Recent tests of the temperature dependence of the flow stress of Ferrovac iron in the range 200 °C to -70 °C show that carbon (and/or nitrogen) impurities produce a drag stress which influences the flow stress around room temperature. Drag stress influence provides a more satisfying explanation of the mechanism by which irradiation changes the activation volume. Quantitative results are difficult to obtain from a temperature change test because dynamic strain aging causes serrated flow above 60 °C, and from 50 to 10 °C the time between tests influences the magnitude of $\Delta\tau/\Delta T$. However, it was clear that $\Delta\tau/\Delta T$ reached a maximum at about 35 °C and that the drag

stress from this source became inconsequential at about 0 °C.

This effect was not previously considered because Cottrell's theory of atmosphere drag⁽¹¹⁾ predicts a condensed atmosphere in this temperature range. Arsenault⁽¹²⁾ had previously studied stress-induced ordering in creep tests of low-carbon iron and concluded the effect could be observed at creep rates of $2.5 \times 10^{-5} \text{ min}^{-1}$ at 274 °K but would not be observed in tensile tests at common strain rates. Estimates of the plausibility of the drag mechanism using Cottrell's equations for an uncondensed atmosphere at 50 °C were made with the following equations:

$$v_D = \left(\frac{D}{kT}\right)F = \left(\frac{D}{kT}\right)\left(\frac{A}{r^2}\right) = \frac{\dot{\gamma}}{\rho b}$$

where v_D is the drift velocity, D is the diffusivity, F is the force, r is the atmosphere radius, and the other quantities have their usual meanings. A relation between ρ and r was determined from the other experimental parameters, and a value of $A = 1.5 \times 10^{-20} \text{ dyn/cm}^2$ was used. An atmosphere radius of $r = \frac{A}{2kT}$ was consistent with a reasonable dislocation density of $2 \times 10^{11} \text{ cm/cm}^3$. The fraction of carbon atoms condensed on the dislocation line was estimated, and the maximum drag stress was calculated from $\sigma_0 = \frac{17A}{b} \frac{C_0}{b}$. This result gave 2.7 kg/mm^2 , which was of the same magnitude as the serrations, 1.4 kg/mm^2 . The problem of atmosphere drag has recently been studied in detail by Keh et al.⁽¹³⁾ who concluded that such effects do indeed

influence the flow stress of iron-carbon and iron-nitrogen alloys.

Returning now to the application of this information to the interpretation of the changes in activation volume observed in radiation damage experiments, we find a more consistent picture. Figures 12.11 through 12.14 show the changes in activation volume with strain observed in Ferrovac E and Battelle zone-refined iron after various irradiations and in the unirradiated condition. Figures 12.11 and 12.13 have been reported previously⁽⁸⁾ while Figures 12.12 and 12.14 show new data for a higher fluence, $2 \times 10^{20} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) and a higher temperature, 400 °C. First, it should be noted that in the unirradiated controls, Ferrovac E, with more carbon, has a higher activation volume than zone-refined iron (compare Figures 12.12 and 12.13). Quenching both Ferrovac E and BMI iron increases the activation volume slightly while aging lowers the activation volume. Next, note that irradiation to fluences of 1.4×10^{18} , 8.3×10^{19} , and $2 \times 10^{20} \text{ n/cm}^2$ causes about the same initial decrease in activation volume for all fluences for both Ferrovac E (Figures 12.11 and 12.12) and for BMI iron (Figures 12.13 and 12.14). This indicates that the initial decrease in activation volume observed at low strains is caused by the removal of carbon from solution. The interaction of dislocations with the defects where the carbon is trapped releases carbon, and the activation volume approaches that of the unirra-

Neg 0671129-1

FIGURE 12.11. Activation Volume at 295 °K As a Function of Strain for Ferrovac E Iron, Unirradiated, Irradiated to Fast Fluences of $1.4 \times 10^{18} \text{ n/cm}^2$ and $8.3 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) at 60°C and Irradiated to a Fast Fluence of $8.3 \times 10^{19} \text{ n/cm}^2$ at 60°C Followed by a Postirradiation Anneal of 2 hr at 350°C .

Neg 0682162-13

FIGURE 12.12. Activation Volume at 295 °K As a Function of Strain for Ferrovac E Iron, Unirradiated, Irradiated to a Fast Fluence of $2 \times 10^{20} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) at 60°C and Irradiated to a Fast Fluence of $8 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) at 400°C .

Neg 0671129-2

FIGURE 12.13. Activation Volume at 295 °K As a Function of Strain for Battelle Zone Refined Iron, Unirradiated, Irradiated to Fast Fluences of 1.4×10^{18} n/cm² and 8.3×10^{19} n/cm² ($E > 1$ MeV) at 60 °C and Irradiated to a Fast Fluence of 8.3×10^{19} n/cm² at 60 °C Followed by a Post-irradiation Anneal of 2 hr at 350 °C.

Neg 0682162-11

FIGURE 12.14. Activation Volume at 295 °K As a Function of Strain for Battelle Zone Refined Iron, Unirradiated, Irradiated to a Fast Fluence of 2×10^{20} n/cm² ($E > 1$ MeV) at 60 °C and Irradiated to a Fast Fluence of 8×10^{19} n/cm² ($E > 1$ MeV) at 400 °C.

diated sample as the samples are strained. Postirradiation annealing (Figures 12.11 and 12.13) also causes dissociation of the defects and release of carbon and, therefore, the activation volume recovers.

McRickard⁽¹⁴⁾ has reported that dynamic strain aging of irradiated iron requires a prestrain to produce serrated flow curves, which also indicates that carbon is released by the deformation. Also noteworthy is that the recovery of BMI iron usually is greater than the control samples' initial value. This would be consistent with small carbide precipitates going into solution during the irradiation.

Thus, it can be concluded that the activation volume measured at 295 °K is influenced by Cottrell atmosphere drag, and that the changes produced by irradiation are a consequence of the trapping of interstitial carbon and/or nitrogen in the atmosphere by the defects produced during the irradiation. Furthermore, the interaction of moving dislocations with the impurity defect complex destroys the complex and releases the interstitial impurity atoms which again form an atmosphere.

Saturation Effects

Several observations have shown saturation of radiation damage at high exposures; intuitively, one would expect this to occur as the volume of atoms perturbed by displacement spikes approaches the volume of the lattice. Among the relationships suggested to describe the fluence dependence of radiation

hardening are $(\phi)^{1/3}$, observed experimentally at low exposures;⁽¹⁵⁾ $(\phi)^{1/2}$, predicted by Seeger from his theory of hardening by depleted zones;⁽¹⁶⁾ and $A[1-\exp(-B\phi t)]^{1/2}$, suggested by Makin and Minter.⁽¹⁷⁾ The latter expression was derived by assuming new clusters would only be formed in an undamaged volume of the crystal and by using Seeger's estimate to relate number of clusters formed to the increase in yield strength. The constant A is the reciprocal of (depleted zone volume)^{1/2}.

McRickard⁽¹⁸⁾ has recently shown that the saturation of radiation hardening in Ferrovac iron occurs at a fast fluence of 2×10^{19} n/cm² ($E > 1.45$ MeV) with a maximum increase in yield stress of 8.8 kg/mm², fitting Makin and Minter's expression for a damage volume of 4.4×10^{-19} cm³ ($d = 94$ Å).

Recent observations of the increase in yield strength of Ferrovac iron, in the fluence range from 1.4×10^{18} n/cm² to 2×10^{20} n/cm² ($E > 1$ MeV), confirm that saturation occurs with a maximum increase in strength of 8.9 kg/mm², although at a somewhat higher fluence (8×10^{19} n/cm²) than observed by McRickard. The difference in the saturation fluence is probably due to the lower threshold value for neutron damage (1 MeV versus 1.45 MeV). Battelle zone-refined iron also saturates at about the same fluence.

The greatest significance in these two investigations, however, is the observation that oversaturation or softening occurs at fluences beyond saturation. This indicates that not only does the efficiency of

damage production decrease at high exposures, but dynamic recovery can also occur. The degree of recovery is quite substantial, as the data show from the present investigation and McRickard's results (Figure 12.15). Among the mechanisms by which this recovery might take place are loop interaction to form networks, dissolution of precipitates, or a change in the state of aggregation of the defects. The interaction of loops to form dislocation networks has been observed in molybdenum⁽¹⁹⁾ irradiated to a fluence of 5×10^{19}

n/cm^2 at 60 °C. The influence of network formation on the yield strength in molybdenum is not well characterized because of the onset of embrittlement at these exposures. Preliminary examinations of the BMI samples irradiated to a fluence of $2 \times 10^{20} \text{ n/cm}^2$ show no indication of network formation, so this mechanism of oversaturation does not appear to be operative.

The dissolution of small carbide precipitates might also result in a decrease in strength. These precipitates presumably strengthen the

Neg 0682162-9

FIGURE 12.15. Increase in Shear Yield Stress of Iron As a Function of Fast Neutron Fluence Showing the Oversaturation or Softening Observed at High Fluences (by McRickard in Ferrovac E iron and by Smidt in Ferrovac E and Battelle zone refined iron).

material by the Orowan mechanism of precipitation hardening in the same manner postulated for defect clusters. It is well known that precipitation hardening is influenced by precipitate spacing and that maximum hardness results from aging at low temperatures where many small, closely-spaced precipitate particles form. Carbon would appear to play some role in this process because McRickard did not observe oversaturation in an Fe-0.1% C alloy irradiated to comparable fluences. In addition, the increase in yield strength in the Fe-0.1% C alloy was less than for the Ferrovac. Presently, it is not clear whether dissolution of precipitates is an important factor in oversaturation.

A change in the state of aggregation of the defects might result in oversaturation in several ways. The dynamic rearrangement at high fluences could cause a growth of large defect aggregates at the expense of smaller aggregates, with an attendant decrease in the density. One possible effect of this is that the larger aggregate might have a different configuration producing a weaker interaction with the dislocations. Another possibility is that, due to their size, the larger aggregates are less effective hardeners. Smidt and Bement⁽²⁰⁾ have suggested that the Orowan mechanism of hardening is the only one which predicts hardening of the observed magnitude, $\tau = 1/2 \mu b (N_v d)^{1/2}$, where τ is the increase in shear stress, μ is the shear modulus, N_v is the volume density of clustered defects, and d

is the diameter of the cluster. For a fixed number of point defects arranged in spherical clusters, it can be shown that the volume density varies inversely with d^3 . Substituting this result in the hardening equation shows that the hardening will be proportional to $1/d$, so that rearrangement of the same number of point defects into larger spherical clusters will result in a decrease in yield strength.

Unfortunately, the defects observed in iron are so small that their size and type cannot be resolved by transmission electron microscopy with sufficient accuracy to check these predictions.

Perhaps the most informative experiments to date have been Beeler's damage simulation by computer experiments.⁽²¹⁾ Although absolute values of the parameters are sensitive to the potential functions used in the calculations, relative results should be valid, and the general features of the process of radiation damage are revealed. These models confirm the displacement spike-depleted zone model of the primary damage event, showing that most interstitials are produced as single interstitials while small vacancy clusters frequently survive the damage.

Interstitial clusters were observed to form and collapse into loops, but unfortunately the lattice model was not large enough to follow their growth in great enough detail to estimate the density of loops in a saturated damage state. The size of the vacancy clusters increased as subsequent displacement spikes occurred

in the same region previously disrupted. In addition, interstitials and vacancies were created in close proximity to those formed by earlier damage events and the number eliminated by annihilation increased as saturation was approached. Beeler estimated that saturation would occur at about 2.6×10^{19} n/cm² ($E > 1$ MeV) if 50% of the defects annealed out at room temperature. Even though interstitial clusters were not considered, it is safe to assume that they too will grow in size as saturation is approached. It, thus, appears that a growth of large aggregates at the expense of smaller ones is possible, and that a point of saturation is reached where no additional point defects survive the damage event. The rearrangement into larger aggregates could lead to oversaturation or softening simply because of the growth of the aggregate or because of a conversion to a defect less effective in causing hardening.

Three mechanisms have been considered in the above discussion that could produce the oversaturation or softening effects observed in pure iron at exposures above 8×10^{19} n/cm² ($E > 1$ MeV). Network formation was not observed in preliminary studies of iron irradiated to these exposures, so this mechanism can be eliminated. Precipitate dissolution may be important in alloys aged under conditions where many fine precipitates form but is not believed to be important in the present experiments. The dynamic rearrangement of defect-aggregates into larger clusters, with an attendant decrease

in the number of point defects surviving the damage event at high fluences, appears to be the best explanation of oversaturation.

HIGH PRESSURE STUDIES

G. L. Kulcinski

The purpose of this phase of the program is to study the effects of ultrahigh pressures (1000 to 60,000 atm) on the production, retention, and subsequent microscopic configuration of the damage produced in irradiated solids. In addition, the high-pressure synthesis of new phases and the high-pressure alteration of mechanical properties will be investigated as possible methods of providing more radiation damage-resistant materials for nuclear reactors.

Irradiation of Materials Under Pressure

The effect of stress on irradiation-promoted phase transitions in zirconium dioxide, which has both an irradiation- and a pressure-induced phase transition, is being studied. Monoclinic ZrO₂ doped with natural uranium has been found to convert to a tetragonal (or pseudo-cubic) structure under irradiation.⁽²²⁾ Whittles et al.⁽²²⁾ have shown that detectable amounts of conversion take place during exposures as low as 1.1×10^{15} fissions/cm³, while the transformation is complete by 2.4×10^{16} fissions/cm³. As reported previously, monoclinic ZrO₂ can be converted to

the tetragonal form at room temperature and 37 kbars.⁽²³⁾ The pressure-induced transition was also shown to be reversible, and the tetragonal structure could not be retained at one bar and room temperature.

Since the tetragonal phase is more dense than the monoclinic structure, it was postulated that the application of pressure during irradiation might accelerate the irradiation-induced transition; or from another viewpoint, irradiating during compression might lower the transformation pressure at room temperature. To test this hypothesis, monoclinic ZrO_2 doped with 800 ppm of natural uranium was irradiated to 1.5×10^{15} fissions/cm³ while under 24 kbars of pressure and no restraint.

The sample material, high-pressure cell, and analytical techniques have been previously described.⁽²⁴⁾ X-ray patterns of the sample at 1 bar and under 24 kbars pressure showed the crystal structure to be monoclinic before irradiation (Figures 12.16a and 12.16b). The pressure cell, with the sample still under pressure, was then encapsulated along with 1 g of starting material in an aluminum irradiation capsule. The assembly was irradiated to 1.5×10^{15} fissions/cm³ at $\sim 40^\circ\text{C}$ in a Hanford production reactor.

After an appropriate decay period, the X-ray pattern of the sample was obtained while still under 24 kbars of pressure. Because the radiation-induced fogging of the X-ray film was so extensive, a 1 in. thick lead

shield was placed in the camera⁽²⁵⁾ between the pressure cell and the film. A small slit was machined into the shield to allow the diffracted X-rays to strike the film. Figure 12.16c represents a typical X-ray pattern of the sample after irradiation, while it was still under 24 kbars of pressure.

Comparison of the X-ray pattern in Figure 12.16c with previously established standard X-rays of known monoclinic-tetragonal mixtures reveals that approximately 30% of the monoclinic phase was converted to the tetragonal structure. Analysis of the powder irradiated to the same exposure at only 1 bar pressure revealed that essentially no (<10%) tetragonal phase had been formed.

Not only did the ZrO_2 show enhanced conversion under pressure, but the X-ray patterns, obtained after the pressure was released, revealed that the metastable tetragonal phase was retained at ambient conditions (Figure 12.16d). This is particularly significant considering that the pressure-induced transition had been shown to be completely reversible.

Although the results are preliminary in nature, it is obvious that the application of pressure during irradiation does accelerate the fission fragment-induced phase transition in ZrO_2 . Irradiations at higher fluences are being conducted to more clearly define the nature of this effect.

		Pressure kbar	% Monoclinic	% Tetragonal
a)		0.001	100	0
	↓ Apply Pressure			
b)		24	100	0
	↓ Irradiation - 1.5×10^{15} fissions/cc			
c)		24	70	30
	↓ Release Pressure			
d)		0.001	70	30

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FIGURE 12.16. X-Ray Analysis of ZrO_2 Doped with 800 ppm Uranium

a) Unirradiated, Uncompressed

b) Unirradiated Under 24 kbar

c) Irradiated to 1.5×10^{15} fissions/cm³ Under 24 kbar

d) Sample in c) After Release of Pressure

IN-REACTOR CORROSION OF ZIRCONIUM ALLOYS

A. B. Johnson, Jr.

In-flux corrosion and hydrogen absorption data were reported previously⁽²⁵⁾ for seven specimens representing three lots of Zircaloy-2 in various prefilmed and heat treatment conditions. The specimens were exposed in the Engineering Test Reactor G-7 Loop in pH-10 NH_4OH . Specimens from the same three lots were exposed

in a previous G-7 loop test series in pH-10 $LiOH$, ~ 1 ppm O_2 .⁽²⁶⁾ The data from the pH-10 NH_4OH system generally showed relatively low hydrogen absorption. However, one lot HT sample that had a thick prefilm oxide (~ 250 mg/dm²) showed a large hydrogen pickup in terms of ppm and percent. Additional specimens from the ammoniated test have now been sampled for hydrogen. The complete data, including those previously reported,⁽²⁵⁾ appear in Table 12.4.

TABLE 12.4. In-Flux Hydrogen Absorption in the ETR G-7 Loop in pH-10 NH_4OH (a)

		As-Etched		Prefilmed		
		As-Fab.	β -Quenched	300°	400 °C-72 hr	400 °C-210 d
ppm	AT	9	14	14 ^(b)	28	241
	CT	19	3	12 ^(b)	34	72
	HT	24	15	--	49	501
%	AT	28	43	14 ^(b)	--	47
	CT	51	9	10 ^(b)	18	16
	HT	51	31	--	--	97

(a) Exposure Conditions: 131 days, $\sim 1 \times 10^{14}$ n/cm² sec; 1.7×10^{21} n/cm > 1 MeV; 270-280 °C.

(b) Corresponding values from the pH-10 LiOH G-7 loop exposure are as follows: AT - 46 ppm (10%); CT - 23 ppm (4%).

The data in Table 12.4 show relatively low hydrogen pickup for the six as-etched specimens. Specimens prefilmed at 300 °C also picked up relatively little hydrogen in-flux; the corresponding percentage values were in reasonable agreement with values from specimens with a 300 °C prefilm exposed in the lithiated G-7 loop system. Specimens prefilmed at 400 °C showed moderate increases in hydrogen absorption over the as-etched and 300 °C prefilm conditions. However, hydrogen absorption on specimens with a thick prefilm were very much larger than the corresponding comparison samples in terms of ppm. In terms of percentage, two of the three values are high (AT and HT) relative to values for the comparison samples.

Viewed in the context of previous G-7 loop hydrogen absorption results,^(26,27) those in Table 12.4 are reasonably consistent. Hydrogen

absorption on as-etched specimens in the ammoniated system has generally been low⁽²⁷⁾ in agreement with values in Table 12.4. In terms of percentage, the values for 300 °C-prefilm samples are in reasonable agreement in the two systems (Table 12.4). In the aggressive lithiated G-7 loop system, hydriding of the three Zircaloy-2 lots increased in the following order: CT < AT < HT. Values for the thick-film samples in Table 12.4 follow the same order. Transfer of specimens from the lithiated to the ammoniated G-7 loop system provided evidence that a combination of a thick oxide and the in-flux ammoniated coolant would cause increased hydrogen absorption.⁽²⁶⁾ The response of the thick-film samples exposed in the ammoniated system also indicated a thick-film/coolant interaction. Percentage values were

higher on the thick-film samples than on corresponding specimens exposed only in the lithiated system.⁽²⁶⁾

The conclusions of the hydriding examination are:

- Specimens exposed as-etched or with a thin prefilm did not undergo marked flux-induced hydriding in an ammoniated coolant under normal exposure conditions.
- Ammoniated (<0.05 ppm oxygen) and lithiated (~1 ppm O₂) G-7 loop environments had substantially different corrosion properties⁽²⁵⁻²⁷⁾ but the proportion of hydrogen absorbed was not appreciably different on specimens having the same prefilm treatment (300 °C water).
- A combination of a thick film and an ammoniated coolant (~45 cc H₂/kg of coolant) appears to increase the absolute and proportional amount of hydrogen absorbed.
- Under accelerated hydriding conditions, Zircaloy-2 lots differ in susceptibility to hydriding.

EVALUATION OF ZIRCONIUM-2.5 WT% NIOBIUM PRESSURE TUBING

P. J. Pankaskie

The objective of this program is to evaluate zirconium-2.5 wt% Nb alloy tubing as a pressure tube material with reference to tests and reactor experience that has been obtained with Zircaloy-2 pressure tubes. Creep and stress-rupture testing, crack growth and propagation studies and in-reactor service evaluation are the major activities in this effort.

A series of creep and stress-rupture tests on zirconium-2.5 wt% Nb tubing at 300 and 400 °C have been in progress. Tubing in the cold-worked and two heat-treated conditions have been used in these tests. One heat treatment consisted of an 880 °C soak, water quench, and aging for 24 hr at 500 °C and the other consisted of an 880 °C soak, water quench, 15% cold reduction, and aging for 24 hr at 500 °C. All creep tests were done in air. The series of creep and stress-rupture tests are complete except for further testing of heat treated tubing at 300 °C.

The results of the creep and stress rupture testing are tabulated in Table 12.5 and shown graphically in Figure 12.17. As seen in Figure 12.17, at 400 °C essentially no difference is apparent in the creep and stress-rupture properties of the two heat treatments. The addition of the 15% cold reduction between the water quench and aging does not appear to provide added creep strength nor extend rupture time at 400 °C. Only one cold-worked tube was tested at 400 °C. This tube had previously been tested at 300 °C for 10,725 hr and had sustained an average strain of about 1.3%. It subsequently failed in 660 hr, which is slightly less than the predicted rupture time for heat treated tubing at comparable hoop stresses.

In those tests where failure occurred, the onset of tertiary creep and rupture is accompanied by localized deformation (Figure 12.18). In those long-term tests wherein local bulging occurs, there is generally

TABLE 12.5. Tubular Stress-Rupture Tests on Zirconium-2.5 wt% Nb Alloy

Specimen Identity	Test Temperature, °C	Hoop Stress, ksi	Test Duration, hr	Total Strain, %	Average Secondary Creep Rate, %/hr
30-2-SR	300	61	1,453 (failed)	>2.4	8×10^{-4}
30-3-SR	300	36	10,725 (disc)	~1.3	5×10^{-6}
	400	25	660 (failed)	3.7 to 10.0	5×10^{-3}
30-11-SR	300	50	11,984 (disc)	2.1	4×10^{-4}
HT-1	400	30	3,176 (failed)	1.1 to 10.1	1×10^{-3}
HT-2	400	25	14,736 (failed)	>3.5	5×10^{-4}
HT-3	400	20	6,784 (in test)	0.75	---
HT-7	400	16.5	~7,250 (failed)	>3.1	2×10^{-4}
HT-1A	400	40	27 (failed)	6.5 to 7.2	---
HT-2A	400	30	295 (failed)	2.7 to 8.3	2×10^{-3}
HT-3A	400	25	742 (failed)	>2.2	3×10^{-4}
HT-4A	400	15	5,720 (in test)	2.8 to 10.0	---

HT-1 material was 60% cold-worked, solution heat-treated at 880 °C, water-quenched and aged 24 hr at 500 °C.

HT-1A material was solution heat-treated at 880 °C, water-quenched, 15% cold-worked and aged 24 hr at 500 °C.

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FIGURE 12.17. Creep Rate and Rupture Times Versus Stress for Zirconium-2.5 wt% Nb Alloy Pressure Tubing

Neg 0683288-2

FIGURE 12.18. Typical Creep Failure in Zirconium-2.5 wt% Nb Alloy Tubing

little or no further creep deformation in areas other than the bulged area. The uniform creep strain generally ranges from about 2 to 3%. In the bulged area where fracture occurs, creep strains range from about 7 to 10%.

Crack growth and propagation investigations have been temporarily suspended pending receipt of test material from CRNL. In the last two of a series of tests on heat treated tubing after hydriding, very low fracture toughness was observed. Unstable fracture was observed at hoop stresses of 14,000 to 15,000 psi and crack lengths of less than 1/2 in. For this reason PRTR Test 148, which was to provide for the installation and operation of a heat treated zirconium-2.5 wt% Nb pressure tube as a normal process channel in the PRTR, is being held in abeyance until the fracture properties of this specific material are more clearly defined. The test material from CRNL will be used to resolve the question of ade-

quate fracture toughness in heat treated tubing after hydriding.

The four cold-worked zirconium-2.5 wt% Nb tubular test specimens being irradiated in the PRTR for postirradiation crack growth and propagation tests have accumulated a fast fluence of about 9×10^{19} n/cm². The cold-worked zirconium-2.5 wt% pressure tube being irradiated under PRTR Test 125 has reached a maximum fast fluence of 6.5×10^{20} n/cm².

FAST REACTOR SUPPORTING STUDIES

SWELLING STUDIES ON STRUCTURAL MATERIALS

J. J. Holmes

Recent experiments^(28,29) concerning the effects of fast reactor irradiation on stainless steels have demonstrated that a considerable volume change occurs during neutron bombardment. Unlike volume changes in fuel materials, swelling in structural alloys and pure metals is caused by

the condensation of irradiation-produced excess vacancies in the form of three-dimensional cavities. Cawthorne and Fulton⁽²⁸⁾ found as much as 0.8% volume change in 316 SS irradiated to 7.8×10^{22} n/cm² total fluence. Volume changes of this magnitude, if not taken into account in component design, are sufficient to cause catastrophic distortion in reactor components. Unfortunately, existing swelling data are too fragmentary to provide a useful relationship between density, fluence, and irradiation temperature.

The objective of this effort is to develop a description of the swelling process that can be used in FFTF and LMFBR designs. The upper fluence limit, which can be achieved in reasonable time with existing technology, is about 10^{23} n/cm². Fast reactor applications, however, range as high as 10^{24} n/cm². Therefore, any useful swelling relationship must be based on extrapolation of low-fluence data to high-fluence applications. The present approach in this study is to obtain the required low-fluence experimental data and develop physical models, based on density measurements and transmission electron microscopy, that can be used to extend the data.

Since the swelling phenomena have only recently been found, no experiments are currently in progress specifically aimed at obtaining swelling data. Experiments are currently being designed for this purpose and should begin in January, 1969. In the meantime, early swelling data are being developed through the examination of various EBR-II components. In particular, a 304L SS control rod

thimble (5C3) and a 304L SS fuels-irradiation capsule (X-011) have been examined in detail. Both were irradiated at temperatures ranging from 704 to 866 °F. The peak fast fluence for the thimble was 4.8×10^{22} n/cm² ($E > 0.1$ MeV); that for the capsule was 2.3×10^{22} n/cm² ($E > 0.1$ MeV).

Density measurements were made on specimens taken at various positions along these components to take advantage of the different fluences and temperatures along their lengths. The densities were determined by an Archimedes method that utilized CCL₄ as the immersion medium. The 95% confidence limits for all the data was $\pm 0.05\%$. The base (unirradiated density) was determined by two techniques: one that involved annealing the density specimen until all voids were removed (thought to be the point where no further density decrease occurs with increasing annealing temperature); a second that involved measuring the density of archives specimens. The two techniques agreed well for the capsule. However, for the control rod thimble, the annealed density was 7.898 ± 0.002 g/cm³ while the density of the archives specimen was 7.905 ± 0.002 g/cm³. The reason for this discrepancy is not apparent at this time. For comparison purposes, we take the annealed density at the "true" base density.

Volume change data for the capsule and thimble core are presented in Figure 12.19 together with all other available swelling data. (PNL data are also summarized in Table 12.6.) The Dounreay data have been multiplied by 1.6 to correlate with EBR-II

FIGURE 12.19. Summary of Available Data on Fast Reactor-Induced Swelling

calculated fluences. It is thought that calculated EBR-II fluences are greater by a factor of 1.6 than actual dosimetry data.⁽³⁰⁾ The data, plotted in logarithmic coordinates, suggest a concave upward relationship between density and fluence. However, the volume change appears to be independent of fluence below about 0.4×10^{22} n/cm², suggesting that volume changes in this region are caused by some process other than void growth. Possibly, the indicated swelling values in this range are due to residual error in the technique or surface effects caused by long sodium exposure. If the swelling below 0.4×10^{22} n/cm² is subtracted from all PNL values (subtract 0.05% from all PNL points),

TABLE 12.6. Summary of PNL Swelling Data

Component	Fluence, (E > 0.1 MeV)	Irradiation Temperature, °F	ΔV/V, %	Sample No.	ΔV/V-0.05, %
Capsule (X-011)	1.9×10^{22} n/cm ²	760	0.13	--	0.07
304L SS	2.3	786	0.27	--	0.22
	2.3	819	0.33	--	0.28
	1.3	840	0.34	--	0.29
	0.8	844	0.13	--	0.08
Control Rod	1.16	704	0.12	A	0.07
Thimble (5C3)	2.81	704	0.24	B	0.19
304L SS	3.96	728	0.51	C	0.46
	1.17	792	1.15	D	1.10
	3.06	866	0.88	E	0.83
	0.70	866	0.063	F	0.013
	0.027	866	0.127	G	0.077
	1.70	704	0.29	2M	0.24
	2.18	866	0.31	2Q-1	0.26
	1.00	866	0.19	2Q-2	0.14
	0.40	866	0.05	2R-1	0
	0.25	866	0.05	2R-2	0
	0.10	866	0.05	2R-3	0
Tensile Specimen (X-009)	1.6	1000	0.24	--	0.19
304 SS					

a linear relationship between fluence and volume change results (Figure 12.20). Extrapolation to target FFTF fuel clad fluences of 1.3×10^{23} n/cm² indicates about a 20% volume

increase. Volume increase in excess of 100% are predicted at calculated EBR-II fluences greater than 4×10^{23} (or probable dosimetry fluences of $4/1.6 \times 10^{23} = 2.5 \times 10^{23}$ n/cm²).

FIGURE 12.20. Data in Figure 12.19 Corrected for 0.05% Residual Error in PNL Values

Extrapolation of the swelling data in this manner is not necessarily valid. It is not yet known that swelling is a function of integrated flux. In reality, flux and time may need to be considered independently. Preliminary attempts at modeling the swelling process at PNL suggest $\frac{\Delta V}{V} \propto \phi^n t^m$, where n is not necessarily equal to m ; although, in some cases $m = n$. Until the mechanism of swelling is better understood, the above extrapolation should be considered as one among many yet unexplored possibilities.

TRANSMISSION ELECTRON MICROSCOPY OF IRRADIATED AUSTENITIC STAINLESS STEEL

H. R. Brager

Transmission electron microscopy studies on austenitic stainless steels are being conducted to provide information for interpreting fast reactor irradiation effects, for formulating material irradiation behavior models, and for improving the performance of austenitic stainless steel in reactor environments.

The electron micrographs of the Type 304L SS samples cut from an EBR-II control rod thimble, published in the previous report,⁽¹⁾ are being analyzed. The specimens have been subjected to fast fluences ranging from 3×10^{20} to 5×10^{22} n/cm² ($E > 0.1$ MeV) at temperatures from 700 to 870 °F (371 to 466 °C). To observe cavities in highly irradiated austenitic stainless steels, it is necessary to adjust the matrix crystal orientation to minimize the

contrast due to the extensive displacement damage in the microstructure. For this diffraction condition, the transmitted beam intensity is a function of the foil thickness. The cavity distribution of the Type 304L SS irradiated at 790 °F (421 °C) to 5×10^{22} n/cm² was determined for foil thicknesses ranging from 250 to 1200 Å, Figure 12.21. The mean diameter of the cavity distribution tended toward larger values for thicker foils. These results appear to be consistent in that, with the transmitted beam intensity being a function of the foil thickness, the very small cavities (~ 100 Å) would be difficult to observe in thick foils (≥ 1000 Å), and the measured distribution would be biased toward larger diameter cavities. However, the cavity distribution measured in thin foils (~ 200 -300 Å) containing cavities whose size approaches the thickness of the foil would, as observed, be expected to be biased toward the smaller sizes. Therefore, it appears that the determination of the cavity distribution in highly irradiated steels requires measurements upon foils having significantly different thicknesses.

The cavities in irradiated steel should appear as bright regions in bright field electron micrographs. Such bright regions are observed but are surrounded by a dark border. The dark border around the cavities has not, as yet, been explained by conventional diffraction theory. This diffraction condition places a lower limit of about 90 Å on the resolution of the cavities. Below this value,

Neg 0682654

FIGURE 12.21. As-Irradiated Cavity Distribution of Type 304L SS Irradiated in the EBR-II at 790 °F (421 °C) to a Fast Fluence of 4.8×10^{22} n/cm² ($E > 0.1$ MeV) Measured in Foils of Thickness from 250 to 1200 Å.

the variation in intensity of the bright center region and the dark border tend to blend together and are not discernible.

It appears that the "true" diameter of the irradiation-produced cavity is the outside diameter of the black border surrounding the cavity. The validity of this statement has been confirmed by calculations of the expected change in the density of the irradiated specimens based on the observed cavities. The "microscopic" density change of the Type 304L SS irradiated at 790 °F (421 °C) to 5×10^{22} n/cm² was calculated to be

0.5% or 1.0%, depending on whether the outside diameter of the bright interior or the dark border of the cavities, respectively, was used in the calculations. The bulk density change of the same sample was measured to be $1.17 \pm 0.04\%$.

Analysis of the electron diffraction patterns from areas such as that shown by the dark field micrograph (Figure 12.22) has revealed that the small precipitates (50-300 Å) present at a high concentration ($\sim 3 \times 10^{15}$ cm³) are M₂₃C₆. The production of this fine coherent phase is probably caused by dynamic

Neg 5820-C

96,000X

FIGURE 12.22. Dark-Field Electron Micrograph of $M_{23}C_6$ Precipitates in Type 304L SS Irradiated in the EBR-II at 750 °F (399 °C) to a Fast Fluence of 4.0×10^{22} n/cm² ($E > 0.1$ MeV).

re-solution and precipitation of particles in the 304L SS during irradiation--in this instance, 4×10^{22} n/cm² ($E > 0.1$ MeV) at 750 °F (399 °C)--in a high neutron flux. A sample of the steel irradiated to a fast fluence of only about 3×10^{20} n/cm² ($E > 0.1$ MeV) for the same period of time contained only a moderate concentration ($\sim 10^{13}$ /cm³) of much larger (0.1-0.5 μ diam) M₂₃C₆ particles.

The diffraction pattern of a second kind of precipitate has been indexed in this highly irradiated steel and has been identified to have a bcc structure with a lattice spacing of 6.9 Å. The spacing of this phase does not correspond to the crystal structure of any normally expected intermetallic phase.⁽³¹⁾ It is much larger than the 3.6 Å lattice spacing of the Type 304L SS fcc matrix structure.

A sample of the Type 304L SS that had received a fast fluence of 4.8×10^{22} n/cm² ($E > 0.1$ MeV) at 790 °F (421 °C) was vacuum-annealed 1 hr at 1200 °F (649 °C). Annealing was done to determine the effect of postirradiation heat treatment on the cavities and the general microstructure; 1 hr at 1200 °F was chosen to scope future annealing conditions. Preliminary analysis indicated that the cavity size and distribution did not change significantly due to this postirradiation anneal. However, the microstructure was affected. The as-irradiated displacement damage and faulted loop substructure (Figure 12.23) was transformed into a random dislocation network (Figure 12.24). This result is substan-

tially identical to that observed for fully annealed Type 304 SS that had been irradiated at 1000 °F (538 °C) to 1.4×10^{22} n/cm² ($E > 0.1$ MeV) in the EBR-II and then postirradiation annealed 1 hr at 1200 °F (649 °C).⁽³²⁾

EBR-II IRRADIATIONS - TENSILE TESTING

R. E. Robbins

The objectives of this program are to determine the combined effects of fast reactor irradiation and environment on the tensile properties of candidate stainless steel fast reactor vessel and core structural alloys and to evaluate the metallurgical stability of these materials after aging at elevated temperatures.

AISI Type 316 SS tensile specimens irradiated at about 900 °F to a total fluence of 0.8×10^{22} n/cm² in the first EBR-II Row 7 subassembly have been tested at temperatures from RT to 1400 °F at strain rates of 0.002 and 0.2/min. The tensile properties of these specimens, together with those for unirradiated as-fabricated specimens, are given in Table 12.7. The data are presently being analyzed; however, several comments may be made. Increased yield strength due to irradiation was observed at all test temperatures. However, the postirradiation yield-strength values at the lower temperatures are much less than those previously reported for 304 SS.⁽⁸⁾ The effect of strain rate on the strength properties is quite small but greatly influences the ductility values (Table 12.7).

Tensile specimens and uniaxial and biaxial stress-rupture specimens are

Neg 5673-D

96,000X

FIGURE 12.23. Bright Field Electron Micrograph of Type 304L SS Irradiated in the EBR-II at 790 °F (421 °C) to a Fast Fluence of 4.8×10^{22} n/cm² ($E > 0.1$ MeV).

Neg 5903-F

96,000X

FIGURE 12.24. Bright Field Electron Micrograph of Same Material Shown in Figure 12.23. After a 1 hr Vacuum Anneal at 1200 °F (649 °C).

TABLE 12.7. Tensile Properties of AISI Type 316 Stainless Steel Irradiated to a Total Fluence of 0.8×10^{22} n/cm² at 900 °F

Test Temperature, °F	Strain Rate	PEL, ksi	0.2% YS, ksi	UTS, ksi	Elongation, %		Reduction of Area, %
					Uniform	Total	
<u>Irradiated</u>							
RT	0.002/min	31.70	42.85	85.04	39.24	46.41	65.04
600		23.64	31.98	71.32	24.72	30.32	59.50
800		23.98	31.30	72.20	22.72	27.60	54.47
900		19.92	29.11	67.64	28.87	33.72	47.97
1000		27.24	33.82	63.33	18.13	19.40	35.77
1100		22.36	33.58	54.07	10.99	12.12	30.89
1200		18.70	31.87	43.90	4.63	5.14	22.76
1400		16.26	24.39	26.50	2.42	5.59	8.13
1000	0.2/min	19.2	28.70	63.52	17.82	23.21	53.60
1100		19.11	29.02	60.73	22.07	32.06	60.16
1200		20.33	30.08	55.61	20.70	28.21	44.72
1400		18.70	26.50	38.86	8.21	16.28	25.20
<u>Unirradiated</u>							
RT	0.002/min	15.3	34.85	77.0	63.80	76.0	77.10
572		19.2	25.9	85.5	47.6	56.5	78.6
700		12.2	18.4	70.6	39.2	44.4	66.2
800		8.5	11.9	65.7	41.8	49.4	60.7
900		11.9	16.7	71.1	39.3	46.9	61.7
1000		12.4	16.9	68.7	46.2	52.8	61.7
1100		12.9	15.9	64.7	42.7	50.6	60.7
1200		11.7	14.3	55.7	35.6	40.1	43.8
1300		12.4	16.7	44.0	23.9	35.3	41.8
1400		10.0	14.0	30.8	14.9	53.8	62.7
1500		14.4	16.3	24.9	10.5	55.9	60.7

being aged in sodium at temperatures from 800 to 1400 °F as part of the aging studies program. Exposure duration will correspond to the in-reactor time of the first EBR-II Row 7 subassembly so that controls for the irradiated specimens will also be provided.

UNIAXIAL CREEP-RUPTURE STUDIES

A. J. Lovell and G. A. Dunagan

The objectives of this program are to evaluate the effects of fast neutron irradiation and environment on the uniaxial creep-rupture properties of candidate vessel and core structural materials for the FTR and to use the information obtained to establish the usable materials limits

to assure their safe and reliable performance.

The uniaxial creep testing of as-fabricated controls is continuing in support of tests to be performed on irradiated specimens from the EBR-II. The emphasis has been placed on obtaining high creep rate (low rupture time) tests on 316 SS from the CANEL heat in the 1950 °F/1 hr solution-treated condition. The data obtained to date on 316 SS are presented in Table 12.8. That obtained from 1100 and 1202 °F tests agree very well with the nationally published data.⁽³³⁾ There are little creep-rupture data for 316 SS at temperatures as low as 1000 °F because the stresses needed to obtain rupture

TABLE 12.8. Uniaxial Creep Data for 316 SS Solution-Treated at 1950 °F/1 hr

Specimen No.	Temperature, °F	Stress, psi	Minimum Creep Rate, in./hr	Time to Rupture, hr	Elongation, %	Reduction of Area, %
M-20/CSR-8027	1000	40,000	0.00009	263.1	14.6	22.8
M-21/CSR-8028	1000	58,000	0.000188	203.9	17.8	29.7
M-22/CSR-8029	1000	60,000	0.000435	129.6	19.9	22.6
M-23/CSR-8030	1000	63,000	0.0010	60.5	23.9	31.4
M-24/CSR-8031	1000	64,000		40 sec	24.0	47.5
M-25/CSR-8032	1100	38,000	0.00054	157.8	13.1	23.1
M-17/CSR-8024	1100	50,000	0.01090	11.2	18.8	26.0
M-14/CSR-8019	1202	25,000	0.00112	373.5	65.7	36.0
M-16/CSR-8023	1202	28,000	0.00226	168.4	30.7	52.3
M-18/CSR-8025	1202	35,000	0.0102	24.9	29.3	53.6
M-19/CSR-8026	1202	22,000	0.00019	1294.2	20.6	11.2

lives of less than 100 hr are greater than 95% of the ultimate strength of the alloy and because of the considerable plastic strain during loading that is associated with the 1000 °F test. A plot of log-stress versus log-time to rupture indicates a nonlinear relation. There is less stress sensitivity at the higher rupture times.

The testing of 304 SS specimens irradiated in space-available pins in the EBR-II and in the hot water loop of the ETR is continuing. The creep-rupture data obtained to date are presented in Table 12.9. Though post-test elongation measurements on tested specimens are not complete, measurements made on the pull-rods during the tests indicate all irradiated specimens suffered considerable loss in total elongation. Deformation (taken from pull-rod motion) versus time data for the 1000 °F tests indicate that irradiation in the EBR-II (to an estimated total fluence of 4×10^{21} n/cm²) has

little effect on the creep rate. In comparison, specimens that were irradiated to a fast fluence of $\sim 8 \times 10^{20}$ n/cm² ($E > 1$ MeV) in a thermal (ETR) reactor at 540 °F, then tested at 1000 °F, show reduced creep rates compared to as-fabricated controls.

Three auto-stress test units and one mini-stress unit were fabricated and placed into service. Two other mini-stress and 10 auto-stress units are partially fabricated. The prototype inert-gas test chamber for use in the CANEL stress-rupture units in 321A was evaluated and found to have an adequate isothermal temperature profile. The stress versus rupture-life data for 304 SS at 1000 °F are shown in Figure 12.25. Considering the relatively low fluences involved, the lowering of rupture life of specimens in both irradiation conditions is marked.

Specimens of 304 SS irradiated in EBR-II space-available pins to a fast fluence of $\sim 1.4 \times 10^{22}$ n/cm² at about 900 °F have been tested at a stress

TABLE 12.9. Uniaxial Creep-Rupture Data on Irradiated 304 SS

Specimen No.	Fluence, n/cm ²	Reactor	Test Temperature, °F	Stress, psi	Minimum Creep Rate, in./in./hr	Rupture Time, hr
GEH-20-280-1	$\sim 8 \times 10^{20}$	ETR	1000	45,000	$< 1 \times 10^{-4}$ (6.0×10^{-4})	19.19 (88.2)
GEH-20-280-2	$\sim 8 \times 10^{20}$	ETR	1000	40,000	1×10^{-4} (1.08×10^{-4})	37.15 (217.9)
A-38-5	$\sim 4 \times 10^{21}$	EBR-II	1000	45,000	1.3×10^{-3} (6.0×10^{-4})	25.33 (88.2)
A-38-6	$\sim 4 \times 10^{21}$	EBR-II	1000	40,000	5×10^{-4} (1.08×10^{-4})	57.05 (217.9)
A-6-5	$\sim 1.4 \times 10^{22}$	EBR-II	1250	20,000	1.6×10^{-3} (4.2×10^{-3})	21.9 (83)
A-2-2	$\sim 1.4 \times 10^{22}$	EBR-II	1300	20,000	8.9×10^{-3} (1.2×10^{-2})	5.9 (25)
A-2-3	$\sim 1.4 \times 10^{22}$	EBR-II	1350	20,000	2.9×10^{-2} (4.5×10^{-2})	1.4 (7.7)
A-1-3	$\sim 1.4 \times 10^{22}$	EBR-II	1400	20,000	1.7×10^{-1} (1.3×10^{-1})	0.2 (2.94)

() As-fabricated control

Neg 0683374-6

FIGURE 12.25. Rupture Data for 304 SS at 1000 °F

of 20,000 psi and various test temperatures in an effort to evaluate the temperature dependence of the creep properties (Table 12.9). The deformation versus time curves for tests at 1250, 1300, 1350, and 1400 °F are shown in Figures 12.26-12.29 for as-fabricated controls and irradiated specimens. The curves indicate that irradiation at 900 °F in the EBR-II decreased the creep rate for postirradiation tests at 1250, 1300, and 1350 °F but possibly increased the creep rate at 1400 °F. An Arrhenius plot of the creep rate data is shown in Figure 12.30. The few data points available indicate in-

creased activation energy for the irradiated specimens over the as-fabricated controls. There are insufficient data to make any conclusions but indications are that:

- EBR-II irradiation to a fast fluence of 1.1×10^{22} n/cm² decreases the postirradiation creep rate of 304 SS for tests below 1350 °F.
- The activation energy for creep in EBR-II irradiated 304 SS steel is increased over that of as-fabricated controls.
- The irradiation-induced defects effecting the creep rate are removed by heating to 1400 °F.

Neg 0683374-4

FIGURE 12.26. Deformation Versus Time Curve at 1250 °F for 304 SS Irradiated at ~900 °F to a Fast Fluence of Approximately 1.1×10^{22} n/cm²

Neg 0683374-1

FIGURE 12.27. Deformation Versus Time Curve at 1300 °F for 304 SS Irradiated at ~900 °F to a Fast Fluence of Approximately 1.1×10^{22} n/cm²

Neg 0683374-3

FIGURE 12.28. Deformation Versus Time Curve at 1350 °F for 304 SS Irradiated at ~900 °F to a Fast Fluence of Approximately 1.1×10^{22} n/cm²

Neg 0683374-2

FIGURE 12.29. Deformation Versus Time Curve at 1400 °F for 304 SS Irradiated at ~900 °F to a Fast Fluence of Approximately 1.1×10^{22} n/cm²

Neg. 0683374-5

FIGURE 12.30. Arrhenius Plot of As-Fabricated Control and Postirradiation Creep Tests on 304 SS

BIAXIAL CREEP RUPTURE STUDIES

R. W. Barker

Because of expected difficulties in the cleaning of biaxial test specimens irradiated in EBR-II, it has been decided that no attempts will be made to remove the sodium from the internal cavities of these biaxial test specimens. Instead, the specimens will be extracted from their sodium-filled irradiation capsules and capped in an inert atmosphere. Tests are currently being performed to determine the feasibility of utilizing a hot cell glove-box to disassemble the capsules and cap the specimens. Whether the irradiated sodium can be salvaged for post-irradiation chemical analysis, and the degree to which the sodium will

become contaminated as a result of the required transfer procedure, will also be determined.

These preliminary efforts are being conducted at Idaho Nuclear Corporation (INC) at their test reactor area (TAN) site at the National Reactor Testing Station (NRTS), Idaho Falls, Idaho. INC has an inert atmosphere glove-box that can be placed in a hot cell and operated remotely. The glove-box is not instrumented for monitoring of the atmospheric conditions; therefore, the quality of the atmosphere must be determined by other methods. It has been determined that atmosphere purities of <200 ppm oxygen are attainable; with additional improvements, it is expected that the impurity level of oxygen can be reduced to <100 ppm. At that time, dummy sodium capsules will be opened and processed to determine the degree of contamination of the sodium by the ~100 ppm oxygen atmosphere.

Once the specimen has been extracted from its irradiation capsule and capped, the specimen exterior may then be easily cleaned. Prior to test, the cap will be removed

and the specimen attached to the biaxial test rig in an inert atmosphere--with the pure sodium remaining in the specimen cavity. On this basis, initiation of the biaxial stress-rupture program is expected by October 15, 1968.

The objective of the program is to determine the effects of stress, temperature, and fast neutron irradiation on the biaxial creep-rupture properties of fuel cladding material. The portion of the program to be initiated this fall will determine these properties on Type 304 and 316 SS fast fluences of irradiated to 0.5, 0.8, and 1×10^{22} n/cm² ($E > 0.1$ MeV) at ~ 1000 °F in Row 7 of the EBR-II. The test outline is shown below in Table 12.10.

The fuel clad test facility, located in the 324 Building, is very nearly completed, Figure 12.31. The final items of construction in progress are the high pressure-low pressure-vacuum piping used for specimen stressing and for other manipulations of the test rig (Figure 12.32) and an inert atmosphere handling box that will be used to protect the sodium in the test

TABLE 12.10. Test Outline for EBR-II Irradiation

Temperature, °F		Fluence, n/cm ²	Rupture Time, hrs
Irradiation	Test		
1000	1000	1×10^{22}	$10^{-1}, 10^0, 10^1, 10^2, 10^3, 10^4$
1000	1200		$10^{-1}, 10^0, 10^1, 10^2, 10^3, 10^4$
1000	1400		$10^{-1}, 10^0, 10^1, 10^2, 10^3, 10^4$
1000	1000	0.5 to 0.8 $\times 10^{22}$	$10^{-1}, 10^0, 10^1, 10^2, 10^3, 10^4$
1000	1200		$10^{-1}, 10^0, 10^1, 10^2, 10^3, 10^4$
1000	1400		$10^{-1}, 10^0, 10^1, 10^2, 10^3, 10^4$

Neg 0683059-1

FIGURE 12.31. Interior of Fuel Clad Test Facility, Showing General Arrangement of Furnaces, Transmission Leads, and Manipulators and Other Handling Devices

specimen while the specimen is being attached to the test rig.

A "shakedown" operation will be performed to identify deficiencies in the complex and to enable their rectification prior to initiation of the test program.

A request has been submitted for approval of space in Row 2 of EBR-II to conduct irradiations on fuel cladding material. Two fuel cladding programs are of interest: a study of the effects of cold-work on the biaxial stress-rupture properties of

Type 316 SS, and a comparison of non-fueled and fueled tubes.

For the effects study, the conditions of interest are 5, 10, and 20% cold-work versus annealed fuel cladding. Available data show that cold-work in tubular specimens can enhance the biaxial stress-to-rupture properties of stainless steels.⁽³⁴⁻³⁶⁾ It is apparent that cold-work can raise the stress required to cause rupture for a period of time depending on temperature. Beyond that time, the stability of the annealed

Neg 0683059-2

FIGURE 12.32. Biaxial Stress-Rupture Rig

structure is the controlling factor, and the advantage of the cold-work appears to be lost. Data are not available for irradiated and unirradiated specimens in various cold-work levels but with all other variables constant. Furthermore, meaningful extrapolations and interpolations to obtain this data are

difficult at best because of the variations in heats, temperatures, environments, etc. It will be a further objective, then, to define that period of time for which cold-work enhances properties for each of the fluences, test temperatures, and material conditions described below. Referring to that period of time at which annealed and cold-worked curves cross as the intercept, the locus of the intercepts can then be plotted to determine for each condition whether the benefits of cold-work will for a period of time be sufficient for FFTF design criteria.

These irradiations will be performed in 1000 and 1200 °F sodium to fast fluence levels of 0.6, 1 and 6×10^{22} n/cm² ($E > 0.1$ MeV). The proposed test program is given in Table 12.11.

For the comparison study, peak fluences of PNL subassembly numbers 1, 5, and 6, as well as fuel clad material and condition, will be duplicated, and irradiation temperatures will be duplicated as far as possible. Sufficient biaxial stress-to-rupture tests will be performed on fueled cladding to permit meaningful comparison with nonfueled tube properties. The technical problems to be encountered in removal of fuel from irradiated cladding and post-irradiation testing of fueled cladding are being studied. The proposed test program is outlined in Table 12.12.

Preliminary tasks are being performed in support of this irradiation. Irradiation subcapsules are being fabricated, using gamma heat values

TABLE 12.11. Proposed Test Program for Cold-Worked 316 SS Fuel Clad

Temperature, °F		Exposure, n/cm ² (a)	Rupture Time, hrs
Test	Irradiation		
1000	1000	$\sim 6.0 \times 10^{22}$	$10^0, 10^1, 10^2, 10^3, 10^4$
1000	1200	$\sim 6.0 \times 10^{22}$	$10^0, 10^1, 10^2, 10^3, 10^4$
1200	1200	$\sim 6.0 \times 10^{22}$	$10^0, 10^1, 10^2, 10^3, 10^4$
1000	1000	$0.6 - 1.0 \times 10^{22}$	$10^1, 10^2, 10^3, 10^4$
1200	1200	$0.6 - 1.0 \times 10^{22}$	$10^1, 10^2, 10^3, 10^4$

Total Tests: 92

(a) $E > 0.1 \text{ MeV}$

NOTE: Each item represents four tests: annealed and 5, 10, and 20% cold-worked fuel cladding.

TABLE 12.12. Non-Fueled Versus Fueled Cladding Comparison Study

Type 316 SS 0.250 in. diam x 0.016 in. wall-fuel clad

Temperature, °F		Rupture Times, hr
Irradiation	Test	
900	900	$10^0, 10^1, 10^2, 10^3$
1000	1000	$10^0, 10^1, 10^2, 10^3$
1100	1100	$10^0, 10^1, 10^2, 10^3$
1200	1200	$10^0, 10^1, 10^2, 10^3$

[Companion to PNL fueled subassembly number 6--peak fluences
 $\sim 3.72 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$)]

Type 304 SS 0.250 in. diam x 0.016 in. wall-fuel clad (Lot 'F')

Temperature, °F		Rupture Times, hr
Irradiation	Test	
900	900	$10^0, 10^1, 10^2, 10^3$
1000	1000	$10^0, 10^1, 10^2, 10^3$
1100	1100	$10^0, 10^1, 10^2, 10^3$
1200	1200	$10^0, 10^1, 10^2, 10^3$

[Companion to PNL fueled subassembly number 5--peak fluence
 $3.72 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$)]

Type 304 SS 0.250 in. diam x 0.016 in. wall-fuel clad (Lot 'E')

Temperature, °F		Rupture Times, hr
Irradiation	Test	
1000	1000	$10^0, 10^1, 10^2, 10^3$
1100	1100	$10^0, 10^1, 10^2, 10^3$

[Companion to PNL fueled subassembly number 1--peak fluence
 $\sim 1.0 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$)]

for Row 2 of EBR-II, that will provide the irradiation temperatures desired. Fabrication of annealed and 5, 10, and 20% cold-drawn Type 316 SS tubing of a 0.235 in. diam by 0.014 in. wall dimension is progressing and specimens will be available by October 1, 1968. Subassembly insertion in the EBR-II is planned for about mid-December, 1968.

CREEP TEST FACILITIES DEVELOPMENT

R. H. Todd

The comprehensive investigation of the effects of irradiation and environment on the creep-rupture properties of candidate structural and cladding materials for use in the FFTF has created a need for a facility that can provide the necessary test environments. Such a facility, now under construction by Battelle-Northwest, will provide means for testing control and irradiated specimens in sodium and controlled gas atmospheres. The final phases of construction now in progress involve installation of the controls for the heating and ventilation system. Test frames from the CANEL equipment pool have been moved into place and are now being activated.

The CANEL test stands temporarily located in the mezzanine workshop produced 12 uniaxial tests during the past three months; 2 tests in static sodium (843 hr total); and 10 tests in air (1978 hr total). Because of the increased emphasis on creep and stress-to-rupture tests in inert atmosphere rather than sodium, one of the CANEL units in the

workshop is being converted for testing in a helium atmosphere. The changes necessary will be applicable to similar units obtained from the CANEL equipment pool.

The support section of the liquid metal loop was operated intermittently for seven days to gain operating experience and to further check out all parts of the loop. This operation disclosed a slow leak in the main drain valve. Flow of sodium in the loop had to be halted periodically as the level of sodium dropped, and more sodium had to be charged back into the loop from the sump tank. This valve was removed and inspected. Numerous small bits of metal were found from drilling or lathe operations, and the valve seats were badly scored. A new valve was installed, and the welds X-rayed for integrity. Subsequently, no leakage has been detected. Parts for the expansion tank level probe were received from the vendor, but this instrument did not work during the initial test run and has been under intensive study by the vendor for several months. When the supplementary parts were properly installed, an operational check of the probe demonstrated that the low side worked satisfactorily while the high side did not. Because a careful check of the circuitry established that the probe for the high side had been shorted out during fabrication, a new probe was obtained and installed in the expansion tank. Additional heater elements were received from the vendor and installed along the return line

from the cold trap. During high-temperature operation, the sodium returning along this line was not within the temperature-difference limits allowable for mixing with the main-stream sodium.

All of the changes described above have been tested in operation up to 1000 °F for short time periods and, within the time and temperature conditions of these tests, the changed or added parts have performed satisfactorily. A sustained test run of about 100 hr at a temperature of 1200 °F is planned to complete the reliability tests on the support section of the liquid metal loop. Present plans call for addition of one test leg to the loop having a single test station that will be capable of testing five biaxial unirradiated specimens at one time. Drawings for construction of the test loop will be issued within the next month.

The Prototype hot-cell passed a radiation-integrity test. A load of ten of the most radioactive specimens available was used to simulate the most severe operating conditions. Some parts of the cell were found to have less shielding efficiency than the average, but they were sufficient to maintain an acceptable radiation level on the outside of the cell. Work is now being directed toward fitting the cell with test furnaces and appropriate supporting devices and tools.

LMFBR FUEL CLADDING INFORMATION CENTER

F. R. Shoher

An information center using a computerized storage and retrieval system has been started at Battelle-Northwest to collect, classify, and catalogue all information and data pertaining to the utilization of materials for liquid metal fast breeder reactor fuel cladding. Initially, this information will pertain only to austenitic stainless steels, and particularly AISI Type 316, in direct support of FFTF fuel pin experiments and FTR cladding fabrication. Ultimately, information for all materials with potential LMFBR cladding applications will be included. The information and data to be stored will consist of primary material processes, composition, fabrication, properties, fuel element fabrication, and irradiation history. The center will further perform a data analysis function to relate the effect of manufacturing and composition variables to lot-to-lot variation in properties, to evaluate fuel pin behavior during irradiation in terms of lot-to-lot variation in properties and irradiation history, and to generate from these data information necessary for the reliability analysis of FTR cladding.

A computer program has been designed to store all information

and data relating cladding manufacture to mechanical properties for all fabrication stages through the finished tubing. To aid in the design of the program, a flow diagram, identifying each step in the manufacturing process, was prepared. Figure 12.33 shows a schematic diagram of all successive steps in the manufacturing processes and the quality control inspections for the fabrication of seamless or welded stainless steel tubing. An attempt to anticipate future needs has been made by selecting several commercially feasible processes for fabrication of small diameter tubing, including those requested in current procurement specifications. A second schematic diagram (Figure 12.34) lists the information and data to be stored in conjunction with the various manufacturing stages. Data from two lots of 316 seamless stainless steel tubing received from Superior Tube are being stored in the system. These data, although incomplete, will serve to test the accuracy, adequacy, and functional characteristics of the system. The results of these tests will establish the need for any modifications that appear necessary to complete this portion of the system. The retrieval program for this portion of the system will be designed concurrently with the storage program to provide information from the fuel fabrication, fuel pin fabrication, fuel subassembly fabrication, irradiation history, and post-irradiation examination.

A series of computer programs, the Biomed Series, are being used on the mechanical property data from REM to demonstrate their effectiveness in analyzing data in terms of several dependent and independent variables. A similar analysis will be made to determine the dependency of lot-to-lot variation in properties on manufacturing variables for LMFBR cladding when sufficient data are available.

Data Processing

The Radiation Effects on Reactor Materials (REM) Computer Program has undergone some changes since the last reporting period. The master file, consisting of two reels of magnetic tape containing some 5600 tests on several different alloys, was divided and placed on five master files according to alloy type. This was done to reduce search time for retrieval of information on any given alloy. Plotting routines have been written--and are currently being checked out--to display the data retrieval in a variety of forms such as property versus test temperature, property versus fluence, and stress versus strain. The addition of new subroutines to handle creep and stress-rupture data has been discussed and parameters described. There were 83 tensile tests added to Master File No. 1, Stainless Steel File, which brings the cumulative total to 1792 for this file. With no action on the other master files, the total for all files is now 5680.

FIGURE 12.33. Flow Diagram for Manufacturing Processes and Quality Control Inspection of Stainless Steel Tubing

RECORD NAME	RECORD TYPE	Material Code	Type Material
MATERIAL	205		
PRODUCER	210	Producer Code	Name of Producer Location of Producer
HEAT	215	Melt (Heat) Number	Number of Ingots Date of Melt Number Times Melt Type Melt Gas Condition Furnace Liner Melt Size Parts Virgin Metal Parts Scrap Metal Prior Heat Number Melt Sequence Chemical Content Units Code
TUBE HOLLOW PRODUCER	220	Producer Code	Name of Producer Location of Producer
INGOT	225	Ingot No.	Number of Tube Hollows Fabrication Method Ingot Breakdown Temperature Temperature Variation Time at Temperature Diameter or Thickness Finished Length Finished Width Reduction Temperature Temperature Variation Time at Temperature Thickness Finished Length Finished Width
		Reduction	Shear Thickness Shear Length Shear Width Roll Pierced Number of Passes Temperature Temperature Variation Time at Temperature Outside Diameter Inside Diameter Length Lubricant Reduction Type of Weld Temperature Temperature Variation
		Inside Diameter	Outside Diameter Length Lubricant Parts Original Parts Final Chemical Content Units Code The Last 2 Words of this Record Repeat. See Data Input Definition and Appendix A For Details.
TUBE HOLLOW	230	Tube Hollow Number	Number of Tube Blanks Surface Conditioned By Drilling Surface Conditioned By Machining Surface Conditioned By Honing Surface Conditioned By Grinding Surface Conditioned By Honing Surface Conditioned By Grinding Pickling Agent Cleaning Agent Number of Passes Temperature Temperature Variation Lubricant Reduction NDT Test NDT Type Fabrication Sequence
		2nd Surface Cond. By Drilling	2nd Surface Cond. By Machining 2nd Surface Cond. By Honing 2nd Surface Cond. By Grinding Second Cleaning Agent NDT Test NDT Type
TUBE BLANK	235	Tube Blank Number	Number of Finished Tubes Outside Diameter Inside Diameter Length Chemical Content Units Code The Last 2 Words of this Record Repeat. See Data Input Definition and Appendix A For Details.
TUBING MANUFACTURER	240	Producer Code	Tube Producer Producer Location
LOT	245	Lot Number	Number of Analytical Lots Beginning Site Sequence Number Ending Site Sequence No. Number of Pins Feet of Tubing Tensile Test Temperature Tensile Test Temperature Variation Tensile Test 0.2 Strength Yield Tensile Test Ultimate Strength Tensile Test Uniform Elongation Tensile Test Total Elongation Tensile Test Area Reduction
		Burst Test Temperature	Burst Test Temperature Variation Burst Test Loading Rate Burst Pressure Burst Stress Burst Test Uniform Ductility Burst Test Total Ductility Flattening Test Pass or Reject Hardness Test Load Hardness Vickers Reading Flaring Test Pass or Reject Smallest Grain Size Largest Grain Size
		Metallograp ASTM Plate Number	Metallograp Inclusion Rating Metallograp Inclusion Type Metallograp Inclusion Thin or Heavy Corrosion Test Tensile Test Temperature Variation Tensile Test 0.2 Strength Yield Tensile Test Ultimate Strength Tensile Test Uniform Elongation Tensile Test Total Elongation Tensile Test Area Reduction Burst Test Temperature
		Burst Test Temperature Variation	Burst Test Loading Rate Burst Pressure Burst Stress Burst Test Uniform Ductility Burst Test Total Ductility Flattening Test Pass or Reject Hardness Test Load Hardness Vickers Reading Flaring Test Pass or Reject Smallest Grain Size Largest Grain Size Metallograp ASTM Plate Number
		Metallograp Inclusion Rating	Metallograp Inclusion Type Metallograp Inclusion Thin or Heavy Corrosion Test Processing Sequence Cold Worked % Cold Worked Final Anneal Temperature Final Anneal Temperature Variation Time at Temperature Hand Straightening Machine Straightening Type Machine
		Type Sanding	Internal Grit Blasting External Grit Blasting Hydrostatic Test Temperature Hydrostatic Test Temp. Variation Hydrostatic Test Pressure Time PNL or Manufacturing Specifications Anneal Atmosphere Lubricant Anneal Temperature Anneal Temperature Variation Time at Anneal Temperature
		Reduction	The Last 4 Words of this Record Repeat Several Times. See Data Input Definition For Details.
TUBING	250	Finished Tubing No.	Number of Pins Outside Diameter Inside Diameter Length Ovality Eccentricity Hydrostatic Test Result NDT Test NDT Type NDT Test Result Final Sizing Length
CLADDING PIN	255	Cladding Pin Number	Outside Diameter Inside Diameter Pin Length Ovality Eccentricity Hydrostatic Test Temperature Hydrostatic Test Temp. Variation Hydrostatic Test Pressure Time Hydrostatic Test Result
		NDT Test	NDT Type NDT Reject Size NDT Test Result

12.55

BNWL-919

FIGURE 12.34. LMFBR Information Center Master File

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