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**QUARTERLY PROGRESS REPORT
IRRADIATION EFFECTS
ON REACTOR STRUCTURAL MATERIALS
FEBRUARY, MARCH, APRIL, 1967**

MAY 15, 1967

**AEC RESEARCH &
DEVELOPMENT REPORT**

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PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

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QUARTERLY PROGRESS REPORT
IRRADIATION EFFECTS
ON REACTOR STRUCTURAL MATERIALS
FEBRUARY, MARCH, APRIL, 1967

Participating Organizations:

Argonne National Laboratory
Battelle Memorial Institute
Brookhaven National Laboratory
General Atomic Division
Idaho Nuclear Corporation
Materials Research Laboratory
Naval Research Laboratory
Nuclear Materials and Propulsion Operation
Oak Ridge National Laboratory
Pacific Northwest Laboratory

May 15, 1967

Edited at:

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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SUMMARYARGONNE NATIONAL LABORATORY

Room temperature tensile tests were performed on specimens of V-20 Ti and V-15 Ti-7.5 Cr irradiated to a nominal fluence of 5×10^{21} n/cm². The results were compared with those obtained from unirradiated specimens given a 30 day anneal at 550 °C. The strength of V-20 Ti was unaffected by irradiation at calculated temperatures of 540 and 600 °C but was increased by irradiation at a calculated temperature of 660 °C. The ductility of V-20 Ti irradiated at all temperatures was not significantly different from that of the unirradiated material annealed at 550 °C for 30 days. Irradiation of V-15 Ti-7.5 Cr at a calculated temperature of 630 °C did not

affect the strength. The ductility of V-15 Ti-7.5 Cr was reduced by approximately 10% by irradiation at 630 °C and by approximately 33% at 500 and 570 °C.

Tube-burst tests were made at 550, 650, and 750 °C on V-15 Ti-7.5 Cr, both unirradiated and irradiated to a nominal fluence of 5×10^{21} n/cm². The maximum increase in burst strength at test temperatures of 550 and 650 °C was 27 and 13%, respectively. There was no increase in strength due to irradiation at a 750 °C test temperature. The ductility of the unirradiated specimens showed about twice the ductility of the irradiated specimens at test temperatures of 650 and 750 °C.

BATTELLE MEMORIAL INSTITUTE

The radiation-induced mechanical property changes in AISI Type 347 SS after irradiation are being studied. The primary objective of the program is to determine the irradiation-induced embrittlement at elevated temperatures which occurs in stainless steels and nickel alloys. A metallographic examination, on an irradiated (2.1×10^{22} nvt) stainless steel specimen which had been tested at room temperature

and then annealed at 1350 °C for one hour, revealed the presence of a second phase at the grain boundaries. The second phase could not be identified by standard etches on stainless steel. The degree of grain growth undergone by the irradiated specimen was minimal when compared to the extensive grain growth by an unirradiated control specimen which was annealed at the same conditions.

Four capsules were inserted into ETR to establish the effect of nickel on the irradiation embrittlement at elevated temperatures. These capsules contained specimens of a manganese austenitic stainless steel (nonnickel), pure nickel, nickel with minor impurities and two cobalt alloys, one of which contained nickel, while the other did not.

An attempt is being made to correlate the effects of a predominantly fast flux and a mixed thermal and fast flux on the mechanical properties of AISI 347 SS. To accomplish this, irradiations will have to be carried out in EBR-II and ETR on the same lot of materials.

The effect of 1000, 5000, and 10,000 hr heat treatments at 426, 538 and 649 °C on the mechanical proper-

ties of AISI 347 and 348 SS at room and elevated temperatures is being investigated. The specimens were annealed in air, nitrogen-0.5 vol% oxygen, and in one case, were periodically immersed in water every 1000 hr between anneals in nitrogen-0.5 vol% oxygen. The heat treatments did not result in any significant changes in the room temperature, 426, 538 and 649 °C tensile properties of the annealed material. The heat treatments at 426 °C resulted in increased strength and ductility for the 30% cold worked material. The room temperature ductility of the welded material was also found to be decreased by long time annealing at 426 °C. Metallographic examinations indicate that some sigma phase may be formed due to the long time heat treatment.

BROOKHAVEN NATIONAL LABORATORY

Electron transmission studies of structures produced by straining Ferrovac E iron in tension at 78 °K showed very little difference between unirradiated specimens and specimens embrittled by fast-neutron ($E > 1$ MeV) irradiation to 2×10^{18} nvt at 60 °C. The only exception was that unirradiated specimens contained internal dislocation sources while irradiated specimens did not. The internal dislocation sources are attributed to ϵ -carbides, which are removed from the structure by the irradiation. Optical microscopic examination revealed that low-temperature fracture of radiation embrittled specimens was ini-

tiated along grain boundaries. The observations indicate that the embrittling irradiation decreased the low-temperature fracture stress of the Ferrovac E by depleting the iron of available carbon through combination of carbon with radiation defects.

Field ion microscopy has been used to determine the type and size of defects in neutron irradiated platinum and tungsten. For exposures up to 1×10^{18} nvt, the defects observed increase linearly with exposure. For higher exposures, the vacancy concentration saturate at approximately 2×10^{-3} at%. Stage III recovery in

tungsten has been attributed to annihilation of tungsten interstitials. Arguments are presented to substan-

tiate that Stage III recovery in neutron irradiated tungsten is an intrinsic recovery stage.

GENERAL ATOMIC DIVISION

The four ambient temperature irradiation capsules were opened, and temperature and flux monitors were examined. The out-of-cell Naval Research Laboratory Charpy impact machine was instrumented, and several success-

ful tests were made. The elevated temperature (290 °C) irradiation capsule was assembled, inserted in the LITR, and removed with a fluence of about 9×10^{18} n/cm².

IDAHO NUCLEAR CORPORATION

A program outline for investigation of the effect of hydrogen on HY 80, A302B, and A542 steels is presented. Specimen preparation, hydrogen charging, and analysis procedures are discussed. The fracture surface of the

PM-2A pressure vessel has been prepared for examination and initial observations of the fracture region in the area of the artificial defect are given.

MATERIALS RESEARCH LABORATORY, INC.

Since only a limited number of tests and only a modest range of specimen thicknesses have been tested to date, conclusions drawn from these data must be considered tentative. Nevertheless, if one were to assume the 2-in. thick specimens yielded valid toughness values at room temperature and the 1 in. thick specimens valid plane strain results at 0 °F, it must be concluded that loading

rate exerts only a modest influence on toughness over the range studied.

However, rapid loading rates do appear to promote plane strain fracturing, at least, in this mild steel. Again, it must be tentatively concluded that the measuring capacity of fracture toughness specimens could be increased by the use of very high loading rates.

NAVAL RESEARCH LABORATORY

COMPARISON OF SELECTED STEELS AFTER IRRADIATION AT 550 °F AND AT 650 °F TO A HIGH NEUTRON FLUENCE

Charpy-V specimens of six steels, including A302-B, A517-E*, 3 1/2 Ni-Cr-Mo, 5 Ni-Cr-Mo-V, and 12 Ni-5 Cr-3 Mo maraging (two heats: 162 and 184 ksi YS) steel, have been irradiated simultaneously at 550 °F and at 650 °F to neutron fluences of 4.8×10^{19} n/cm² and 4.1×10^{19} n/cm² >1 MeV, respectively. Specimens of A212-B steel were also included in the 550 °F experiment but were replaced by specimens of A542 Class II steel in the 650 °F assembly. A general trend of decreasing radiation embrittlement sensitivity with increasing yield strength was depicted by 550 °F irradiation test data; however, the 5 Ni-Cr-Mo-V steel proved a significant exception. Major differences with respect to individual tendencies toward radiation damage saturation were also found for the 550 °F exposure condition when referencing earlier experimental data. In contrast to the 550 °F service condition, experimental results for 650 °F irradiation suggest that embrittlement developed at this temperature by a fluence of $\sim 4 \times 10^{19}$ n/cm² >1 MeV will not be of great engineering concern as absolute transition temperatures were below 200 °F in all cases.

THROUGH-THICKNESS CHARPY-V NOTCH DUCTILITY PERFORMANCE OF A 10 1/2 IN. PLATE OF IRRADIATED A302-B STEEL

Charpy V-notch specimens machined from the surface and from the 1/4T and

1/2T locations in a 10 1/2 in. thick plate of A302-B steel have been irradiated at <250 °F to a fluence of 1.7×10^{19} n/cm² >1 MeV in a core lattice position of the Low Intensity Test Reactor (LITR) at Oak Ridge. A 60 °F difference in initial Charpy-V 30 ft-lb transition temperatures developed by surface and 1/2T locations was observed to be maintained after irradiation. Specimens from each of the three layers showed transition temperature increases of 240 to 245 °F. It was therefore concluded that the radiation sensitivity of the 10 1/2 in. plate was very uniform through the entire thickness, and furthermore, was somewhat less than that of other plates of the same nominal composition evaluated previously.

INVESTIGATION OF THE EFFECTS OF OXYGEN AND NITROGEN CONTENTS ON THE RADIATION EMBRITTLEMENT SENSITIVITY OF 7 1/2 Ni-Cr-Mo STEEL AT 250 °F

The effects of a series of oxygen and nitrogen contents on the radiation embrittlement sensitivity of 7 1/2 Ni-Cr-Mo steel at an exposure temperature of <250 °F have been evaluated. Test plates for this investigation were provided by a three-way split heat of a laboratory air-induction melt. Charpy-V specimens representing each test plate and a plate from a commercial electric furnace vacuum carbon deoxidized (VCD) heat were simultaneously irradiated to a fluence of 1.7×10^{19} n/cm² >1 MeV. The experimental results did not reveal a critical effect of either oxygen or nitrogen

* Armco designation: SSS-100

contents on embrittlement sensitivity of this steel composition at the low exposure temperature. The data also suggested that the choice of melting practice (air induction versus electric furnace VCD) is not critical to the level of embrittlement of this steel in its high strength condition. An elevated temperature (~550 °F) irradiation is now contemplated to further evaluate the effects of these variables on the irradiation performance of the 7 1/2 Ni-Cr-Mo composition at more representative reactor structural component service temperatures.

NOTCH DUCTILITY BEHAVIOR OF IRRADIATED 12 Ni-5 Cr-3 Mo MARAGING STEEL WELDMENTS

Two 1 in. thick weldments of 12 Ni-5 Cr-3 Mo maraging steel base plate prepared by the TIG process at NRL using 12 Ni-5 Cr-3 Mo and 17 Ni-2 Co-3 Mo weld wire have been irradiated at <250 °F to a neutron fluence of about 6×10^{18} n/cm² >1 MeV. Tests of irradiated Charpy V-notch specimens showed neither an increase in transition temperature nor a change in full shear energy absorption level from the behavior of unirradiated specimens. The absence of any radiation-induced transition temperature increase for the weld metals coupled with the high yield strength and small radiation effect on base metal properties for this exposure condition serves to reemphasize the potential value of the maraging steels for future use as a nuclear structural material.

RESPONSE OF A350-LF1 (MODIFIED) STEEL TO POSTIRRADIATION ANNEALING AT TEMPERATURES IN THE RANGE OF 550 TO 590 °F

Six groups of Charpy V-notch specimens from an A350-LF1 (Mod.) steel ring forging, representative of the Army SM-1A reactor pressure vessel, have been irradiated at 430 °F to a neutron fluence of 1.3×10^{19} n/cm² >1 MeV for determinations of the sensitivity of annealing response to temperature in the range of 550 to 590 °F. Experimental results indicate that the recovery of initial Charpy-V 30 ft-lb transition temperatures is not highly dependent upon the exact postirradiation annealing temperature within the range of 560 to 590 °F; however, annealing at 550 °F for the same 168 hour period was measurably less effective. Furthermore, while embrittlement relief from 580 or 590 °F heat treatments was not significantly different from 560 or 570 °F treatments on comparing 30 ft-lb transition temperatures, the higher temperature heat treatment would be preferred in terms of overall annealing response.

NOTCH DUCTILITY BEHAVIOR OF A350-LF1 (MODIFIED) STEEL WITH CYCLIC 430 °F IRRADIATION-168 HOUR, 585 °F ANNEAL

The effectiveness of periodic 585 °F-168 hour annealing treatments for embrittlement relief of the Army SM-1A reactor pressure vessel has been determined experimentally using steel representative of the actual vessel. Irradiation temperature and fluence conditions duplicating those in service

prior to initial vessel annealing operations were also investigated. Two complete cycles of irradiation and annealing were examined with transition temperature behavior being established at each stage of cyclic operations. The major significance of the data lies in the illustration of relief of embrittlement for the maximum fluence conditions of 3.9×10^{19} and 4.6×10^{19} n/cm^2 >1 MeV coupled with the observation that embrittlement is not accelerated during a second cycle exposure compared to the rate established for virgin material.

PREIRRADIATION MECHANICAL PROPERTIES OF MH-1A PRESSURE VESSEL STEEL

A series of material surveillance specimen capsules for irradiation in the Army MH-1A, barge-mounted reactor has been prepared by NRL. AISI Type 316 SS from the same steel heat as the pressure vessel was incorporated in the surveillance program through the use of two nozzle cutouts for the specimen stock material. Charpy V-notch specimens tested over the temperature range -250 to 500 °F to develop preirradiation characteristics of the steel forging showed energy absorption values in excess of 200 ft-lb. Tensile properties have been measured at room temperature and at the service temperature of 500 °F.

NOTCH DUCTILITY OF SEVERAL REACTOR STRUCTURAL STEELS AFTER IRRADIATION IN A HEAVY WATER MODERATED REACTOR

Charpy V-notch specimens representing two reactor pressure vessel steels,

A212-B and A302-B, and one potential reactor structural steel, A517-E, have been irradiated at a temperature <250 °F in a position adjacent to the fuel core of the Carolinas-Virginia Tube Reactor (CVTR) at Parr, South Carolina. The A212-B and A302-B steel Δ NDT data compare well with other data from irradiations at temperatures <450 °F when interrelated on the basis of n/cm^2 >1 MeV assuming a fission spectrum neutron distribution. When interrelated on the basis of n/cm^2 >0.5 MeV from calculated neutron spectrum, however, the CVTR data compare poorly with the body of data from light water moderated reactors.

MODIFICATION, INSTALLATION, AND INITIAL OPERATION OF A REMOTELY OPERATED, TAPE-CONTROLLED MILLING MACHINE

During this quarter a tape-operated, numerically controlled milling machine was modified for remote operation and installed in the hot cell machine shop. A thorough description of the modifications performed and the resulting operational capabilities are presented.

REMOTELY OPERATED PRECISION MEASURING EQUIPMENT

Two instruments have been developed at NRL which complement the Charpy-V impact testing operations in the High Level Radiation Laboratory (HLRL). These include a device for locating the position of the notch with respect to the center of the specimen length and for lateral expansion measurements. The description of both instruments is presented in detail.

NUCLEAR MATERIALS AND PROPULSION OPERATION

Irradiating tungsten to 1.4×10^{20} n/cm² ($E_n \geq 1$ MeV) at 1100 to 1350 °C indicated creep-rupture ductility was a function of irradiation and test temperatures. Specimens irradiated at reactor ambient temperature and tested at 1100 °C showed the effect of different reactor spectra was mainly due to the production of rhenium by the (n,γ) reaction. Tensile tests of irradiated tungsten at 400 °C showed the yield-strength increase was proportional to the square root of fast neutron fluence. Annealing an irradiated (1.4×10^{20} n/cm²) specimen at 1200 °C for 1 hr resulted in about 80% recovery of yield and ultimate strengths but very little recovery of ductility; annealing specimens irradiated to 8×10^{19} n/cm² at 1300 °C produced complete recovery of both strength and ductility. Radiation effects on tungsten resistivity appeared to reach saturation at a fast neutron fluence of about 1.5×10^{21} n/cm². A fluence threshold region existed in tungsten between 5.9×10^{18} and 3.8×10^{19} n/cm² ($E_n > 1$ MeV) over which an abrupt increase in size and concentration of clusters occurred, leading to a drastic loss in tensile ductility. Irradiated molybdenum tested at 580 °C evidenced accelerated creep at a fluence level of 5×10^{18} n/cm² ($E_n \geq 1$ MeV) but not at a higher fluence level of 3×10^{19} n/cm². Delayed creep was observed at 600 and 700 °C for irradiated molybdenum but not at 580, 850, or 900 °C. In resistivity studies of irradiated molybdenum, the radiation-induced resistivity showed a maximum at 10^{20}

n/cm², the first evidence of saturation of neutron irradiation damage. Recovery studies of the hot-hardness and tensile properties of Mo, Mo-TZM, Mo-50Re, W, W-25Re, and W-25Re-30Mo, irradiated at reactor ambient temperature, showed that these properties are generally recovered at temperatures greater than $0.22 T_m$ for the alloys and above $0.35 T_m$ for the unalloyed metals.

The creep rupture life Hastelloy N was reduced by irradiation by a factor of 20 to 30 at 650 °C and 2 to 3 at 815 °C; the loss in rupture life was apparently affected by the maximum temperature regardless of irradiation or subsequent testing. Pre- and post-irradiation annealing indicated rupture life may be recoverable; however, techniques that improved rupture life did not improve postirradiation ductility. The fracture of unirradiated Hastelloy N was transgranular at 650 °C but intergranular at 815 °C. The radiation-induced embrittlement of A-286 was not caused by coalescence of helium bubbles on grain boundaries; rather, it appeared that the helium bubbles were pinning dislocations. Irradiated Hastelloy R-235 containing a fixed boron content but varying amounts of B¹⁰ showed a loss in rupture life at 870 °C, with the loss increasing with increasing B¹⁰ content. Tensile tests at temperatures to 1000 °C of Fe-15Cr-4Al-1Y alloy, irradiated at reactor ambient temperature or 700 °C to a fluence of 6 to 8×10^{19} n/cm², showed no radiation-induced embrittlement.

OAK RIDGE NATIONAL LABORATORY

The creep-rupture properties of type 304 SS at 650 °C have been investigated in the presence of a neutron flux of 6×10^{13} neutrons/cm²-sec, after irradiation and in the unirradiated condition. No change in the creep rate was found that could be attributed to the effects of irradiation. However, as expected the rupture time and ductility were decreased.

Work involving the introduction of helium into stainless steels by use of the Oak Ridge Isochronous Cyclotron has continued. The results for a 1 ppm helium concentration in fine-grained and coarse-grained type 304 SS show essentially the same grain-size effect as has been observed in reactor irradiated type 304 SS. Samples of the 0.2% titanium-modified stainless steel, containing 1 ppm helium, show no loss in ductility when tested in the fine-grained condition. However, the mate-

rial annealed at 1038 °C shows a slight decrease in ductility at test temperatures above 700 °C.

Electron microscopic observations of helium bubbles in a titanium-bearing stainless steel have led to several interesting conclusions regarding their location in the metal; the most important of which is that bubbles do attach themselves to precipitates in the alloy.

A systematic study has been conducted of the mechanical properties of Hastelloy N irradiated in the ORR and the Molten-Salt Reactor Experiment to similar fluence. The results of tests from 20 to 850 °C indicate that the effects produced were similar for both irradiation facilities.

Our experiments designed to measure the nuclear heating rate in EBR-II have been delivered to the EBR-II site and are awaiting insertion.

PACIFIC NORTHWEST LABORATORY

ALLOY SELECTION

Production of a new program material, Incoloy Alloy 800, is completed. The rod and flat stock will be stored at Battelle-Northwest. The tube blanks will be stored at Atomics International.

Six tensile specimens from the GE-San Jose superheat program, irradiated in the EBR-II, show almost no effects of the irradiation on Incoloy 800 (0.17×10^{20} n/cm²) but substantial changes in strength and ductility for Inconel 625 and Hastelloy X ($2.7 - 2.9 \times 10^{20}$ n/cm²).

IRRADIATION FACILITIES OPERATION

A total of 205 specimens was discharged from the ETR at the conclusion of Cycles 86 and 87. The variety of specimens included tensile, double cantilever beam (DCB), thin foils and single crystals, and corrosion blanks.

Tensile data reports have been recorded, processed, and stored in the Radiation Effects on Metals (REM) computer program for 5009 individual tests.

Thermal aging of an inert atmosphere out-of-reactor capsule, GEH-22-3,

containing tensile specimens of AISI 304 SS and 316 SS is in progress.

The results of melting point tests conducted on a series of irradiated Tempil melt pellets show that the pellets are not suitable as thermal monitors in neutron environments.

IRRADIATION DAMAGE TO STAINLESS STEELS

Annealed AISI 304 SS tensile specimens have been irradiated at 650 °C in a NaK environment to an exposure of 3.5×10^{20} fast fluence. It appears that carbide precipitation in these specimens is inhibited by neutron irradiation but that carbides are partly responsible for the loss of high-temperature ductility. An exposure of 6.0×10^{21} fast fluence has been obtained on 304 SS. This is the highest exposure to date obtained in the ETR G-7 hot water loop. These data have been used to test the reliability of predicting tensile properties, in particular uniform elongation, from lower exposures. It has also been illustrated that the data at this exposure provide an important link for the comparison of fast-to-thermal irradiations. In this regard, it has been shown that the tensile properties of 304 SS converge at 6.0×10^{21} fast fluence for a test temperature of 750 °C. It has also been shown that the nature of the radiation-induced displacement damage at this exposure is probably different from that observed at lower exposures.

IRRADIATION DAMAGE TO NICKEL-BASE ALLOYS

Electron transmission microscopy of a standard solution treated specimen of Hastelloy X-280 that was irradiated

at 540 °F (280 °C) to 1×10^{20} nvt (>1 MeV) and annealed at 600 °C for 2 hr showed zones 1-2 μ in width around matrix and grain boundary precipitates that were denuded of cluster defects. This had not been observed in either the as-irradiated condition or after a 500 °C anneal.

Phase analysis of Inconel 600 confirms that more chromium carbides result from the experimental aging treatments than from the standard solution treatment.

Phase analysis of Inconel X-750 was instituted to illuminate mechanisms of ductility improvement.

IRRADIATION DAMAGE TO SINTERED ALUMINUM PRODUCT

A program to determine the effect of irradiation on Sintered Aluminum Products (SAP) has been initiated. Irradiation at high temperature to a fluence of 1×10^{22} ($E \geq 1$ MeV) is planned. Test material is on hand; characterization of material has been initiated.

IN-REACTOR MEASUREMENTS OF MECHANICAL PROPERTIES

In-reactor creep tests conducted on annealed 304 SS are showing a greater reduction of rupture life due to irradiation at temperatures above 700 °C than at temperatures below 700 °C.

An in-reactor creep test conducted on Hastelloy X-280 ruptured in a shorter time than predicted from unirradiated data for this alloy.

Specimens of Zry-2.5 wt% Nb pre-crept in-reactor at 350 and 400 °C and then tested in tension showed no appreciable loss of ductility.

DAMAGE MECHANISMS

This investigation of the influence of neutron radiation damage on two purities of alpha iron has shown that:

1. The major effect is an increase in internal stress which is attributed to small defect clusters.
2. Carbon impurity atoms serve as traps for interstitials and nucleate the majority of the clusters.
3. Defects in irradiated iron produce a small change in activation volume that recovers upon annealing at 350 °C. This is a modification of the basic rate controlling process rather than a new rate process.
4. Activation volumes recover with strain, reflecting the interaction between defects and dislocations and the localized nature of the deformation.

A system for measuring the electrical resistivity of materials has been set up and is presently being checked out and evaluated.

HIGH PRESSURE STUDIES

Calibration of the In-Reactor Diamond High Pressure Cell to 50 kbars has demonstrated that the actual pressure on a weak sample surrounded by a strong washer may deviate by a factor of two or more from that predicted by a simple force/area ratio. The proposed fcc-bcc phase transition in NaCl at 17 kbars was not observed which is in accordance with the more recent findings of high pressure scientists.

A description of the out-of-reactor high pressure-temperature equipment is presented. This equipment will be used in pre- or postirradiation studies of nuclear reactor materials.

IN-FLUX OXIDATION AND HYDRIDING OF TITANIUM AND NIOBIUM

Specimens of titanium, niobium, niobium-1% zirconium, and Zircaloy-2 were exposed 37 days in the ETR G-7 loop in pH-10 NH_4OH at 280 °C. Titanium weight gains were lower than Zircaloy-2 weight gains. Niobium weight gains were variable, but were generally considerably higher than Zircaloy-2. Titanium absorbed hydrogen influx, while niobium did not.

PRESSURE-VESSEL DOSIMETRY

The possibility of using fission-track detectors as a fluence monitoring technique for pressure vessel surveillance is being investigated. Glass, mica, and plastic samples have been irradiated in test facilities in the Hanford K-Reactor to determine the limiting exposure level at which the detectors become useless due to radiation damage. Plastics were found to be unsuitable for use at expected pressure vessel fluences. Fast-neutron irradiation damage limited the use of the micas investigated to fluences less than $\sim 5 \times 10^{18} \text{ n/cm}^2$, $E > 1 \text{ MeV}$, (about 10 yr at the surface of a typical pressure vessel). Above this exposure, optical microscopy methods could not be used successfully.

Mica and glass samples were coated with a thin layer of highly depleted uranium and irradiated. This technique appeared an attractive way to monitor fast neutrons and minimize the tracks formed by thermal-neutron-induced fission of ^{235}U . This coating technique yielded satisfactory results only at fluences less than $\sim 10^{15} \text{ n/cm}^2$.

($E > 1$ MeV). Therefore, coating fissionable material on mica and glass is not suitable for pressure-vessel applications.

Natural mica is a good candidate for a detector material within its exposure limits. Fission fragments are registered with 100% efficiency in mica, and processing techniques, including etching and data analysis, have been established. However, extension of the fluence range will require a more radiation resistant material. To determine whether the accuracy of the method will meet pressure-vessel requirements, further investigation is required for detector calibration, determination of the reproducibility of the fission tracks, and effects on nonhomogeneous distribution of the uranium in the material.

INTERCALIBRATION TEST ON COUNTING LABORATORIES

An intercalibration test has been initiated among several counting laboratories in order to insure that experimenters involved in the Irradiation Effects on Reactor Structural Materials Program have comparable fluence determinations.

FAST NEUTRON MECHANISMS

The possible effects of nonideal effusion conditions on expected neutronic-hydrogen concentration profiles in fast reactor core components have been evaluated for endothermically occluding alloys systems. The formation of surface permeation regions with unfavorable hydrogen diffusivity or solubility characteristics can substantially increase the steady-state

concentrations calculated for ideal effusion behavior; however, the magnitude of these concentration increments can be readily predicted using simple permeation tests. The maximum neutronic-hydrogen concentrations calculated for the transient period preceding steady-state effusion behavior approximate those for complete hydrogen retention.

EBR-II IRRADIATIONS

Initial tensile testing of specimens of annealed AISI 304 SS irradiated in EBR-II over the temperature range room temperature to 1600 °F (870 °C) has been completed. Results show that radiation hardening persists to temperatures as high as $0.67 T_m$ after fast reactor irradiation to 1.7×10^{22} nvt. This radiation hardening results in high-temperature ductility loss.

Two subassemblies containing AISI 304, 316, 321, and 348 and various nickel base alloys have been placed in the EBR-II.

LIQUID METAL CAPSULE IRRADIATIONS

Liquid metal capsule GEH-22-3 was discharged during shutdown of ETR Cycle 87 accumulating a total fast fluence of 1.38×10^{21} .

GEH-22-4 began irradiation in Cycle 87. Design modifications have permitted capsule sodium temperatures of 1400 °F (760 °C) at 50-50 gas mixture control.

EXPERIMENTAL TESTING FACILITIES

The building that will house the testing facilities has been improved sufficiently for office spaces and workshop to be occupied. Renovation

of the remainder of the building will begin when a project proposal for that purpose receives final approval. Four test stands from the CANEL equipment pool have been overhauled and will be ready for service shortly. The prototype cell for testing irradiated samples in vacuum or controlled atmosphere is in place and nearly complete. The remotely operable test stands, which are to be used in this cell, are ready for testing under cell conditions with manipulators. The sodium loop which will provide high temperature flowing sodium for the test program, will not be shipped until April--a delay caused by late delivery of essential parts. The special creep test apparatus to be used in the sodium loop has been designed and a prototype is being made ready for performance tests. The final unit is approximately half the physical size and half the cost of the original working model. The scoping design package of the section of the sodium loop for testing of irradiated samples is nearly complete. A special dispenser has been designed for dispensing high-purity liquid sodium without contamination to other enclosed containers.

RADIATION EFFECTS ON METALS - 05 PROGRAM

Studies have continued on the effect of irradiation temperature on molybdenum. Unirradiated molybdenum has been annealed for a time and at a temperature equivalent to that pertaining to molybdenum irradiated at 600 °C. No carbide formation was ap-

parent in either the high purity or carbon-bearing molybdenum although it is possible that carbide growth occurred in the carbon-bearing molybdenum.

No denuding of loops near boundaries is observed in molybdenum irradiated at 60 °C and annealed at elevated temperatures, where as significant denuding is observed in molybdenum irradiated at elevated temperatures. Also, considerably smaller loops are observed in material irradiated at low temperatures and annealed at elevated temperatures. While under observation in the electron microscope, a small cluster was observed to move in a glissile fashion in the irradiated molybdenum. This was probably due to beam heating, and may explain the increased concentration of the small clusters inside the large interstitial loops.

Nickel, irradiated to a higher fluence than previously reported (1.7×10^{19} n/cm²), shows slightly larger loops than that at a lower fluence. No loop interaction, either in the as-irradiated state or after annealing, was noted. A short note describing the phenomena of increased damage accumulation at coherent twin boundaries in nickel has been submitted to The Journal of Applied Physics for publication.

A stored-energy calorimeter has been designed, assembled, and tested. Preliminary tests show that heat effects as small as 0.01 cal/g of copper are detectable. This calorimeter will be employed in the study of annealing processes in irradiated molybdenum, nickel, and iron. It may be used with

both single-crystal and polycrystalline specimens.

Molybdenum samples for the isobaric-isochronal annealing experiments have been irradiated to 5×10^{18} n/cm² (fast). A model of the In-Reactor Hydrostatic Pressure Cell has operated successfully at 17 kbars for a month in an out-of-reactor test. The out-of-reactor high pressure facility is described.

MODEL GAS LOOP

The model gas loop was operated a total of 3110 hr since installation of the new heater; this includes 920 hr this quarter. Exposure of superalloys and refractory metal corrosion specimens at two locations in the loop is continuing. No major maintenance problems were encountered during the quarter.

A study of a method to upgrade the temperature under which the vertical specimens can be evaluated was made.

TEST ASSEMBLY EVALUATION

Exposure of a prototype ATR test assembly in the model gas loop demonstrated that the present design performs satisfactorily. Surface contamination of molybdenum, chromium, manganese, iron, and oxygen was found on the assembly after exposure at 940 °C. Contamination resulted in significant strengthening of niobium and modest strengthening of tantalum, but good room temperature ductility was retained.

SUPERALLOY AND REFRACTORY METAL CORROSION STUDIES

The niobium, niobium-1 zirconium, and tantalum corrosion samples exposed to ATR Model Loop "clean run" conditions readily pick up impurities from loop atmospheres pure enough to permit evaporation of superalloys. Molybdenum, TZM, and tungsten samples remain comparatively unaffected. Continued exposure of all samples is planned.

ATR GAS LOOP OPERATION AND MAINTENANCE

Examination of test assemblies and corrosion coupons exposed in the Model Gas Loop show evidence of in-leakage of air even though the gas impurity level was maintained at less than 1.0 ppm total impurities. Tests are planned to determine the source and amount of in-leakage. Studies are also being conducted to develop methods of increasing the temperature of specimens to 2500 °F (1370 °C) which will more closely approximate the temperatures anticipated during ATR operation.

Minor weaknesses in the gas chromatographs are being corrected and work is continuing towards achieving maximum sensitivity and assurance that equipment will measure total impurities in the gas to less than 1 ppm. The gas analysis equipment for measuring dew point in the Transfer Facility was inspected and accepted.

Examination of corrosion coupons after 504 hr exposure in the Model Gas

Loop is in progress. Hastelloy X-280 and Haynes 25, which incurred weight loss, show metal loss from grain boundaries. Niobium and tantalum, which incurred weight gains, show surface contamination. In the case of

one niobium sample, the surface contamination layer was shown to be Nb-

$N_{0.9}-O_{0.1}$.

Review of vendor data has continued and is currently 60% complete.

IRRADIATION EFFECTS ON V-20 Ti AND V-15 Ti-7.5 Cr ALLOYS

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INTRODUCTION

In the development of vanadium alloys as candidates for LMFBR fuel jackets, a study of the effects of fast neutrons at reactor temperatures is essential. A program of irradiations in EBR-II was begun in late 1965 for this purpose. Initially, only V-20 Ti alloy was irradiated, but in the summer of 1965 V-15 Ti-7.5 Cr was included in the program.

The program consists of irradiations of tensile type specimens and tube sec-

tions. The tensile specimens serve for short-time tensile tests and creep rupture tests at temperatures up to 750 °C. The tube sections are used for short-time burst tests and, eventually, will serve for creep rupture tests. The neutron exposures will range from 5×10^{21} to 1×10^{23} n/cm² and irradiation temperatures will range from 500 to 700 °C. Specimens currently in the reactor are shown in Table 3.1. Specimens already removed from the reactor are shown in Table 3.2.

TABLE 3.1. Status and Identification of ANL-Vanadium-Base Alloy Irradiations in EBR-II

Material	Specimen-Type	Heat Treatment, 1 hr at	No. of Specimens	Maximum Specimen Temp., °C	Current Fluence, n/cm ²	Target Fluence, n/cm ²
V-20 Ti	Tensile	900 °C	9	650	5.3×10^{20}	1×10^{22}
V-20 Ti	Tensile	900 °C	9	650	8.1×10^{20}	2×10^{22}
V-20 Ti	Tensile	900 °C	74	500	1.6×10^{22}	4×10^{22}
V-20 Ti	Tensile	900 °C	32	580	3.7×10^{22}	7×10^{22}
V-20 Ti	Tube-burst	900 °C	24	540	3.4×10^{22}	7×10^{22}
V-20 Ti	Tensile	900 °C	15	700	5.3×10^{21}	1×10^{23}
V-20 Ti	Tensile	1000 °C	9	650	5.3×10^{22}	1×10^{22}
V-20 Ti	Tensile	1000 °C	9	650	8.3×10^{20}	2×10^{22}
V-20 Ti	Tensile	1000 °C	8	700	5.3×10^{21}	1×10^{23}
V-20 Ti	Tensile	1100 °C	9	650	8.2×10^{20}	2×10^{22}
V-20 Ti	Tensile	1100 °C	6	700	5.3×10^{20}	1×10^{23}
V-15 Ti-7.5 Cr	Tensile	850 °C	9	650	8.6×10^{20}	2×10^{22}
V-15 Ti-7.5 Cr	Tensile	850 °C	17	700	5.3×10^{21}	1×10^{23}
V-15 Ti-7.5 Cr	Tensile	1100 °C	9	650	8.6×10^{20}	2×10^{22}
V-15 Ti-7.5 Cr	Tube-burst	1100 °C	16	650	8.6×10^{20}	2×10^{22}
V-15 Ti-7.5 Cr	Tube-burst	1100 °C	36	700	5.3×10^{21}	1×10^{23}
V-15 Ti-7.5 Cr	Tensile	1100 °C	10	700	5.3×10^{21}	1×10^{23}
V-15 Ti-7.5 Cr	Tensile	1250 °C	9	650	8.9×10^{20}	2×10^{22}
V-15 Ti-7.5 Cr	Tensile	1250 °C	8	700	5.3×10^{21}	1×10^{23}

TABLE 3.2. Identification of ANL-Vanadium-Base Alloy Specimens Irradiated in EBR-II

<u>Material</u>	<u>Specimen-Type</u>	<u>Heat Treatment, 1 hr at:</u>	<u>No. of Specimens</u>	<u>Maximum Specimen Temp., °C</u>	<u>Target Fluence, n/cm²</u>
V-20 Ti	Tensile	900 °C	18	650	5×10^{21}
V-20 Ti	Tensile	900 °C	26	670	2.5×10^{22}
V-20 Ti	Tensile	900 °C	16	585	4×10^{22}
V-20 Ti	Tensile	1000 °C	18	650	5×10^{21}
V-20 Ti	Tensile	1100 °C	18	650	5×10^{21}
V-15 Ti-7.5 Cr	Tensile	850 °C	18	650	5×10^{21}
V-15 Ti-7.5 Cr	Tensile	1100 °C	18	650	5×10^{21}
V-15 Ti-7.5 Cr	Tube-burst	1100 °C	64	650	5×10^{21}
V-15 Ti-7.5 Cr	Tensile	1250 °C	18	650	5×10^{21}

One group of specimens irradiated to a nominal fluence of 5×10^{21} n/cm² at a nominal temperature of 650 °C has been partially examined. These specimens were irradiated in gas-annulus sodium filled capsules. The tensile specimens were inside the specimen capsule tube that provided the tube burst specimens. Tempil pellets were included for temperature monitors over the range of 427 to 704 °C. Iron, nickel, and titanium wires were included as fast flux monitors, but the flux monitors have not yet been examined. Calculated neutron exposures along the length of the capsule are shown in Figure 3.1. Calculated temperatures and the results of evaluating the Tempil pellets are shown in Figure 3.2. The irradiation temperatures are not well defined.

Tensile specimens of both V-20 Ti and V-15 Ti-7.5 Cr are in this group. Tube burst specimens are available only from V-15 Ti -7.5 Cr inner capsule tubes. Both materials have been calcu-

lated to have 20 atoms of hydrogen and one atom of helium per million atoms of alloy as a result of irradiation.

ROOM TEMPERATURE TENSILE TESTS

Procedure

Room temperature tensile tests were performed on V-20 Ti and V-15 Ti-7.5 Cr irradiated in EBR-II to a nominal fluence of 5×10^{21} n/cm² as part of a study to separate the effects of exposure and irradiation temperature, as well as to provide a basis for determining to what extent recovery will effect the observed mechanical property changes during subsequent high-temperature tensile testing.

The tests were conducted at a strain rate of 2%/min on specimens of V-20 Ti alloy that had been given a one hour anneal at 900 °C prior to irradiation, and specimens of V-15 Ti-7.5 Cr alloy that had been given a one hour anneal at 850 °C prior to irradiation. Strain measurements were taken between the grips. The values were compared to

FIGURE 3.1. EBR-II Gas-Annulus Sodium-Filled Jacket Materials Capsule

FIGURE 3.2. Axial Temperature Distribution Along Capsule

those from unirradiated control specimens, some of which were annealed at 550 °C for 30 days to simulate reactor conditions. Specimen preirradiation history is given in Table 3.3. The results are given in Table 3.4. The temperatures given in Table 3.4 are calculated temperatures, represented by the curve in Figure 3.3.

Results

Both the yield strength and the ultimate tensile strength of the unirradiated V-20 Ti were decreased by approximately 14 to 17% by the 30 day 550 °C anneal. Irradiation at calculated temperatures of 540 and 600 °C did not significantly change the yield strength and tensile strength from that shown by the unirradiated control material annealed for 30 days at 550 °C. Irradiation near 660 °C increased the strength slightly above that which might be expected from the 550 °C anneal.

The ductility of the unirradiated V-20 Ti was decreased 25% due to the

30 day 550 °C anneal. Irradiation at all temperatures resulted in ductility decreases of the same order of magnitude as that resulting from the 550 °C anneal.

The yield strength and the ultimate tensile strength of the unirradiated V-15 Ti-7.5 Cr were increased 4 to 6% by annealing for 30 days at 550 °C. In comparison to this annealing treatment, irradiation at calculated temperatures near 630 °C did not significantly change the strength; whereas, irradiation at calculated temperatures between 500 and 570 °C reduced the yield strength and ultimate tensile strength.

The ductility of the unirradiated V-15 Ti-7.5 Cr was not altered by the 30 day 550 °C anneal. Irradiation at calculated temperatures near 630 °C resulted in a slight ductility decrease of approximately 10%. The ductility was reduced up to 33% from that of the unirradiated 550 °C annealed control material by irradiation at calculated temperatures between 500 and 570 °C.

TABLE 3.3. Preirradiation History of V-20 Ti-7.5 Cr Tensile Specimens

Material	Heat Treatment	Grain Size, grains/mm ²
V-20 Ti	1 hr vacuum anneal at 900 °C, furnace cooled	11,000
V-15 Ti-7.4 Cr	1 hr vacuum anneal at 850 °C, furnace cooled	10,000

Material	Chemical Composition, wt%						
	V	Ti	Cr	O	N	H	C
V-20 Ti	bal.	19.65/20.02	-	0.083	0.030	0.0003	0.025
V-15 Ti-7.4 Cr	bal.	14.47/15.12	7.31/7.77	0.070	0.040	0.0014	-

TABLE 3.4. Room Temperature Tensile Properties of Vanadium-Base Alloys Irradiated in EBR-II

Material, wt%	Condition	Exposure at Center of Specimen, $n/cm^2 \times 10^{21}$	Calculated Irradiation Temperature, $^{\circ}C$
V-20 Ti	Unirradiated	-	-
V-20 Ti	Unirradiated, 30 days-550 $^{\circ}C$	-	-
V-20 Ti	Irradiated	1.8	540
V-20 Ti	Irradiated	3.2	600
V-20 Ti ^(a)	Irradiated	4.7	660
V-20 Ti-7.5 Cr	Unirradiated	-	-
V-20 Ti-7.5 Cr	Unirradiated, 30 days-550 $^{\circ}C$	-	-
V-15 Ti-7.5 Cr	Irradiated	1.6	500
V-15 Ti-7.5 Cr	Irradiated	3.2	570
V-15 Ti-7.5 Cr	Irradiated	4.8	630

Material, wt%	0.2% Offset, YS	UTS	Uniform Elongation,	Total Elongation,
	kg/mm ²	kg/mm ²	%	%
V-20 Ti	63.2	75.2	20.0	25.8
V-20 Ti	52.6	64.8	15.0	20.4
V-20 Ti	49.5	60.7	15.8	22.5
V-20 Ti	53.5	64.6	15.4	21.7
V-20 Ti ^(a)	57.2	67.6	13.6	19.0
V-20 Ti-7.5 Cr	64.4	72.3	15.0	20.6
V-20 Ti-7.5 Cr	67.2	76.3	15.0	19.9
V-15 Ti-7.5 Cr	61.3	72.2	10.4	14.8
V-15 Ti-7.5 Cr	58.1	68.9	10.0	14.2
V-15 Ti-7.5 Cr	67.0	80.0	13.6	18.0

(a) Average of two tests.

FIGURE 3.3. Axial Fluence Distribution for X013 Irradiation

Conclusions

Irradiation of V-20 Ti alloy at 50 °C to 1.4×10^{21} n/cm² ($E_n > 1$ MeV) reportedly⁽¹⁾ results in an increase in the room temperature yield strength of 44% and a decrease in the room temperature percent elongation of 90%. Testing of the same material at 650 °C resulted in properties similar to that of the unirradiated material. Irradiation effects, accordingly, are annealed out at the higher testing temperature.⁽¹⁾ Since the maximum irradiation temperature of the two vanadium base alloys tested was calculated at 660 °C and the changes in room temperature properties were considerably less, irradiation effects apparently were

annealed out at the higher irradiation temperature, with the observed changes primarily due to thermal effects.

Elevated Temperature Tests and Irradiations

Tensile testing of irradiated V-20 Ti and V-15 Ti-7.5 Cr at 550 °C is under way, and is to be followed by testing at 650 °C. The yield strength, ultimate tensile strength, uniform elongation, total elongation, and reduction in area are being evaluated as a function of grain size and strain rate. Strain rates are 0.2, 2, and 8%/min. The grain sizes of the V-20 Ti are 11,000, 1850, and 1100 grains/mm², and were obtained by annealing for 1 hr

in vacuum at 900, 1000 and 1100 °C, respectively. The grain sizes of the V-15 Ti-7.5 Cr are 10,000, 3600, and 1050 grains/mm², and were obtained by annealing for 1 hr in vacuum at 850, 1100, and 1250 °C, respectively.

TUBE-BURST TESTS

Eight inner capsule tubes V-15 Ti-7.5 Cr were on hand, and had initially been annealed for 1 hr at 1100 °C. Irradiation conditions are given in Table 3.2 and Figures 3.2 and 3.3. These were each cut into 8 sections about 4 in. long. A few tests have been made on the 4 in. sections at 550, 650, and 750 °C. Unirradiated control specimens were also tested. The results are shown in Table 3.5.

The maximum tangential stress was calculated from the equation for thick-walled tubes. Ductility has been indicated by the percent change in the diameter of the tube. In some cases, the tube shattered to such an extent that no diameter measurements were possible. The time at test temperature is shown because of its possible relation to annealing effects particularly at the higher temperatures. The variation in the time at temperature resulted from problems in getting a tight system.

At 550 °C in the maximum increase in burst strength is 27%; at 650 °C, 13%, and at 750 °C, no increase in strength exists due to the irradiation. At both 650 and 750 °C the unirradiated specimens showed about twice the

TABLE V. Burst Strength and Ductility of V-15 Ti-7.5 Cr Tubing

Test Temp., °C	Specimen Location ^(a)	Max. Tang. Stress		% ΔD/D		Time at Test Temp., min
		psi	kg/mm ²	Range	Avg	
550	Unirradiated	100,100	70.4	(b)	(b)	60
	Unirradiated	101,300	71.3	(b)	(b)	60
	-10 to -20 cm	113,800	80.0	0.8 - 2.3	1.4	130
	0 to -10 cm	128,000	90.0	1.8 - 2.4	2.0	90
	+50 to +60 cm	110,500	77.7	2.2 - 3.6	2.8	30
650	Unirradiated	97,800	68.8	7.4 - 8.8	8.1	45
	-10 to -20 cm	111,000	78.1	2.6 - 3.2	2.9	35
	0 to -10 cm	112,000	78.8	1.2 - 1.7	1.4	120
	+50 to +60 cm	88,000	61.9	(b)	(b)	60
750	Unirradiated	90,200	63.4	6.9 - 8.6	7.8	60
	+50 to +60 cm	89,300	62.8	2.2 - 6.8	3.7	95

(a) Specimen location above or below reactor midplane in cm.

(b) Shattered

ductility of the irradiated. Presumably, the shattered specimens had negligible ductility.

Bohm, et al.,⁽¹⁾ indicated that complete recovery of both strength and ductility of tensile specimens irradiated at 50 °C occurred with vanadium alloys tested at 650 and 750 °C. The

results of our biaxial tests are at variance with these data.

REFERENCE

1. H. Bohm, et al. "Irradiated Effects on the Mechanical Properties of Vanadium-Base Alloys," presented at the Sixty-Ninth Annual Meeting of ASTM, Atlantic City, New Jersey, June 27-July 1, 1966, Paper No. 75.

RADIATION-INDUCED PROPERTY CHANGES IN STRUCTURAL MATERIALS

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The radiation-induced mechanical property changes in AISI Type 347 SS after irradiation are being studied. The primary objective of the program is to determine the irradiation-induced embrittlement at elevated temperatures which occurs in stainless steels and nickel alloys. A metallographic examination, on an irradiated (2.1×10^{22} nvt) stainless steel specimen which had been tested at room temperature and then annealed at 1350 °C for one hour, revealed the presence of a second phase at the grain boundaries. The second phase could not be identified by standard etches on stainless steel. The degree of grain growth undergone by the irradiated specimen was minimal when compared to the extensive grain growth by an unirradiated control specimen which was annealed at the same conditions.

Four capsules were inserted into ETR to establish the effect of nickel on the irradiation embrittlement at elevated temperatures. These capsules contained specimens of a manganese austenitic stainless steel (nonnickel), pure nickel, nickel with minor impurities, and two cobalt alloys, one of which contained nickel, while the other did not.

An attempt is being made to correlate the effects of a predominantly fast flux and a mixed thermal and fast flux on the mechanical properties

of AISI 347 SS. To accomplish this, irradiations will have to be carried out in EBR-II and ETR on the same lot of materials.

The effect of 1000, 5000, and 10,000 hr heat treatments at 426, 538 and 649 °C on the mechanical properties of AISI 347 and 348 SS at room and elevated temperatures is being investigated. The specimens were annealed in air, nitrogen-0.5 vol% oxygen, and in one case, were periodically immersed in water every 1000 hr between anneals in nitrogen-0.5 vol% oxygen. The heat treatments did not result in any significant changes in the room temperature, 426, 538 and 649 °C tensile properties of the annealed material. The heat treatments at 426 °C resulted in increased strength and ductility for the 30% cold worked material. The room temperature ductility of the welded material was also found to be decreased by long time annealing at 426 °C. Metallographic examinations indicate that some sigma phase may be formed due to the long time heat treatment.

THE EFFECT OF IRRADIATION ON THE MECHANICAL PROPERTIES OF CLADDING MATERIALS

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The objectives of the cladding materials program at Battelle-Columbus are: (1) to determine the cause of limited

ductility at elevated temperatures (750 °C) of irradiated stainless steels and nickel alloys and (2) to determine the effect of large fluences on the mechanical properties of AISI Type 347 SS at pressure loop operating conditions in the ETR (up to 315 °C).

The magnitude of the radiation-induced property changes of AISI Type 347 SS, as measured by tensile tests, has been determined after fluences of 5.7×10^{21} , 1.1×10^{22} , 1.6×10^{22} , and 2.1×10^{22} nvt (1 MeV).⁽¹⁾ Unirradiated specimens tested in tension at a strain rate of 0.005 in./in./min at room temperature, 316 °C, and 750 °C all failed by ductile, transgranular fracture. Irradiated specimens (2.1×10^{22} nvt) tested under the same conditions at room temperature and 316 °C also failed by ductile transgranular fracture. However, irradiated specimens tested at 750 °C failed by brittle, intergranular fracture and, in addition, displayed a drastic reduction in ductility.

Previously reported⁽²⁾ electron microscopy has been performed to study fracture surfaces of both unirradiated specimens and specimens irradiated to fluences of 4×10^{20} nvt and 2.1×10^{22} nvt; all of these specimens had been tested in tension at 750 °C. The fracture surface of a ductile fracture is characterized by a dimpled appearance which results from tearing of the matrix. The fracture surface of brittle fracture is characterized by sharp facets which indicate grain boundary separation. The unirradiated specimen tested at 750 °C in tension was found to exhibit a completely ductile fracture. However, the specimen irradiated

to 4×10^{20} nvt had some areas where ductile fracture had occurred, while brittle fracture had occurred in other areas. The specimen that had received a fluence of 2.1×10^{22} nvt had failed by a completely brittle fracture when tested in tension at 750 °C. The fracture surfaces of the highly irradiated specimen also exhibited a bumpy appearance, the origin of which could not be explained.

A metallographic examination was performed on a section of an irradiated (2.1×10^{22} nvt) of 347 SS tensile specimen which had been tested at room temperature and then annealed at 1350 °C for one hour in vacuum. An unirradiated 347 SS specimen was annealed together with the irradiated specimen to establish a control as to possible microstructural changes. Figure 4.1 shows the unirradiated, unannealed material. Previous metallographic studies had indicated that irradiation at 50 °C did not alter the microstructure. Figure 4.2 shows that the one hour anneal at 1350 °C resulted in considerable grain growth of the unirradiated specimen. Figures 4.3 and 4.4 show the effect of the 1350 °C anneal on the microstructure of the irradiated 347 SS. A second phase is present in the grain boundaries, and grain growth has taken place although it is rather minimal compared to the grain growth in the unirradiated material. The nature of the second phase could not be determined by standard etches for stainless steels.

Initial work was started to grow single crystals of 347 SS for irradiation studies. In growing these single crystals by the zone leveling technique, special emphasis will be given

Neg 9A977

80X

FIGURE 4.1. Microstructure of the Unirradiated, 347 SS in the as Received Condition (Solution annealed)

Neg HC19663

80X

FIGURE 4.2. Microstructure of the Unirradiated 347 SS After Annealing for 1 hr at 1350 °C

Neg HC19671

80X

Etched

*FIGURE 4.3. Microstructure of the Irradiated
(2.1×10^{22} nvt) 347 SS After Annealing for
1 hr at 1350 °C*

Neg HC19659

400X

As Polished

*FIGURE 4.4. Microstructure of the Irradiated
(2.1×10^{22} nvt) 347 SS After Annealing for
1 hr at 1350 °C*

to possible composition changes, especially boron.

Preliminary studies were initiated to predict the magnitude of compositional change that can be expected in stainless steel due to neutron-induced transmutations during the long time irradiation. This study will consider the gaseous as well as the solid fission products. The effect of thermal neutrons on possible transmutations of the various silicon and nickel isotopes is being considered, initially as a means of evaluating the sensitivity of such calculations. If these studies prove successful, a computer program will be undertaken.

To study the role of nickel in the irradiation-induced elevated temperature embrittlement, four capsules were inserted into the ETR.⁽³⁾ These capsules contain the following tensile specimens.

<u>Capsule Designation</u>	<u>Number of Specimens</u>	<u>Materials</u>
BMI-24-23	9	UMCo-50
	9	Haynes #25
BMI-24-24	9	Tenelon
	6	Nickel-200
	3	Pure Nickel
BMI-24-25	9	UMCo-50
	9	Haynes #25
BMI-24-26	9	Tenelon
	6	Nickel-200
	3	Pure Nickel

Plans for Future Work

Irradiated (2.0 to 2.4×10^{22} nvt) 347 SS specimens will be tensile tested in the temperature range of 315 to 750 °C to establish the temperature where the fracture mode changes from transgranular to intergranular.

Further attempts will be made to identify the second phase in the grain

boundaries of the irradiated 347 SS annealed for 1 hr at 1350 °C. To identify the second phase, etches and microhardness measurements will be utilized. If it is possible to reduce the radiation level of the specimen to a low level by thinning or shielding, then a microprobe analysis of the second phase will be performed.

Attempts to grow single crystals of 347 SS will be continued. Also, specimens with different grain orientations will be fabricated. Single crystals and specimens with preferred grain orientations will then be irradiated.

After completion of the preliminary studies to predict neutron transmutation-induced composition changes in silicon and nickel, the program will be expanded to include other elements contained in Type 347 SS. To predict the compositional change of all the elements, a computer program will be utilized.

A capsule containing 347 SS tensile and fatigue specimens will be removed after it accumulates a fluence of 2.5×10^{22} nvt. The tensile and fatigue specimens will be tested at 315 and 750 °C.

A capsule containing the cobalt alloys (UMCo-50 and Haynes #25) and another capsule containing Tenelon and nickel specimens will be removed after they accumulate a fluence of 8×10^{20} nvt. These specimens will be tested mainly at elevated temperatures after irradiation. The test results are expected to show the role of nickel in irradiation-induced elevated temperature embrittlement.

CORRELATION OF IRRADIATION EFFECTS
ON MECHANICAL PROPERTIES AS CAUSED
BY PREDOMINANTLY FAST FLUX AND A MIXED
FAST AND THERMAL FLUX

M. Kangilaski and R. A. Wullaert

The objective of the program is to compare the effect of irradiation on the mechanical properties of cladding materials in a predominantly fast flux and in a mixed fast and thermal flux. To accomplish this, specimens of the same lot of Type 347 SS that were irradiated in the ETR to fluences of up to 3×10^{22} nvt will be irradiated in the EBR-II to equivalent fluences. The irradiated specimens will be tested at room temperature, 315 and 750 °C to correlate the effects of predominantly fast neutrons, and mixed thermal and fast neutrons, on the mechanical properties. Since the EBR-II irradiation will take place at elevated temperatures (400 and 650 °C) and the original ETR irradiation was conducted at 50 °C, there is a possibility that the different irradiation temperatures will not allow a direct comparison of the postirradiation mechanical properties. Therefore, another capsule will be irradiated in ETR with the specimens being at 400 and 650 °C during irradiation so that a direct correlation between the effects of irradiation on the mechanical properties of 347 SS can be made. These specimens will be tested at room temperature, 315 °C, and 750 °C after irradiation.

Sheet specimens will be prepared from an irradiated Fermi wrapper tube which has received a fluence of 3×10^{20} nvt in a predominantly fast flux. This wrapper tube is made of 347 SS, is 2 1/2 in. square, has a weld down one side, and has a thickness of

100 mils. Specimens will be cut from both the longitudinal and transverse directions and some specimens will contain the weld in their gage section. These specimens will be tested in tension at room temperature, 315, 650, and 750 °C in the asirradiated condition and after annealing. Later tests will be performed in creep and on notched tensile specimens. Results from these tests will be compared to irradiation data obtained on Type 347 SS in a mixed thermal and fast flux spectrum and to unirradiated control specimens from Fermi wrapper tubes.

The tensile specimens which are to be inserted in EBR-II and ETR have been machined.

EVALUATION OF TYPE 347 AND 348
STAINLESS STEEL

D. E. Price and D. P. Moak

The objective of this program is to investigate the influence of long-time heat treatments at 426, 538, and 649 °C on selected properties of AISI Type 347 SS. The effect of two gaseous atmospheres, periodic immersion in water, and a tensile stress during heat treatment on the mechanical properties of Type 347 SS in three conditions (mill annealed, cold worked, and welded) is being investigated. Annealed and cold-worked Type 347 SS rod from the same heat was machined into tensile specimens. Tensile specimens are being annealed for 1000, 5000, 10,000 and 15,000 hr at 426, 538, and 649 °C under stresses of 10,000 and 5,000 psi, respectively, for time periods of 5,000 and 10,000 hr.

Tensile tests at room temperature, 426, 538, and 649 °C and metallographic examinations of the tested samples are

being used to evaluate the influence of the heat treatments and variables. Attempts will be made to correlate observed structural changes, such as carbide precipitation and possible sigma formation, with changes in mechanical properties.

Results of the tensile tests are given in Tables 4.1 through Table 4.15. Tables 4.1, 4.2, and 4.3 give the mechanical properties of annealed Type 347 SS heat treated at 426, 538, and 649 °C, respectively. In general, none of the heat treatments changed the mechanical properties significantly.

Tables 4.4 and 4.5 give the mechanical properties of annealed Type 348 SS after heat treatments at 538 and 649 °C, respectively. Again there is no significant change in the mechanical properties, and the strength and ductility

are very close to that of Type 347. Both Type 347 and 348 show a slight loss of elongation with increased annealing time at 1200 °F.

Table 4.6, 4.7, and 4.8 are for Type 347 SS cold worked 30% prior to the heat treatments. The 426 °C heat treatment caused an increase in strength in general but a significant drop in elongation in the 426 °C tests. After 5,000 hr at 426 °C the elongation dropped to roughly 9% but a portion of the ductility seems to be recovered with increasing time.

The 538 °C and 649 °C heat treatments both decreased the strength of cold worked material through stress relief and recrystallization. The elongation changed little with time at 538 °C and increased slightly at 649 °C.

TABLE 4.1. Annealed Type 347 SS Heat Treated at 800 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
1-8-1-R	1,000	RT	97,500	57,900	58.5	80.4
1-8-5-R	5,000	RT	--	--	--	--
1-8-10-R	10,000	RT	95,700	57,800	62.0	80.4
1-8-15-R	15,000	RT	95,600	57,400	63.0	80.4
1-R-0-8	0	800	61,100	42,000	32.3	70.7
1-8-1-8	1,000	800	62,000	43,100	30.6	80.3
1-8-5-8	5,000	800	--	--	--	--
1-8-10-8	10,000	800	64,600	46,000	33.0	71.3
1-8-15-8	15,000	800	65,000	44,200	35.0	70.5
1-R-0-10	0	1,000	46,100	31,300	33.6	69.8
1-8-1-10(a)	1,000	1,000	47,000	37,700	30.7	72.2
1-8-5-10	5,000	1,000				
1-8-10-10	10,000	1,000				
1-8-15-10	15,000	1,000				
1-R-0-12	0	1,200	51,700	36,100	29.1	71.2
1-8-1-12	1,000	1,200	51,400	36,300	44.1	69.0
1-8-5-12	5,000	1,200				
1-8-10-12	10,000	1,200				
1-8-15-12	15,000	1,200				

(a) Based on one sample

TABLE 4.2. Annealed Type 347 SS Heat Treated at 1,000 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
1-10-1-R	1,000	RT	97,400	57,000	60.9	81.3
1-10-5-R	5,000	RT	95,300	57,100	63.0	80.5
1-10-10-R	10,000	RT	94,800	54,100	62.0	79.6
1-10-15-R	15,000	RT	96,600	53,200	60.0	79.6
1-R-0-8	0	800	61,100	42,000	32.3	70.7
1-10-1-8	1,000	800	68,700	48,900	29.4	71.3
1-10-5-8	5,000	800	65,200	44,300	33.0	49.8
1-10-10-8	10,000	800	61,900	44,100	32.0	72.4
1-10-15-8	15,000	800	63,200	43,100	33.0	72.2
1-R-0-10	0	1,000	46,100	31,300	33.6	69.8
1-10-1-10	1,000	1,000	44,800	29,900	31.8	69.4
1-10-5-10	5,000	1,000				
1-10-10-10	10,000	1,000				
1-10-15-10	15,000	1,000				
1-R-0-R	0	1,200	51,700	36,100	29.1	71.2
1-10-1-12	1,000	1,200	--	--	--	--
1-10-5-12	5,000	1,200				
1-10-10-12	10,000	1,200				
1-10-15-12	15,000	1,200				

TABLE 4.3. Annealed Type 347 SS Heat Treated at 1,200 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
1-12-1-R(a)	1,000	RT	99,900	56,400	55.8	81.0
1-12-5-R	5,000	RT	95,700	47,300	57.0	68.8
1-12-10-R	10,000	RT	97,200	49,100	54.0	65.1
1-12-15-R	15,000	RT	98,600	49,700	50.0	61.9
1-R-0-8	0	800	61,100	42,000	32.3	70.7
1-12-1-8	1,000	800	60,500	41,900	28.1	80.6
1-12-5-8	5,000	800	62,100	39,200	32.0	64.3
1-12-10-8	10,000	800	61,500	36,600	33.0	64.4
1-12-15-8	15,000	800	60,900	34,700	32.0	61.0
1-R-0-10	0	1,000	46,100	31,300	33.6	69.8
1-12-1-10	1,000	1,000	45,200	33,100	29.2	69.4
1-12-5-10	5,000					
1-12-10-10						
1-12-15-10						
1-R-0-12	0	1,200	51,700	36,100	29.1	71.2
1-12-1-12	1,000	1,200	50,800	39,300	37.3	72.2
1-12-5-12						
1-12-10-12						
1-12-15-12						

(a) Based on one sample

TABLE 4.4. Annealed Type 348 SS Heat Treated at 1,000 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
2-R-0-R(a)	0	RT	100,400	62,100	53.5	75.0
2-10-1-R	1,000	RT	99,400	59,000	56.6	52.1
2-10-5-R	5,000	RT	99,700	61,400	57.0	74.7
2-10-10-R	10,000	RT	100,700	60,700	57.0	73.4
2-10-15-R	15,000	RT	102,900	62,000	55.0	72.0
2-R-0-10	0	1,000	63,200	39,500	36.9	76.8
2-10-1-10	1,000	1,000	66,900	42,200	31.6	62.8
2-10-5-10	5,000	1,000				
2-10-10-10	10,000	1,000				
2-10-15-10	15,000	1,000				
2-R-0-12(a)	0	1,200	53,400	33,900	45.3	61.2
2-10-1-12	1,000	1,200	54,200	38,200	33.9	71.7
2-10-5-12	5,000	1,200				
2-10-10-12	10,000	1,200				
2-10-15-12	15,000	1,200				

(a) Based on one sample

TABLE 4.5. Annealed Type 348 SS Heat Treated at 1,200 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
2-R-0-R	0	RT	100,400	62,100	53.5	75.0
2-12-1-R	1,000	RT	98,300	53,600	54.3	65.2
2-12-5-R	5,000	RT	98,900	45,500	47.0	54.7
2-12-10-R	10,000	RT	100,000	45,700	44.0	52.2
2-12-15-R	15,000	RT	100,200	45,300	42.0	49.4
2-R-0-10	0	1,000	63,200	39,500	36.9	76.8
2-12-1-10	1,000	1,000	62,100	41,200	27.0	60.1
2-12-5-10	5,000	1,000				
2-12-10-10	10,000	1,000				
2-12-15-10	15,000	1,000				
2-R-0-12	0	1,200	53,400	33,900	45.3	61.2
2-12-1-12	1,000	1,200	54,600	37,600	33.9	65.9
2-12-5-12	5,000	1,200				
2-12-10-12	10,000	1,200				
2-12-15-12	15,000	1,200				

TABLE 4.6. Cold Worked Type 347 SS Heat Treated at 800 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
6-R-0-R	0	RT	132,100	105,500	23.5	72.8
6-8-1-R	1,000	RT	141,200	119,800	25.2	64.5
6-8-5-R	5,000	RT	148,400	121,900	26.0	68.2
6-8-10-R	10,000	RT	147,800	121,600	27.9	70.4
6-8-15-R	15,000	RT	144,800	119,900	29.0	68.3
6-R-0-8	0	800	127,200	90,600	25.0	50.5
6-8-1-8	1,000	800	110,800	96,800	12.2	55.1
6-8-5-8	5,000	800	110,000	98,000	9.0	52.0
6-8-10-8	10,000	800	113,200	90,800	13.0	49.5
6-8-15-8	15,000	800	--	--	--	--
6-R-0-10	0	1,000	89,100	78,200	11.5	47.7
6-8-1-10	1,000	1,000	102,000	89,900	11.1	51.3
6-8-5-10	5,000	1,000				
6-8-10-10	10,000	1,000				
6-8-15-10	15,000	1,000				
6-R-0-12	0	1,200	86,600	68,800	11.0	37.7
6-8-1-12	1,000	1,200	85,900	69,700	12.8	28.9
6-8-5-12	5,000	1,200				
6-8-10-12	10,000	1,200				
6-8-15-12	15,000	1,200				

TABLE 4.7. Cold Worked Type 347 SS Heat Treated at 1,000 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
6-R-0-R	0	RT	132,100	105,500	23.5	72.8
6-10-1-R	1,000	RT	141,600	116,700	28.3	70.7
6-10-5-R	5,000	RT	141,500	108,200	28.0	63.1
6-10-10-R	10,000	RT	133,400	96,200	22.0	45.5
6-10-15-R	15,000	RT	130,400	93,200	25.0	44.0
6-R-0-8(a)	0	800	114,800	100,000	13.0	57.3
6-10-1-8	1,000	800	109,400	96,200	14.8	53.2
6-10-5-8	5,000	800	108,600	90,800	16.0	49.8
6-10-10-8	10,000	800	99,400	78,200	16.0	44.0
6-10-15-8	15,000	800	94,300	68,900	17.0	44.3
6-R-0-10	0	1,000	89,100	78,200	11.5	47.2
6-10-1-10	1,000	1,000	99,500	88,300	12.5	48.8
6-10-5-10	5,000	1,000				
6-10-10-10	10,000					
6-10-15-10	15,000					
6-R-0-12	0	1,200	86,600	68,800	11.0	37.7
6-10-1-12	1,000	1,200	86,100	71,800	11.9	16.8
6-10-5-12	5,000	1,200				
6-10-10-12	10,000	1,200				
6-10-15-12	15,000	1,200				

(a) Based on one sample

TABLE 4.8. Cold Worked Type 347 SS Heat Treated at 1,200 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
6-R-0-R	0	RT	132,100	105,500	23.5	72.8
6-12-1-R	1,000	RT	127,200	90,600	25.0	50.5
6-12-5-R	5,000	RT	110,900	53,200	32.6	44.0
6-12-10-R	10,000	RT	106,000	44,200	37.0	45.6
6-12-15-R	15,000	RT	107,300	44,800	38.0	44.9
6-R-0-8	0	800	114,800	100,000	13.0	57.3
6-12-1-8	1,000	800	87,300	69,500	13.1	40.1
6-12-5-8	5,000	800	74,400	39,800	19.0	43.0
6-12-10-8	10,000	800	70,600	31,800	25.0	51.0
6-12-15-8	15,000	800	70,200	31,400	25.0	46.0
6-R-0-10	0	1,000	89,100	78,200	11.5	47.2
6-12-1-10	1,000	1,000	62,900	51,500	11.7	46.1
6-12-5-10	5,000	1,000				
6-12-10-10	10,000	1,000				
6-12-15-10	15,000	1,000				
6-R-0-12	0	1,200	86,600	68,800	11.0	37.7
6-12-1-12	1,000	1,200	68,400	53,800	19.8	24.2
6-12-5-12	5,000	1,200				
6-12-10-12	10,000	1,200				
6-12-15-12	15,000					

TABLE 4.9. Welded Type 347 SS Heat Treated at 800 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
3-R-0-R	0	RT	84,700	38,600	32.6	26.2
3-8-1-R	1,000	RT	82,600	39,200	28.2	35.2
3-8-5-R	5,000	RT	85,200	39,000	30.0	41.8
3-8-10-R	10,000	RT	85,300	38,700	26.0(a)	42.3
3-8-15-R	15,000	RT				
3-R-0-8	0	800	58,500	27,600	25.7	67.2
3-8-1-8	1,000	800	60,800	27,900	22.3	34.6
3-8-5-8	5,000	800	62,700	24,500(a)	35.0	73.0
3-8-10-8(a)	10,000	800	61,100	25,100	27.0	40.4
3-8-15-8	15,000	800				
3-R-0-10	0	1,000	56,500	26,100	27.6	65.2
3-8-1-10	1,000	1,000	54,900	25,200	17.8	36.7
3-8-5-10	5,000	1,000				
3-8-10-10	10,000	1,000				
3-8-15-10	15,000	1,000				
3-R-0-12	0	1,200	49,200	25,700	25.3	34.3
3-8-1-12	1,000	1,200	50,200	25,600	27.7	54.1
3-8-5-12	5,000	1,200				
3-8-10-12	10,000	1,200				
3-8-15-12	15,000	1,200				

(a) Based on one sample

Tables 4.9, 4.10, and 4.11 contain the mechanical property data on the welded Type 347 SS. The 649 °C heat treatments have caused a decrease in elongation with time; however, even after 15,000 hr, there was about 18% elongation in one inch at room temperature. Whether the trend toward decreased elongations will continue, embrittlement will occur after even longer periods of time is unknown. Other mechanical properties were not significantly affected by the heat treatments.

Tables 4.12 and 4.13 give the room temperature mechanical properties of Type 347 SS that was loaded during the heat treatment. Samples heated at 538 °C and 649 °C were stressed at 10,000 psi and 5,000 psi respectively.

These samples performed mechanically almost identically to the Type 347 SS samples given the same heat treatments without stress.

Tables 4.14 and 4.15 list the mechanical property data for Type 347 SS heat treated at 649 °C under special conditions. The data in Table 4.14 are for samples heated in an atmosphere containing 1/2 vol% O₂-99 1/2 vol% N₂. Data reported in Table 4.15 are from samples that were periodically (every 1000 hr) removed from the furnace, air cooled, immersed in water, and reheated to 649 °C. Neither of these heat treatments changed the mechanical properties significantly from those obtained on samples heated continuously at 649 °C in air.

TABLE 4.10. Welded Type 347 SS Heat Treated at 1,000 °F

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
3-R-0-R	0	RT	84,700	38,600	32.6	26.2
3-10-1-R	1,000	RT	85,700	40,200	32.4	37.4
3-10-5-R	5,000	RT	88,900	40,500	37.0	47.8
3-10-10-R	10,000	RT	85,100	41,100	27.0	40.0
3-10-15-R	15,000	RT	85,800	38,600	28.0	36.9
3-R-0-8	0	800	58,500	27,600	25.7	67.2
3-10-1-8(a)	1,000	800	61,800	28,300	29.9	71.3
3-10-5-8	5,000	800	59,800	27,200	20.0	64.3
3-10-10-8(a)	10,000	800	58,900	30,300	20.0	41.2
3-10-15-8(a)	15,000	800	58,300	26,600	18.0	34.3
3-R-0-10	0	1,000	56,500	26,100	27.6	65.2
3-10-1-10	1,000	1,000	56,500	27,400	21.7	38.9
3-10-5-10	5,000	1,000				
3-10-10-10	10,000	1,000				
3-10-15-10	15,000	1,000				
3-R-0-12	0	1,200	49,200	25,700	25.3	34.3
3-10-1-12	1,000	1,200	51,000	24,500	26.3	59.8
3-10-5-12	5,000	1,200				
3-10-10-12	10,000	1,200				
3-10-15-12	15,000	1,200				

(a) Based on one sample

TABLE 4.11. Welded Type 347 SS Heat Treated at 1,200 °F

Identifi- cation	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
3-R-0-R	0	RT	84,700	38,600	32.6	26.2
3-12-1-R	1,000	RT	88,600	41,700	34.1	34.0
3-12-5-R	5,000	RT	89,300	38,000	27.0	37.5
3-12-10-R	10,000	RT	89,400	35,400	24.0	31.2
3-12-15-R	15,000	RT	86,700	39,000	18.0	19.5
3-R-0-8	0	800	58,500	27,600	25.7	67.2
3-12-1-8(a)	1,000	800	59,000	29,100	23.2	22.2
3-12-5-8	5,000	800	59,600	29,800	29.0	65.7
3-12-10-8	10,000	800	59,200	29,100	28.0	64.4
3-12-15-8	15,000	800	60,200	29,800	25.0	61.0
3-R-0-10	0	1,000	56,500	26,100	27.6	65.2
3-12-1-10	1,000	1,000	56,600	28,500	27.9	58.9
3-12-5-10	5,000	1,000				
3-12-10-10	10,000	1,000				
3-12-15-10	15,000	1,000				
3-R-0-12	0	1,200	49,200	25,700	25.3	34.3
3-12-1-12	1,000	1,200	48,900	26,900	24.6	52.3
3-12-5-12	5,000	1,200				
3-12-10-12	10,000	1,200				
3-12-15-12	15,000	1,200				

(a) Based on one sample

TABLE 4.12. Annealed Type 347 SS Heat Treated at 1,000 °F While Stressed at 10,000 psi

Identifi- cation	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
7-10-5-R	5,000	RT	89,300	55,000	63.0	81.0
7-10-10-R	10,000	RT	96,800	57,500	61.0	79.0
1-R-0-12	0	1,200	51,700	36,100	29.1	71.2
7-10-5-12	5,000	1,200				
7-10-10-12	10,000	1,200				

TABLE 4.13. Annealed Type 347 SS Heat Treated at 1,200 °F While Stressed at 5,000 psi

Identifi- cation	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
7-12-5-R	5,000	RT	97,500	51,000	53.0	66.2
7-12-10-R	10,000	RT	97,800	47,100	53.0	61.3
1-R-0-12	0	1,200	51,700	36,100	29.1	71.2
7-12-5-12	5,000	1,200				
7-12-10-12	10,000	1,200				

TABLE 4.14. *Annealed Type 347 SS Heat Treated at 1,200 °F in a N₂ + 1/2 vol% O₂ Atmosphere*

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
4-12-1-R	1,000	RT	95,500	52,400	58.5	78.6
4-12-5-R	5,000	RT	97,100	48,900	52.0	58.7
4-12-10-R	10,000	RT	95,800	44,900	50.0	50.0
4-12-15-R	15,000	RT	98,600	48,600	45.0	50.6
1-R-0-12	0	1,200	51,700	36,100	29.1	71.2
4-12-1-12	1,000	1,200	49,400	36,400	36.3	73.1
4-12-5-12	5,000	1,200				
4-12-10-12	10,000	1,200				
4-12-15-12	15,000	1,200				

TABLE 4.15. *Annealed Type 347 SS Heat Treated at 1,200 °F, Air Cooled and Immersed in Water Every 1,000 hr, and Reheated*

Identification	Time, hr	Testing Temperature, °F	Tensile Strength, psi	0.2% Yield Strength, psi	% Elongation	% Reduction in Area
1-R-0-R	0	RT	95,200	55,400	61.5	69.9
5-12-1-R	1,000	RT	94,600	51,600	58.5	81.2
5-12-5-R	5,000	RT	97,100	52,400	56.0	71.5
5-12-10-R	10,000	RT	96,700	46,700	52.0	65.1
5-12-15-R	15,000	RT	99,900	50,300	51.0	62.3
1-R-0-12	0	1,200	51,700	36,100	29.1	71.2
5-12-1-12	1,000	1,200	50,500	36,500	37.5	67.7
5-12-5-12	5,000	1,200				
5-12-10-12	10,000	1,200				
5-12-15-12	15,000	1,200				

The 15,000 hr samples were examined metallographically and a more detailed examination of selected 10,000 hr samples was carried out. It was noted previously that the samples heated at 538 and 649 °C contained a fine dispersed precipitate that came out preferentially at the grain boundaries. Earlier this precipitate was thought to be (Cr,Fe)₂₃C₆. However, by comparing the volume percent of this

phase present (10.5 vol% by point count) in the cold-worked Type 347 SS sample heated 15,000 hr at 1200 °F and the carbon content of the alloy (0.04 wt% C), it was apparent that there was not enough carbon in the alloy to account for the quantity of precipitate.

In the Type 348, large (Nb,Ta)C exists as a result of the thermal history of the annealed material. After receiving a 649 °C heat treatment, the

new precipitate forms randomly throughout the alloy with some particles very near the (Nb,Ta)C particles originally in the alloy. These (Nb,Ta)C particles are frequently fractured as a result of mechanical rolling at the mill prior to shipment and are readily identified. The new precipitate etches much differently from the (Nb,Ta)C and, in general, seems more reactive. Although at the present time it is not substantiated, it is believed that the new precipitate is sigma, which is essentially a FeCr intermetallic compound. At the present time work is underway to identify the new phase by X-ray diffraction by extraction techniques. Unfortunately, there is not a large enough quantity in the heat treated alloys to give a good pattern.

Several other metallographic observations parallel the behavior commonly exhibited by sigma. For example, cold-worked material contains more of the precipitate after heat treatment than the annealed material. This can be seen in Figure 4.5. In Figure 4.6 most of the precipitate is seen adjacent to mechanical threads and not in the matrix. This sample was heated at 538 °C which is below the range sigma is normally observed. However, it is suggested that the heavy deformation near the threads promoted the formation of the precipitate.

In Figure 4.7, which shows the surface of a sample not heavily deformed, there is a zone adjacent to the surface that is free of the precipitate. Since Cr_2O_3 forms preferentially on stainless steels, it appears as if this zone is depleted in chromium to a sufficient extent to prevent the formation of the precipitate.

This same precipitate also forms in the weld metal. Figure 4.8 shows this phase along the grain boundaries and intergranular fracture. Welds in stainless steel frequently contain ferrite, the bcc form of iron. It is established that the presence of ferrite promotes the formation of sigma. In fact, it has been proposed that in order for sigma to form in austenite, the bcc structure must be obtained as an intermediate step.

Sigma present in a steel can affect the mechanical properties in several ways, depending on the amount, size, and shape. In the steels with higher chromium contents than Type 347, there is frequently a large loss in ductility. In the alloys studied in this program, there appears to be a maximum of about 5 vol% of uniformly dispersed particles which are roughly 3 μ in the largest dimension. Based on the mechanical property data, the precipitate is apparently having little effect on the alloys, with the possible exception of the welded samples. However, even after 15,000 hr the welds exhibit 18% elongation. Improved weld properties could possibly be obtained by giving the weld a ferrite-to-austenite heat treatment prior to the low temperature exposures.

Plans for Future Work

Tensile tests will be carried out at 538 and 649 °C. A few remaining lower temperature tests will be completed. Metallographic work on the 15,000 hr samples will be completed and an effort will be made to identify the precipitate.

Neg 6B971

600X

Annealed

Neg 6B977

600X

Cold Worked

FIGURE 4.5. Structure of Type 347, Both Annealed and Cold Worked, and Then Heated 15,000 hr at 649 °C.

Neg 6B970

600X

*FIGURE 4.6. Surface Near Threaded
Portion of Sample Heated 15,00 hr
at 538 °C*

Neg 6B972

600X

*FIGURE 4.7. Surface of Type 347 SS
Heated in Air for 15,000 hr at 649 °C.*

Neg 6B975

80X

FIGURE 4.8. Weld Zone and Room Temperature Fracture Surface of a Welded Type 347 SS Heated 15,000 hr at 649 °C.

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IRRADIATION EFFECTS ON REACTOR STRUCTURAL MATERIAL PROGRAM

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OPTICAL AND ELECTRON MICROSCOPIC
STUDY OF THE EFFECTS OF NEUTRON
IRRADIATION ON IRON

J. S. Bryner

A study was made of substructures produced in unirradiated and 2×10^{18} nvt-irradiated ($E > 1$ MeV) Ferrovac E by tensile strain below the ductile-to-brittle transition temperature of the irradiated iron. Electropolished polycrystalline specimens approximately 0.020 in. thick were subjected to simple bending at 78 °K to limited deflections calculated to produce a surface stress slightly higher than the yield stress. Residual deflections, due to plastic strain, were measured. The as-bent surfaces strained in tension were etched lightly and examined by optical microscope. Then the half of each specimen strained in compression was ground away, and the remaining half was electrolytically thinned for electron transmission examination.

Examination in the optical microscope disclosed that in both irradiated and unirradiated specimens, the plastic strain was limited to a narrow band approximately equal in width to the specimen thickness. The amount of local deformation varied from grain to grain and from area to area within individual grains. The bands contained linear strain markings which were revealed by oblique illumination to be surface ledges. The ledges were generally

aligned at angles less than 45° from the bending axis, but otherwise showed no consistent pattern. The heavier markings sometimes continued straight across several grains, sometimes showed sharp or curved changes of direction at grain boundaries or within grains, and sometimes followed grain boundaries for short distances. Within individual grains, additional shallower strain markings were observed which were sometimes straight and parallel and sometimes wavy and intersecting. Some grains were obviously distorted but showed no surface markings. The markings in irradiated and unirradiated specimens were quite similar. In particular, the reported findings of other investigators that the strain markings in unirradiated iron are shallow and uniformly distributed; whereas, could the markings in irradiated iron which are heavier and more widely spaced, could not be duplicated. On all irradiated specimens strained plastically to approximately 0.5% surface elongation within the deformed band, surface cracks were found. No cracks were found in unirradiated specimens after higher strains, indicating that the irradiated specimens were indeed embrittled. All of the surface cracks detected were greater than one grain diameter in length, so that the site of crack initiation could not be pinpointed. Many of the cracks were discontinuous, suggesting that the crack was initiated at a point beneath the

surface. Long cracks were transgranular along most of their length, but always contained intercrystalline segments.

Figure 5.1a shows an as-bent area on an irradiated specimen containing typical strain markings, distorted grains, and a crack. Figure 5.1b shows the same area after light wet lapping to flatten the surface plus a light etch to show grain boundaries. The crack lies partially within one grain parallel to the boundary; after interruption by a thin web of metal, it runs along the boundary between the first grain and a second grain; then after interruption by a portion of a third grain which is heavily distorted, the crack runs partially through a fourth grain. Although the metal at the ends of the crack is heavily deformed, the metal along the sides of the crack shows no deformation, indicating failure by cleavages.

Figure 5.1c shows the same area after electropolishing to remove approximately 0.0002 in. from the surface plus a light etch to bring out grain and twin boundaries. Strain markings due to deformation by slip are eliminated. Comparison of Figures 5.1a and 5.1c shows that a continuous ledge crossing several grains may be due to twinning in some grains and to slip in others. It is apparent that the first and fourth grains adjacent to the crack in Figure 5.1a have a common boundary beneath the surface. Extension of the crack in Figure 5.1c suggests that the crack may have been initiated along this common subsurface boundary. A small grain boundary crack has appeared in the upper

right corner of Figure 5.1c, adding further evidence that cracking is initiated at subsurface grain boundaries. This crack was apparently initiated at a position where Figure 5.1a shows that ledges, due to slip in one of the adjacent grains, end on the grain boundary.

Electron-transmission microstructures produced in irradiated and unirradiated specimens by tensile strain at 78 °K were quite similar to each other and to those previously observed after equivalent strains in compression except for greater variations in the amount of local deformation. After strains up to approximately 0.5% surface elongation, the structures contained twins and narrow bands or lines of concentrated dislocations. Because of twinings and slip, these bands or lines probably corresponded to the surface ledges. Instances of transition from twinning to slip across a grain boundary were noted. High concentrations of dislocations occurred where twins impinged on grain boundaries indicating high local strains. Although these dislocations lay mostly within the contiguous grains, some cases of high concentrations of dislocations within the grain boundary suggested slip along the grain boundary. Additional dislocations distributed randomly throughout the grains probably corresponded to the case of surface grains which were distorted without producing strain markings. In regions of higher strain, networks of relatively straight dislocations were formed within the grains. Representative structures resulting from light

688X

FIGURE 5.1. Deformed and Cracked Area on the Surface of a Ferrovac E Specimen Irradiated to 2×10^{18} nvt ($E > 1$ MeV) and Strained in Tension by Bending at 78 °K. (Nital Etch)

- a. Strain Markings and Interrupted Crack on Surface as Bent*
- b. Same Area After Mechanical Polish to Remove Ledges Showing Relationship Between Crack and Grain Boundaries*
- c. Same Area After Electrolytic Polish to Remove Approximately 0.0002 in. Showing Crack Extension, Twins, and an Additional Intergranular Crack*

strain at 78 °K in unirradiated and irradiated specimens are shown in Figures 5.2 and 5.3.

Attempts to examine the metal in the grains adjacent to cracks have proved difficult, but in a few instances, visual examination of a very narrow region was possible. In these few cases the metal on each side of a transgranular crack and grain boundary crack contained relatively few

dislocations (suggesting brittle failure), but the metal within webs that interrupted cracks contained very high concentrations of dislocations. The only apparent difference between the electron-transmission microstructures of irradiated and unirradiated specimens was that the unirradiated specimens contained "internal dislocation sources" while the irradiated specimens did not.

a.

b.

c.

d.

FIGURE 5.2. Electron Transmission Structure in Unirradiated Ferrovac E Strained Less than 0.5% at 78 °K, Showing Grain Boundaries, Twins, Deformation Bands, Internal Dislocation Sources, and Arrays of Relatively Straight Dislocations

a.

b.

c.

d.

FIGURE 5.3. Electron Transmission Structures in Ferrovac E Irradiated to 2×10^{18} nvt and Strained Less than 0.5% at 78 °K, Showing Similarity to Unirradiated Structures Except for Absence of Internal Dislocation Sources

This same difference between irradiated and unirradiated specimens has been previously observed after tensile or compressive strain at ambient temperature or compressive strain at 78 °K. Attempts have been made to learn more about the "internal dislocation sources" and their relationship to the deformation behavior of iron. These efforts have indicated that the structures are almost certainly due to ϵ -carbide precipitates in the iron. No ϵ -carbide precipitates are visible in Ferrovac E after furnace cooling from 700 °C, and no "internal dislocation sources" are produced by straining after holding for several days at ambient temperature. However, if the furnace-cooled iron is held for months at ambient temperature, the ϵ -carbides can apparently be precipitated by high elastic stress. Thus, it appears that some segregation of carbon in the supersaturated solid solution is necessary before precipitation can be induced by stress. The ϵ carbides are visible as small platelets lying parallel to the {100} planes of the iron. After light plastic strains, they appear as small patches of concentrated dislocations. After heavier plastic strain these patches interact with dislocations from other sources. Heating at 350 °C for two hours causes the ϵ -carbides to disappear, but does not remove the patches of concentrated dislocations, which indicates that the ϵ -carbides do serve as internal dislocation sources. Although the resulting dislocations do not appear to move very far from the source, they should help to distribute the initial deformation during

yielding of unirradiated iron. They might be expected to cause an increased rate of strain hardening by interfering with the motion of slip dislocations.

Fast-neutron irradiation ($E > 1$ MeV) to a fluence of 1×10^{16} nvt at 60 °C causes precipitation of ϵ -carbides in Ferrovac E furnace-cooled or quenched after annealing at 700 °C. It has been found that the ϵ -carbides precipitated by this initial irradiation in Ferrovac E, containing 30 ppm carbon, and in iron, containing up to 100 ppm carbon, are completely removed from the electron-transmission microstructure by a subsequent irradiation at 60 °C to 2×10^{18} nvt. This observation indicates that during the irradiation the carbon combined with radiation defects to produce complex defects in which the carbon atoms are bound more strongly than in the ϵ -carbide. The observation also indicates that internal dislocation sources are not observed in Ferrovac E strained after irradiation to 2×10^{18} nvt because the ϵ -carbides are decomposed by the irradiation.

DISCUSSION

Since the internal friction studies of Wagenblast and Damask⁽¹⁾ showed that carbon is rapidly removed from solid solution in iron by fast neutron irradiation at reactor ambient temperatures, it is evident that the combination of carbon atoms with radiation defects during the irradiation depletes the iron of available carbon. Fast,⁽²⁾ Low and Feustel,⁽³⁾ and a number of subsequent investigators have shown that removal of available carbon from a relatively pure

iron promotes low fracture stress and intercrystalline fracture at low temperatures, presumably due to other impurities such as oxygen or phosphorus segregated at grain boundaries. The condition is aggravated by twinning at low temperatures by stress concentrations where twins impinge on grain boundaries. The present microscopic results indicate that low-temperature fracture in radiation embrittled iron is initially intercrystalline, and confirm that high stress concentrations exist where twins impinge on grain boundaries. Therefore the results suggest that fast-neutron irradiation to 2×10^{18} nvt at reactor ambient temperature reduces the low-temperature fracture stress of Ferrovac E by depleting the iron of available carbon through combination of carbon with radiation defects.

The results indicate that there are no obvious differences between the electro-transmission microstructures of unirradiated and radiation-embrittled specimens of Ferrovac E after equivalent strains at 78 °K, which might explain the abrupt increase in the yield strength of the radiation-embrittled Ferrovac E at the ductile-to-brittle transition temperature. Crystallographic studies of these structures are in progress to seek evidence for effects of radiation on the mechanism of plastic deformation at low temperature.

FIELD ION MICROSCOPY STUDY OF DEFECT STRUCTURE

M. Attardo

Studies of radiation damage in platinum and tungsten have continued, using a liquid helium cooled field ion microscope. Correlations between the den-

sity and sizes of depleted zones introduced by fast neutron irradiation and yield stress have been made. In platinum it has been shown that the smaller sized depleted zones are most effective in hardening the material. A study of the dependence of the defect structure on exposure in tungsten is also being made. Preliminary results indicate that up until a dose of approximately 10^{18} nvt ($E > 1$ MeV), there exists a linear dependence of single vacancy concentration and depleted zone density on fast neutron exposure. At higher doses the single vacancy concentration begins to saturate, and the saturation value for single vacancies has been determined to be approximately 2×10^{-3} at.%. Simultaneously, the depleted zones grow to larger sizes and the largest of which begin to collapse forming small vacancy type dislocation loops (20 to 30 Å in diam) these preliminary results can be used in a qualitative manner to help explain saturation phenomenon in the yield stress of neutron irradiated materials.

We have also studied Stage III recovery in 99.995% tungsten by directly observing the defect structure before and after Stage III recovery in this material. Before Stage III recovery (350 °C), the interstitial concentration after an exposure of 5×10^{19} nvt ($E > 1$ MeV) was approximately 1×10^{-3} at.% these defects appear as bright spots which are not in ordinary lattice positions. Annealing the material at 400 °C for one hour causes a drop in the concentration of these interstitials from approximately 10^{-3} atom fraction to less than 10^{-5} atom fraction. Simultaneously, the vacancy concentration decreases from 2×10^{-3}

to 1.3×10^{-3} atom fraction. The only possible explanation of a drop in the vacancy concentration from 2×10^{-3} to 1.3×10^{-3} , upon annealing through Stage III, is the migration of interstitials to vacancies since it has previously been shown that extensive vacancy motion occurs in Stage IV recovery. Thus, the simultaneous decrease in the concentration of interstitials and vacancies can only be accounted for by the migration and subsequent annihilation of interstitials at vacancy type defects already present in the irradiated material.

The possibility that the observed interstitial atoms are impurity trapped interstitials can be discounted since the ratio of interstitials to total impurities present is approximately 20% for this material. Thus, one impurity atom must trap more than 20 interstitial atoms to account for 10^{-3} atom fraction of interstitials. In our observations we observe single interstitials, but certainly not large interstitial clusters.

The observations thus indicate that Stage III recovery in neutron irradiated tungsten is an intrinsic recovery stage where the removal of a self interstitial type defect occurs.

Work has also been initiated on alloy systems. Preliminary observations indicate that osmium atoms can be distinguished from tungsten atoms in dilute W-Os 1% alloys.

PROPERTIES OF Fe-N ALLOYS

S. B. McRickard

An ingot high in nitrogen content was produced for tensile and internal friction studies. A pure nitrogen

atmosphere was introduced during a vacuum remelt of Ferrovac iron and charged into the melt during induction agitation. Chemical analyses showed 135 ppm of nitrogen and 9 ppm of carbon present. Grain growth was particularly difficult to achieve in this material and an anneal of 825 °C for five hours was required to produce an average grain diameter of 25 μ . The microstructure contains small nitrides in the matrix.

Preliminary tensile tests have shown:

- Strain aging at 298 °K (carbon strain aging begins above 380 °K at this strain rate of 0.02/min)
- The yield stress is almost athermal down to 180 °K
- A brittle transition at approximately 130 °K.

These tensile properties are shown in Figure 5.4 along with those of Ferrovac iron and an iron containing 300 ppm of carbon.

Additional tensile specimens and internal friction wires will be irradiated. If soluble nitrogen is responsible for strain aging at 298 °K, it would appear from these results that the Ferrovac iron we have used contains insufficient nitrogen to affect dislocation mobility since none of the above effects were observed in testing the Ferrovac iron.

FUEL CLADDING MATERIALS FOR FAST REACTORS

J. G. Y. Chow

The cobalt base alloys have been proposed as an advanced fuel cladding material for use in the 1400 to 1500 °F temperature range. The cobalt

EFFECT OF INTERSTITIALS ON TENSILE PROPERTIES OF IRON

FIGURE 5.4. Low Temperature Tensile Properties of Ferrovac Iron and Iron Containing Larger Amounts of Carbon and Nitrogen Respectively

base alloys are believed to have adequate corrosion resistance to sodium and adequate strength for use at these temperatures. To support this program, a study on the effect of irradiation on the mechanical properties was initiated.

Haynes 25 a Co-base alloy, containing 10 Ni, 20 Cr, 15 W, and 3 Fe, and Multimet alloys, an iron base alloy containing 20 Co, 20 Ni, 21 Cr, 3 Mo, 2.5 W, and 1.0 Nb, are being studied. Prior heat treatment on both alloys were solution treated at 2150 °F. The

specimens were irradiated at 100 °C in a core facility of HFBR, and the exposure is estimated to be 2×10^{20} nvt ($E > 1$ MeV). The flux foils have not been analyzed as yet.

The tensile test results obtained for these alloys in the irradiated and unirradiated conditions are shown in Table 5.1.

After an exposure of 2×10^{20} nvt, the yield strength of Haynes 25 at 25 °C increased from 67,000 to 139,000 psi, but the elongation decreased from 55 to 15%. At 800 °C, the elongation

TABLE 5.1. Tensile Properties of Haynes 25 and Multimet Alloys

Alloy	Exposure ^(a)	Test Temp.	YS	TS	Total Elong.
Haynes 25	0	25 °C	67,000	152,000	55%
Haynes 25	2 x 10 ²⁰	25 °C	139,000	142,000	15%
Haynes 25	0	800 °C	35,900	41,500	10.5%
Haynes 25	2 x 10 ²⁰	800 °C	39,200	41,100	2.3%
Multimet	0	25 °C ^(b)	71,000	121,000	43%
Multimet	2 x 10 ²⁰	25 °C	116,000	138,000	31%
Multimet	0	800 °C	23,600	32,900	55%
Multimet	2 x 10 ²⁰	800 °C	30,000	31,300	6.7%

(All specimens solution treated at 2150 °F. Strain rate of tensile test is 0.002 in. per min. Irradiation temperature-100 °C.)

(a) Estimated exp. - flux foils being analyzed.

(b) Tested at 0.02 in. per min.

for unirradiated Haynes 25 is only 10.5%, while the elongation at 800 °C for the irradiated material is only 2.3%.

At 25 °C, the yield strength of the Multimet alloy was increased from 71,000 to 116,000 psi, and the elongation was decreased from 43 to 31% after irradiation. At 800 °C, the yield strength was increased from 23,600 to 30,000 psi, but the elongation decreased from 55 to 6.7% after irradiation.

These preliminary results indicate that Haynes 25 and Multimet alloys are also embrittled at high temperatures after irradiation in a manner similar to austenitic stainless steels and nickle base alloys. Several modifications of the Haynes 25 alloy and other cobalt base alloys will be studied.

SUMMARY

Tensile test on iron with 135 ppm nitrogen has been made. The iron with

nitrogen strain aged at ambient temperature; whereas, the iron with carbon strain aged at 100 °C during a tensile test. Tensile test and internal friction specimens of the nitrogen-bearing iron are being irradiated for further study.

Haynes 25, a cobalt base alloy, and Multimet alloy, which contains 20% cobalt, have been tested at 25 °C and 80 °C after an exposure of 2 x 10²⁰ nvt (E > 1 Mev). Preliminary results indicate that both of these alloys suffer high temperature embrittlement similar to that found in stainless steels and nickle base alloys.

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A RESEARCH PROGRAM ON THE EFFECT OF SUBSTRUCTURE ON
THE IRRADIATION STABILITY OF ASTM A302 GRADE B STEEL

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STATEMENT OF PROBLEM

The program on the effect of substructure on the irradiation stability of ASTM A302-B steel has two related objectives. The objective of the first phase of the program is to evaluate the effect of substructure on the mechanical properties of A302-B steel in the nonirradiated state. The specific purpose of this part of the program is to show that the properties of this typical reactor pressure vessel steel can be significantly improved by a suitable combination of cold work and heat treatment. This improvement has been demonstrated, and the second phase of the program has been started. The objective of this phase is to study changes due to neutron irradiation in the properties of this steel with and without a well-defined substructure. This phase of the program will include tensile tests, instrumented Charpy tests, and multiaxial stress tests on tubular specimens.

TEST MATERIAL AND TECHNIQUE

The test material is ASTM A302-B steel from heat N-31438, plate 190785. The 10 cm thick section was received in the quenched and tempered condition (water-quenched from 900 °C, 4 hr temper at 650 °C). The plate was trimmed and cut by sawing it into slabs

at the one-quarter and three-quarter thickness levels. These slabs were then cut into specimen blanks. The composition of the steel is within ASTM specifications, the hardness is Rockwell B 92, and the grain size is ASTM 7-8. The largest inclusions observed microscopically were less than 5% of the planned specimen cross section for the tubular specimens. The tensile and Charpy specimens are made to and tested in accordance with applicable ASTM specifications. The tube specimen is essentially a tensile bar with an axial hole, and testing is accomplished by internal pressurization that yields a 2 to 1 stress ratio.

EXPERIMENTAL RESULTS AND DISCUSSION

During the reporting period, the four ambient temperature irradiation capsules were opened at the Naval Research Laboratory (NRL). The Charpy impact specimens were retained at NRL, where they will be tested in April with specimens from the elevated temperature capsule. The tensile and tube specimens and the flux and temperature monitors were shipped to the General Atomic Hot Cell Facility. The tensile and tube specimens were easily removed from the aluminum heat transfer blocks and have been stored in bottles. Several tensile bars from Capsule 1

had small amounts of rust on the threaded end grip region, indicating that the capsule may have leaked slightly. However, no rust was visible in the test section or on the tube specimens.

The temperature melt monitors (124 °C for Capsule 1 and 117 °C for Capsules 2, 3, and 4) were examined under the stereo microscope, and no evidence of melting was seen. Figure 6.1 shows a typical monitor in a quartz ampoule. The number on the ampoule is applied as a decal and then the ampoule is fired at 575 °C. No deterioration of the number or the standard-grade quartz was evident at the highest fluence, $\sim 3 \times 10^{19}$. The iron and nickel-cobalt flux monitors, which were in the form of 5 cm long, 0.38 mm diam wires, were counted in a single channel analyzer with a NaI window. The following absolute fluence values for one monitor in each capsule were determined by hand calculation: $\sim 2.2 \times 10^{18}$, $\sim 9 \times 10^{18}$, $\sim 1.5 \times 10^{19}$, and $\sim 3 \times 10^{19}$ n/cm² (<1 MeV). Values for all monitors will be calculated by computer with the program used in previous General Atomic irradiations.⁽¹⁾ The counts per minute values from each monitor were plotted (Figure 6.2) and demonstrate the symmetry of the flux profile over the length of the capsule. The fluences obtained varied slightly from the desired values of 1×10^{18} , 5×10^{18} , 1×10^{19} , and 5×10^{19} n/cm². The fact that two samples received fluences close to 1×10^{19} suggests that it may not be useful to test all the samples from each capsule, since the differences would be small. On the basis of this preliminary fluence information,

a tentative specimen testing schedule has been set up. In addition, several tests of nonirradiated samples will be made. The testing techniques for the tensile and tube tests have been described elsewhere.⁽¹⁾ Quartz radiant heat lamps will be used as the heat source, since liquid baths are difficult to use at the higher temperatures (290 °C).

An instrumented tup was installed on the out-of-cell Testing Machines, Inc., Charpy impact machine at NRL and several Charpy specimens were broken. The results were good, and no problems with the in-cell machine are anticipated.

The elevated temperature irradiation capsule (290 °C) was assembled, inserted in the LITR on February 18, 1967, and removed on March 29, 1967, with an approximate fluence of 9×10^{18} n/cm². The capsule should be received at NRL during the first week of April, when it will be opened.

Figure 6.3 is a drawing of the elevated temperature capsule showing the configuration necessary to allow heater wires, thermocouple wires, and the helium inlet tube to extend to the lower parts of the capsule. Details concerning capsule construction and the heaters used can be found in Reference (2). The partially assembled capsule, including the Charpy specimen section with several of the thermocouple wires visible in the notches, is shown in Figure 6.4. Figure 6.5 is an end view of the tensile and tube section which shows thermocouple wires in grooves in the aluminum block to allow positioning at the center of the test section of the tensile bars. Thermocouples for the tube specimens were

Neg K53058

FIGURE 6.1. Photograph of Melt Monitor Showing Identifying Number; No Evidence of Melting is Present

FIGURE 6.2. Plot of Counts Per Minute Values from Iron Monitors Versus Position

FIGURE 6.3. Elevated Temperature Capsule

Neg 67148

FIGURE 6.4. Partially Assembled Elevated Temperature Capsule Showing Thermocouple Placement in Charpy Section

FIGURE 6.5. Partially Assembled Elevated Temperature Capsule Showing End of Tensile and Tube Section

placed in grooves in the mandrel inserted in each specimen. The thermocouples were not attached directly to either the tensile bar or tube specimen but were spot-welded to the heat transfer sleeve for the tensile bar and to the internal mandrel for the tube specimens (Figure 6.6). The tensile bars had the same type of split heat transfer sleeves that were used in the ambient temperature capsules, and the specimens had similar sleeves. In addition, the tube specimens had steel split mandrels inserted in the inner diameter to provide additional

gamma heating. As in the ambient temperature capsules, flux monitor wires were contained in plugged holes in the blocks or in the notches in the Charpy specimens (Figure 6.7). The completed capsule before and after the inner skin was welded on is shown in Figure 6.4 and 6.8, respectively. Figure 6.9 shows the completed capsule with the outer skin in place.

Temperature control in the capsule is accomplished in several ways. On the basis of previous experience at NRL, information on the temperature reached with gamma heating alone is

Neg K53060

FIGURE 6.6. Photograph Illustrating Method of Attaching Thermocouples to Split Sleeve on Mandrel

Neg G47396

FIGURE 6.7. Partial Assembly of Ambient Temperature Capsule Block Showing Split Sleeves and Flux Monitor Placement

Neg 67145

*FIGURE 6.8. Capsule with Inner Thin
Stainless Steel Skin*

Neg 67144

*FIGURE 6.9. Capsule with Outer Thin
Stainless Steel Skin*

obtained. Then the clamps shown in Figure 6.4 are positioned to give the flattest temperature profile along the specimen axis. The electrical resistance heater below the specimens is controlled by monitoring the temperature 1 cm from the heater. Finally, the rate of heat loss from the capsule can be controlled by pressurizing the space between the inner and outer skins. The temperature profile on the

capsule has been good, with temperatures ranging from 277 to 293 °C. One exception occurred in the bottom of the capsule, where one Charpy specimen at the outside of the specimen bundle had a temperature of 268 °C.

PLANS FOR FUTURE WORK

When the fluence calculations have been completed, the tensile and tube specimens will be tested at 22, 115,

and 290 °C. Instrumented Charpy tests will also be performed.

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IRRADIATION EFFECTS ON REACTOR STRUCTURAL MATERIALS

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HYDROGEN EMBRITTLEMENT OF STEELS

In the previous quarterly report the purpose of the tests was given. In this report plans and progress for the determination of the effect of hydrogen in the pressure vessel steels HY 80 (Ni-Cr-Mo), A542, and A302B are discussed. Specimens of these steels are to be irradiated in simulated prototypical conditions and tested in specified conditions. Selected for these conditions were as-received heat treatment from the 1/4 in. plate thickness, irradiation temperature of 200 to 325 °C and helium bonded irradiation atmosphere. The specimens will be tested in the as-irradiated and hydrogen charged condition. Base line relationships for the hydrogen charged unirradiated condition and hydrogen concentrations are to be established. A few tests will be conducted to determine if the steels can become more susceptible to hydrogen embrittlement.

Chosen as representative of these steels for the additional tests was A542, which will be subjected to various heat and cold working treatments to vary the microstructure and strain within this steel prior to irradiation, hydrogenation, and testing.⁽¹⁾ This steel was selected for these variations because it has the higher yield strength⁽²⁾ and, therefore, should be the steel most prone to become susceptible to the ad-

verse affects of hydrogen. The program is outlined in Figure 7.1.

Sample plates of A302B, U.S.S. heat No. N-31438,⁽³⁾ and HY 80 (Ni-Cr-Mo), U.S.S. heat No. 74L204,⁽⁴⁾ were acquired from the AEC stockpile at PNL. A sample plate of A542 Class II, Melt/Slab B4274-1, 6 1/4 in. thick, was obtained from Lukens Steel Company. All three steels have been cut-up into blanks for machining.

Portions of the A542 steel as 11/16 in. x 11/16 in. x 5 in. blanks have been heat treated as follows:

- Austenitized at 930 °C for 1 hr, quenched in an agitated brine solution, and tempered at 620 °C to a hardness of R_C 23-24
- Austenitized at 930 °C for 1 hr, quenched in an agitated brine solution, and tempered at 620 °C to R_C 37-38
- Austenitized at 930 °C for 1 hr and furnace cooled.

These heat treatments produced:

- An essentially tempered martensite structure at about the same hardness or strength level as the as-received condition (tempered bainite R_C 22-23)
- Tempered martensite at a higher hardness or strength level
- A mixture of fine pearlite and ferrite.

FIGURE 7.1. Hydrogen Embrittlement of Steels Program

In addition, several of the blanks have been cold worked various amounts by rolling

Blanks of these steels, from the one-quarter thickness are being machined into shapes shown in Figure 7.2. The notched specimen is similar to that of Troiano's⁽⁵⁾ and Rossin's⁽⁶⁾ and will be employed for stress-rupture testing while the plain tensile specimen will be used in conventional tests performed at low strain rates.

Plans at present call for encapsulation of the specimens to be irradiated in stainless steel capsules (Figure 7.3) in a stagnant helium atmosphere. Specimen temperature is to be maintained at about 300 °C by using aluminum spacers to control the helium gap. The exact dimensions of the spacers, due to the flux gradient, will be dependent upon its location within the capsule. Each capsule will contain a single thermocouple affixed to the top specimen.

FIGURE 7.2. Notched and Tensile Specimens for Hydrogen Embrittlement of Steel Tests

Hydrogen absorption by the specimens will be accomplished by cathodic charging. Attempts will be made to adjust the concentration in the specimens to two reproducible levels (high and low) below the level where irreversible embrittlement (cracking and blistering) occurs. Hydrogen analysis will be accomplished using an Exhalograph EA-1 gas analyzer.⁽⁷⁾

PM-2A Vessel Fracture Surface Characterization

An introduction to the work being conducted to characterize the fracture produced under controlled conditions

in the PM-2A pressure vessel was presented in the last quarterly progress report. The work is to be completed by the end of the fiscal year.

After completion of the cutting by torch of the main slab from the PM-2A pressure vessel, a 2 1/2 in. wide slice was cut by power hack saw from the edge carrying the artificial defect and the fracture surface. This slice was immersed overnight in "Methachlor" to remove the corrosion inhibiting oil, "Surface Shield," which had been applied several times to preserve the fracture surface. A rinse in "Methachlor" followed by acetone

FIGURE 7.3. Schematic of Specimen Irradiation in the ETR

completed the cleaning process, after which the fracture surface was photographed extensively in both black and white and in color to give overall views plus stereo views at 3X and 10X on the fracture surface in the region of the defect.

An overall view of the fracture surface and a slightly enlarged picture of the region including the origin of the fracture are reproduced in Figure 7.4. Part A in Figure 7.4. delineates the artificial defect and also the 304 SS lining which had been applied by welding. Projections resembling shear lips were found along the

outer edge of the fracture surface in the regions indicated. Figure 7.4 B shows several features of interest, including "chevron" lines inclined at an angled of 60° in the direction of the origin of the fracture (opposite 64 $\frac{3}{8}$ on the nearby tape measure). The broad sloping wall of the artificial defect produced by the rough-cut electrodes terminates in the narrow "tip" produced by the tungsten foil electrode. The columnar structure of the 304 SS weld deposit occupies about $\frac{1}{3}$ of the metal at the base of the artificial defect. In the A350 steel at the base and adjacent to the notch

FIGURE 7.4. (A) Overall View of Fracture Surface Including Artificial Defect (B) Magnified View (2.4X) of Region Containing Origin of Fracture. (Note: the "chevron" lines sloping toward the origin of the fracture and the jagged tear region next to the artificial defects.)

a region resembling a ductile jagged tear region is present. This region is about $3/8$ the width of the A350 metal, and is presumably the stress corrosion area. Corrosion has started in this region at the tops of the jagged peaks and throughout the region.

The hydraulic fluid that flooded the area after the failure and the corrosion inhibiting oil evidently did not wash away all the acidic solution used in the stress corrosion treatment.

Optical and electron microscopy is in progress to determine the depth of penetration by stress corrosion.

Electron microscope replicas have been taken in the region of the fracture origin (which was close to the middle of the artificial defect) and on each side of the defect at regions:

- Midway between fracture origin and end of defect
- At end of artificial defect
- At region midway between end of artificial defect and end of slab
- On sloping surface of "shear lip."

The replicas will be examined with the electron microscope to determine the percentage of brittle fracture.

When it has been ascertained that satisfactory replicas have been obtained from each of these regions, 1/2 in. thick slices will be taken in the same regions for optical metallographic examination.

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DEVELOPMENT OF TEST PROCEDURES, FRACTURE TOUGHNESS OF
REACTOR PRESSURE VESSEL STEELS

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INTRODUCTION

Obtaining plane strain fracture toughness data on irradiated reactor vessel steels requires the development of test procedures that use specimens much smaller than those now considered necessary for valid testing. The obstacle in using small specimens is the fact that the critical specimen dimensions are proportional to the square of the ratio of toughness to yield strength of the material being tested, and this ratio is very high for the metals used in reactor vessels. Additionally, to duplicate service conditions, the temperatures of interest for irradiation studies are ambient and above, preventing the use of very low temperature testing. The most promising method for decreasing size requirements would be to take advantage of the yield strength elevation produced by the use of high loading rates. Consequently, all of the experimental effort during this quarter was concerned with evaluating toughness as a function of loading rate for two different test temperatures, i.e. ambient (75 °F) and 0 °F. Future work will extend this testing range to -75 °F.

IMPORTANCE OF LOADING RATE

For any material, the measured values of K_{IC} are expected to be a

function of the rate at which the stress is increased at the crack tip; the higher this rate the lower the value of K_{IC} over a reasonable range of loading rates. The minimum value in K_{IC} would probably occur when the crack is running freely at a rate exceeding the separation rate of the test machine heads. This lower limit in K_{IC} should be equal to the plane strain arrest toughness value for a DCB specimen. To simulate conditions at the crack tip characteristic of a moving crack, the loading rate for a specimen with a stationary crack might be continuously increased until the initiation toughness is found to have some minimum value.

On the basis of these considerations, one might expect that as the loading rate for a specimen with a stationary crack were continuously increased, the yield strength at the crack tip would be increased while the fracture toughness decreased. Hence, high rate tests should lead to a maximum measuring capacity for any specimen at a particular test temperature.

EXPERIMENTAL PROCEDURE

Fracture toughness tests made during the present report period used linear compliance change DCB specimens contoured to $m = 3$. The specimens were 1 and 2 in. thick with side-groove

ratios varying from $b_n/b = 0.50$ to 0.75 .⁽¹⁾ The cracking plane was parallel to the plate surface and located on the midplane of the 4 in. thick A302-B steel plate. A fatigue precrack, extending into the linear compliance change region, was produced at room temperature in all of the specimens.

To obtain load-to-fracture times ranging down to 10 msec a closed-loop servo-hydraulic testing machine was used. Each specimen was loaded with

a single triangular displacement pulse which had an amplitude much greater than that required for fracture.

Loading rates were varied by adjusting the pulse width. Load displacement (P- δ) and load time (P-t) were obtained on an oscilloscope. Records from tests at 0 °F on two different specimens loaded at two different loading rates are shown in Figure 8.1. Load deflection traces (on the left side of each photograph) show linear behavior up to the initiation of rapid

FIGURE 8.1. Load-and Deflection-Time Traces. (The P- δ curves start from the lower left corner of the photographs: the P-t curves from the lower right.)

fracture, an abrupt drop in load to the value for crack arrest, then re-loading until rapid fracture is again initiated, etc. The record for the lower strain rate test (top figure) gives two initiation values and one arrest value; at the higher strain rate test (bottom figure) there were three initiation and two arrest values. These initiations and arrests were obtained during one continuous loading of the specimen.

The graphs on the right side of the two oscilloscope patterns in Figure 8.1 are load-time traces. To improve the sensitivity of load measurements, zero is suppressed in the P-t traces and each major division is 2000 lb rather than 4000 lb as used for P- δ . It is necessary to plot both of these functions simultaneously for each test to obtain the data being sought. The P- δ trace is used to be certain that the specimen fractures on the elastic portion of the curve; the P-t trace is used both for measuring initiation and arrest loads as well as for obtaining the data on loading rate.

To obtain more rapid loading times than was possible with the hydraulic testing machine, a drop-weight test was developed. The top clevis of the loading train—in series with a load cell—was fastened to the top crosshead of a loading frame. A 2 in. diam bar approximately 3 ft long was fastened to the bottom clevis. To load the specimen, a 100 lb weight was dropped the length of the 3 ft bar and allowed to strike a flange attached to the bottom of the bar. To avoid erratic loading behavior, it was necessary to make the loading times correspond with

the natural frequency of the loading train by cushioning the impact of the dropped weight. A load-time trace obtained with this testing arrangement is shown in Figure 8.2a. The record was obtained on a dummy specimen, of approximately the same compliance as an actual test specimen, but it did not crack under the impact loading. The load monitored by a load cell is plotted up from the bottom left while the output of the clip gage measuring specimen deflection is plotted down from the top left. The fact that the load-time and displacement-time curves were in phase and that spurious peaks were absent is evidence that the load cell provides a meaningful measurement at the loading rate used. The load-time curve obtained on a room temperature test of A302-B steel is shown in Figure 8.2b. A load value of 18,000 lb at the onset of rapid fracture can be clearly read from the record; the loading time is 1 msec. On this test no crack arrest was observed.

EXPERIMENTAL RESULTS

Room temperature measurements of K_{IC} for initiation and arrest are plotted in Figure 8.3. Loading times were obtained from the slopes of the load-time traces as the fracture instability load was approached, and the reciprocal of the loading time is used as a measure of the loading rate. Only K_{IC} values from the re-initiation from fatigue cracks are plotted; those obtained from the re-initiation of arrested rapid fractures are not shown. The data in Figure 8.3 were obtained on 1 and 2 in. thick specimens, with b_n/b varying from = 0.50 to 0.75.

FIGURE 8.2. Load- and Deflection-Time Traces Obtained in Drop-Weight Tests. (The P-t traces start from the lower left corner of the photographs; the δ -t from the upper left.)

Neither the 2 in. nor 1 in. thick specimens fulfill the Brown-Srawley criterion for specimen thicknesses, B , i.e., $B = 2.5 \left(\frac{K_{IC}}{\sigma_0} \right)^2$ if one uses the yield value obtained at a low loading rate (i.e., 60 ksi). The rates used in these tests, however, elevate the yield strength subsequently above this

level so that the two such specimens are probably within this criterion.

From examination of the initiation data for the 2 in. thick specimens, it is seen that K_{IC} over the range of loading rates studied is essentially independent of rate, and the value of toughness is approximately $80 \text{ ksi}\sqrt{\text{in.}}$.

FIGURE 8.3. Room Temperature Initiation and Arrest Toughness of A302-B Steel

The 1 in. thick specimens showed a high level of rate sensitivities, with the measured toughness decreasing to a value of approximately 85 ksi√in. at reciprocal rates of the order of 100 to 1000 sec⁻¹. The arrest toughness for both the thick and thin specimens appears to be identical, and essentially independent of loading rate. The scatter of arrest toughness is small and has an average value of approximately 77 ksi√in.

Data obtained to date on tests at 0 °F are shown in Figure 8.4. All of the specimens were 1 in. thick with $b_n/b = 0.75$ and, again, the open circles represent initiation values of K_{Ic} and the closed circles arrest toughness. Most of the data show initiation toughness to range between 60

and 70 ksi√in. and arrest toughness between 50 and 60 ksi in., both independent of loading rate. Two neighboring points obtained at reciprocal loading rates of approximately 3.0 and 6.0 sec⁻¹, respectively, show significantly higher initiation toughness values and somewhat elevated arrest toughnesses. Further tests will be conducted in this region of loading rates to determine whether these values are, in fact, the result of a strain rate effect or merely indicative of material scatter.

DISCUSSION AND CONCLUSIONS

Since only a limited number of tests and only a modest range of specimen thicknesses have been tested to date, conclusions drawn from these data must

FIGURE 8.4. Initiation and Arrest Toughness of A302-B Steel at 0 °F

be considered tentative. Nevertheless, if one were to assume that the 2-in. thick specimens yielded valid toughness values at room temperature and the 1 in. thick specimens valid plane strain results at 0 °F, it must be concluded that loading rate exerts only a modest influence on toughness over the range studied.

However, rapid loading rates do appear to promote plane strain fracturing, at least, in this mild steel.

Again, it must be tentatively concluded that the measuring capacity of fracture toughness specimens could be increased by the use of very high loading rates.

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RESEARCH PROGRESS

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COMPARISON OF SELECTED STRUCTURAL
STEELS AFTER IRRADIATION AT 550 °F
AND AT 650 °F TO A HIGH NEUTRON FLUENCE

Seven steels, having compositions as listed in Table 9.1, have been irradiated at a controlled temperature of 550 °F to a neutron fluence $\approx 4.8 \times 10^{19}$ n/cm² > 1 MeV for comparisons of Charpy-V notch ductility behavior. The steels represent a broad range of yield strength from 44.7 ksi (A212-B) to 184.4 ksi (12 Ni-5 Cr-3 Mo maraging), as noted from Table 9.2. In a second experiment, the same steels with exception of the A212-B steel were irradiated at 650 °F to a neutron fluence of 4.1×10^{19} n/cm² > 1 MeV. An A542, Class II, steel having a yield strength of 102.9 ksi was also included in this particular experiment. Fluence levels of both experiments are considered maximum if not in excess of lifetime exposures presently estimated for most carbon and low alloy steel reactor pressure vessels in operation or under construction. Results from both materials irradiations are summarized in Table 9.3.

Reviewing the 550 °F irradiation test data, the results suggest four general categories of radiation embrittlement sensitivity as follows: Category 1: (high sensitivity), 5 Ni-Cr-Mo-V; Category 2: (conventional), A212-B, A302-B; Category 3: (moderately

low), 3 1/2 Ni-Cr-Mo, 162.6 ksi YS maraging steel, A517-E; and Category 4: (low), 184.4 ksi YS maraging steel. From these data, the general trend reported earlier of decreasing embrittlement sensitivity with increasing yield strength is again seen.^(1,2) However, the 5 Ni-Cr-Mo-V steel (130/140 ksi YS) is a significant exception to this trend as, to a lesser extent, is the 162.2 ksi YS marage steel. Thus, each new high strength steel composition (or heat) remains suspect for possible anomalous behavior until metallurgical explanations for these behavior differences are established.

On comparing the 550 °F exposure data listed in Table 9.3 with results from earlier experiments receiving a lower fluence,^(3,4) significant differences with respect to tendencies toward radiation damage saturation are observed among the seven steels. For example, the A212-B, A302-B, and A517-E steels would appear to be reaching their maximum embrittlement states for the 550 °F exposure condition over the fluence range from 3 to 5×10^{19} n/cm² > 1 MeV; whereas, the 3 1/2 Ni-Cr-Mo and 5 Ni-Cr-Mo-V steels do not. In the case of the A302 B steel, data from the Yankee reactor pressure vessel surveillance program, wherein the same ASTM reference steel heat was involved, support this analysis

TABLE 9.1. Chemical Composition and Heat Treatment of Selected Steels Irradiated at 550 and 650 °F

Steel	Plate Thickness, in.	Chemical Analysis, %												Heat Treatments	
		C	Mn	Si	P	S	Ni	Cr	Mo	Cu	Ti	N	Al		V
A212-B	4	0.26	0.76	0.24	0.011	0.031	0.22	0.20	0.02	-	-	-	-	-	(1)
A302-B (ASTM Reference)	6	0.20	1.31	0.25	0.012	0.023	0.20	0.17	0.47	-	-	-	-	-	(2)
A517-E (SSS-100)	6	0.18	0.51	0.28	0.017	0.020	0.08	1.72	0.50	0.26	0.085	-	0.036	-	(3)
3 1/2 Ni-Cr-Mo	8	0.17	0.38	0.29	0.013	0.023	3.65	1.88	0.51	-	-	0.010	0.02	-	(4)
5 Ni-Cr-Mo-V	4	0.12	0.75	0.28	0.003	0.005	4.95	0.57	0.46	-	-	-	-	0.065	(5)
12 Ni-5 Cr-3 Mo (162.6 ksi YS)	1	0.005	0.04	0.05	0.005	0.007	11.8	5.16	3.30	-	0.24	-	0.13	-	(6)
12 Ni-5 Cr-3 Mo (184.4 ksi YS)	1	0.007	0.04	0.08	0.005	0.007	11.8	5.08	3.30	-	0.24	-	0.14	-	(7)
A542 (Class II)	6 1/4	0.16	0.38	0.35	0.011	0.004	0.25	2.40	1.18	-	-	-	-	0.02	(8)

Heat Treatments:

- (1) Austenitized at 1650 °F for 2 hr; water quenched; tempered at 1175 °F for 4 hr; furnace cooled to below 600 °F.
- (2) Austenitized at 1650 °F for 2 hr; water quenched; tempered at 1200 °F for 6 hr; furnace cooled to below 600 °F.
- (3) Austenitized at 1650 °F; water quenched; tempered at 1225 °F for 1 1/2 hr; retempered at 1240 °F for 2 hr.
- (4) Austenitized at 1650 °F with 8 hr heating and 2 hr hold; water quenched for 17 min; reaustenitized at 1500 °F with 8 hr heating and 2 hr hold; water quenched for 17 min; tempered at 1185 °F with 8 hr heating and 2 hr hold; water quenched cold.
- (5) Standard mill quenched and temper treatment for 130 ksi minimum yield strength.
- (6) Solution annealed at 1500 °F for 1 hr; water quenched; aged at 900 °F for 2 hr; water quenched.
- (7) Solution annealed at 1500 °F for 1 hr; water quenched; aged at 900 °F for 20 hr; water quenched.
- (8) Austenitized 6 hr at 1725 °F; water dip quenched; tempered 6 1/4 hr at 1100 °F, air cooled; tempered 12 hr at 1150 °F, air cooled.

TABLE 9.2. Tensile Properties of Selected Steels at Quarter Plate Thickness Prior to Neutron Exposure

Steel	Plate Thickness in.	Yield Strength, 0.2% offset, ksi	Ultimate Tensile Strength, ksi	Elongation, 1 in. gage, length, %	Reduction of Area, %
A212-B	4	44.7	79.1	33.6	64.6
A302-B	6	70.7	92.3	27.2	64.8
A517-E (SSS-100)	6	97.5	110.9	22.3	68.9
3 1/2 Ni-Cr-Mo	8	95.0	122.0	24.7	68.7
5 Ni-Cr-Mo-V	4	145.0	154.8	16.8	54.2
12 Ni-5 Cr-3 Mo (162.6 ksi YS)	1	162.6	170.6	15.8 ^(a)	64.1
12 Ni-5 Cr-3 Mo (184.4 ksi YS)	1	184.4	190.0	13.5 ^(a)	55.7
A542 ^(b) (Class II)	6 1/4	102.9	120.5	22.0 ^(a)	71.0

(a) Elongation in 2 in. gage length

(b) Courtesy of Lukens Steel Company

by suggesting that an increase in neutron fluence at ≈ 520 °F from 5×10^{19} to 7×10^{19} n/cm² would increase the transition temperature shift of the heat from 225 to only 260 degrees. In turn, the recently obtained data from the test reactor irradiation serve to reinforce data obtained from the Yankee pressure vessel surveillance program for the 5×10^{19} n/cm² exposure case (ΔT 225 degrees at 520 °F versus 205 degrees at 550 °F).

The most important assessment of the experiment results for the 550 °F exposure of the seven steels, however, is that of the absolute transition temperature and full shear energy absorption level following this

"maximum" service condition. From Table 9.3, it is seen that the post-exposure transition temperatures of the 3 1/2 Ni-Cr-Mo, the A517-E, and the maraging steels are all less than room temperature while that of the 5 Ni-Cr-Mo-V, the A212-B, and the A302-B steels are in the range of 200 to 250 °F. Applying the nil-ductility transition (NDT) temperature plus 60 °F criterion for vessel pressurization, the benefits of the first steel group are readily seen from the combined standpoints of ease in reactor startup and reactor vessel safety. With regard to full shear energy absorption level after irradiation, the data are less clear when the possibilities for low energy absorption shear rupture versus

TABLE 9.3. Charpy-V Notch Ductility Properties of Selected Steels Before and After Simultaneous Irradiation at 550 °F and at 650 °F

Steel	Charpy-V 30 ft-lb Transition Temperature, °F					Approximate Full Shear Energy Absorption, ft-lb		
	Initial	Irradiated				Initial	Irradiated	
		550 °F, 4.8×10^{19} n/cm ² >1 MeV ^(a)	ΔT	650 °F, 4.1×10^{19} n/cm ² >1 MeV	ΔT		550 °F, 4.8×10^{19} n/cm ² >1 MeV	650 °F, 4.1×10^{19} n/cm ² >1 MeV ^(a)
A212-B	5	230	225	-	-	81	50	-
A302-B	15	205	190	105	90	81	66	75
A517-E (SSS-100)	-45	65	110	40	95	114	84	88(95%)
3 1/2 Ni-Cr-Mo	-130	20	150	-80	50	74	52	≈65
5 Ni-Cr-Mo-V	-90	240	330	+25	115	57	36	46
12 Ni-5 Cr-3 Mo (162.6 YS)	-195	-50	145	-50	145	84 ^(b)	55 ^(b)	55 ^(b)
12 Ni-5 Cr-3 Mo (184.4 YS)	-175	-105	70	-105	70	61 ^(b)	61 ^(b)	61 ^(b)
A542 (Class II)	+10	-	-	140	130	60 ^(b)	-	60 ^(b)

(a) $\bar{\sigma}$ 68 mb, Mn⁵⁴ Fission

(b) Transverse specimen orientation

properties, both parallel and transverse to the plate primary rolling direction ("strong" and "weak" plate orientations), are considered. Earlier experimental results⁽⁴⁾ on A212-B and A302-B steels irradiated at <250 °F suggest that the difference between the full shear energy absorption levels given by parallel and transverse specimen orientations is maintained after irradiation. On this basis, ≈30-35 ft-lb and ≈35-40 ft-lb respectively would be projected as maximums for the "weak" orientation of the A212-B and A302-B steels for the described 550 °F exposure condition. Similarly, full shear energy absorption values for the A517-E and 5 Ni-Cr-Mo-V steels in their "weak" orientations would be projected at ≈25-30 ft-lb after irradiation. A high retention of notch toughness is noted, however, for the two maraging steel heats as the values for full shear energy absorption (Table 9.3) represent the transverse orientation. Assessment of the full implication of a significant loss in full shear energy absorption with neutron exposure will be one objective of drop-weight tear test specimen studies planned for the near future.

In contrast to the 550 °F service condition, experimental data generated by the 650 °F irradiation experiment suggest that embrittlement developed at the higher temperature by a neutron fluence of 4.1×10^{19} n/cm² would not be of great engineering concern. As shown in Table 9.3, postexposure transition temperatures were below 200 °F in all cases, and furthermore, were below room temperature for five of the seven steel compositions evaluated. Notably,

however, the A542 composition revealed an embrittlement sensitivity equal to that of the 5 Ni-Cr-Mo-V steel. Aside from its slightly higher strength, the A542 steel would not, therefore, appear to hold an advantage over A302-B steel either in terms of embrittlement sensitivity or from the standpoint of absolute (as-irradiated) transition temperature (+140 versus 105). However, the 162.6 ksi YS maraging steel, while demonstrating the greatest change in notch ductility properties among the seven steels, retains a superior overall performance because its initial transition temperature was very low (-195 °F). The equivalence in transition temperature characteristics following irradiation at either 550 or 650 °F (in the case of the maraging steels) is also an interesting observation from this experimental series.

The relatively good performance of the 3 1/2 Ni-Cr-Mo steel following exposure at 650 °F is significant because the use of the Ni-Cr-Mo steels at temperatures higher than 650 °F may be limited by temper embrittlement susceptibility (notably absent in this case). An experiment to determine the possible alteration of temper embrittlement conditions (i.e., "C-curve" characteristics) of this class of steels by irradiation is now in progress. The possibilities for other thermal and strain aging phenomenon must also be recognized and considered in the long-term irradiation (years) of the above materials at elevated service temperatures; thus, the continuing need for reactor component materials surveillance programs is re-emphasized.

THROUGH-THICKNESS CHARPY-V NOTCH
DUCTILITY PERFORMANCE OF A 10 1/2 IN.
PLATE OF IRRADIATED A302-B STEEL

As pressurized water nuclear reactors are designed for higher power output, stronger and, thus, heavier section pressure vessel walls are required. Variation in mechanical properties through-the-thickness of heavy section steel plates are, therefore, of concern since previous research⁽⁵⁾ has revealed significant differences not only in initial Charpy-V 30 ft-lb transition temperatures for surface versus 1/4T or 1/2T thickness locations, but also in the radiation embrittlement sensitivities of these locations in an ~8 in. thick A302-B plate.

Through-thickness notch ductility characteristics of a 10 1/2 in. thick A302-B steel plate supplied by the Westinghouse Atomic Power Division through the courtesy of the City of Los Angeles has been investigated in an effort to gain further experience regarding the irradiation response of heavy section steel plates. The chemical composition and heat treatment of the test plate are given in Table 9.4. Charpy V-notch specimens

were taken from layers representing the top and bottom plate surfaces, the quarter plate thickness (1/4T), and the half plate thickness (1/2T) location.

Referring to Figure 9.1, data points representing the top and bottom plate surface material are consistent and show low scatter about an average curve for the unirradiated condition. Similarly, data points are also closely grouped from specimens taken in a double layer about the 1/2T location. As expected, the center or 1/2T location exhibited an initial transition temperature higher than either the 1/4T or surface locations. Specimens taken from the 1/4T location exhibited considerably more data scatter, the width of the data band being described approximately by the surface and 1/2T curves.

Charpy-V specimens of all three thickness regions were irradiated at <250 °F to a fluence of 1.7×10^{19} n/cm² > 1 MeV in a core lattice position of the LITR. Data curves referencing individual Charpy-V 30 ft-lb transition temperature increases are reproduced in Figure 9.1. The notch ductility data for both

TABLE 9.4. *Chemical Composition and Heat Treatment of 10 1/2 in. A302-B Plate*

Source	A. Chemistry, wt%										
	C	Mn	P	S	Si	Ni	Cr	Mo	V	Cu	Al
NRL	0.25	1.37	0.006	0.011	0.21	0.27	0.10	0.62	≤0.01	0.15	0.02
WAPD	0.20	1.37	0.009	0.017	0.19	-	-	0.48	-	-	-

B. Heat Treatment

Austenitized: 1550 - 1600 °F for 4 hr, water dip quenched

Tempered: 1225 ± 25 °F for 4 hr, furnace cooled

Stress Relieved: 1150 ± 25 °F for 30 hr, furnace cooled to 600 °F

FIGURE 9.1. Notch Ductility Characteristics of Surface, 1/4 and 1/2T Locations in a 10 1/2 in. Thick Plate of A302-B Steel Before and After Irradiation at $<240^{\circ}\text{F}$ to a Fluence of $1.7 \times 10^{19} \text{ n/cm}^2 > 1 \text{ MeV}$

material conditions are summarized in Table 9.5. The results of this experiment reveal a consistent transition temperature increase (ΔT) for each location through the thickness of the 10 1/2 in. thick plate. Thus, a very uniform degree of through-thickness radiation sensitivity is suggested for this particular test plate, converse to the response of the first A302-B plate assessed in this manner. In addition, a somewhat lower radiation embrittlement sensitivity is depicted for the second plate in reference to the listed irradiation temperature and fluence conditions. Relatively high upper shelf energy levels both in the irradiated and unirradiated conditions also suggest superior performance.

Generalized conclusions concerning radiation-induced changes in through-thickness properties of heavy section plates of A302-B steel cannot be made on the basis of this one experiment, nor on the results of the initial experiment, as observed plate responses were inconsistent with one another. Nevertheless, the two examples do show that the region of center plate thickness, having a higher transition temperature than either the 1/4T or surface locations, should be evaluated at least in a cursory manner in future irradiation test programs. Finally, the somewhat smaller radiation-induced transition temperature increase recorded for this more recently fabricated plate hopefully is indicative

TABLE 9.5. Charpy-V Notch Ductility Properties
of 10 1/2 in. A302-B Plate Before and After Irradiation
at <250 °F to 1.7×10^{19} n/cm² > 1 MeV^(a)

Plate Thickness Location	30 ft-lb Transition Temperature, °F			Full Shear Energy Absorption, ft-lb	
	Initial	Irradiated	ΔT	Initial Average	Radiation-Induced Loss
Surface	-20	220	240	125	≈10
1/4T	5	250	245	120	<10
1/2T	35	275	240	94	<10

(a) $\sigma = 68$ mb, ⁵⁴Mn Fission

of a trend toward better overall performance of pressure vessel steel plate now being fabricated, despite increasing thicknesses and commensurate difficulties in obtaining the uniform properties in depth.

INVESTIGATION OF THE EFFECTS OF OXYGEN
AND NITROGEN CONTENTS ON THE RADIATION
EMBRITTEMENT SENSITIVITY
OF 7 1/2 Ni-Cr-Mo STEEL AT 250 °F

A 300 lb air induction heat of 7 1/2 Ni-Cr-Mo steel was prepared and split three ways to provide a series of oxygen and nitrogen contents as given by Table 9.6. Ingots were broken down to 1/2 in. plates which were subsequently heat treated according to the quenching and tempering schedule listed in the table to attain matching yield strengths in the range of 140 to 150 ksi. Charpy-V specimens representing the three plate materials together with reference specimens from a 20 ton electric furnace VCD heat of the same nominal composition and approximate strength level were irradiated simultaneously at <250 °F to a neutron

fluence of 1.7×10^{19} n/cm² > 1 MeV. Experimental results are shown in Table 9.7.

Referring first to preirradiation properties, good correspondence in initial Charpy-V 30 ft-lb transition temperatures was found among the four plate materials. As expected, the low oxygen-low nitrogen-containing plate developed the highest full shear energy absorption level. Following irradiation, individual transition temperature increases were also found to be in good agreement. Concurrently, the radiation-induced loss in full shear energy absorption was the same (20 to 26 ft-lb) in all cases. By way of comparison, duplicate irradiation conditions applied to ASTM A302-B reference steel produce a transition temperature increase of 300 degrees.

The experimental results, by depicting equivalent radiation embrittlement sensitivity among plates from the laboratory split heat and the reference commercial heat, suggest that for the 7 1/2 Ni-Cr-Mo composition neither oxygen content in the range

TABLE 9.6. Chemical Composition and Heat Treatment of 1/2 in. Laboratory Test Plates and 1 in. Commercial Reference Plate of 7 1/2 Ni-Cr-Mo Steel

U. S. Steel Supplied Heat	Plate	Chemical Analysis, wt%					
		C	Mn	P	S	Si	Ni
Laboratory Melt (300-lb air induction)	A	0.10	0.27	0.009	0.008	0.22	7.49
	B	0.10	0.26	0.009	0.008	0.22	7.48
	C	0.095	0.23	0.010	0.009	0.18	7.47
Commercial Melt (20 ton Electric Furnace-VCD)		0.12	0.27	0.008	0.008	0.21	7.38

U. S. Steel Supplied Heat	Plate	Chemical Analysis, wt%					
		Cr	Mo	Al ^(a)	Al ^(b)	N	O, ppm
Laboratory Melt (300-lb air induction)	A	0.80	0.98	0.024	0.028	0.002	37
	B	0.80	0.97	0.031	0.038	0.001	65
	C	0.79	0.98	0.016	0.027	0.006	90
Commercial Melt (20 ton Electric Furnace-VCD)	-	0.81	0.97	0.034	(c)	0.006	20

(a) Acid soluble

(b) Total

(c) Not Available

Heat Treatment:

Laboratory Plates - Austenitized at 1500 °F for 1 hr, water quenched;
tempered at 1125 °F for 1 hr, water quenched.

Commercial Plate - Austenitized at 1550 °F for 1 hr, water quenched;
tempered at 1025 °F for 1 hr, water quenched.

TABLE 9.7. Charpy-V Notch Ductility Properties and Rockwell Hardness of 1/2 in. Laboratory Test Plates and 1 in. Commercial Reference Plate of 7 1/2 Ni-Cr-Mo Steel Before and After Irradiation at $<250^{\circ}\text{F}$ to $1.7 \times 10^{19} \text{ n/cm}^2 > 1 \text{ MeV(a)}$

U. S. Steel Supplied Heat	Plate	Nitrogen, %	Oxygen, ppm	Charpy-V 30 ft-lb Trans. Temp., $^{\circ}\text{F}$		
				Initial	Irradiated	ΔT
Laboratory Melt (300-lb air induction)	A	0.002	0.37	-265	-70	195
	B	0.001	0.65	-245	-80	165
	C	0.006	0.90	-265	-70	195
Commercial Melt (20 ton Electric Furnance-VCD)	-	0.006	0.20	-215	-65	150

U. S. Steel Supplied Heat	Plate	Full Shear Energy Absorption, avg. ft-lbs			Rockwell Hardness (R_C)	
		Initial	Irradiated	$\Delta \text{ft-lbs}$	Initial	Irradiated
Laboratory Melt (300-lb air induction)	A					
	B	80	57	23	33.0	37.0
	C	68	42	26	34.0	37.0
Commercial Melt (20 ton Electric Furnace-VCD)		58	38	20	32.5	35.5
	-	62	37	25	38.0	41.0

(a) $\bar{\sigma} \ 68 \text{ mb}$, ^{54}Mn , Fission

of 37 to 90 ppm nor nitrogen content in the range of 0.002 to 0.006% are of major significance with respect to irradiation response at $<250^{\circ}\text{F}$. Furthermore, the choice of melting practice (air induction versus electric furnace VCD) would not appear to be critical to the subsequent irradiation performance of this particular steel in the high strength, quenched and tempered condition. Continuing experimental evaluations of this particular steel series, utilizing 550°F irradiation temperature, will determine

if oxygen and nitrogen contents as well as melting practices are or are not of major importance in irradiation response at realistic Pressurized and Boiling Water Reactor vessel service temperatures.

NOTCH DUCTILITY BEHAVIOR OF IRRADIATED 12 Ni-5 Cr-3 Mo MARAGING STEEL WELDMENTS

Initial evaluations of the effect of low temperature ($<250^{\circ}\text{F}$) irradiation upon the notch ductility properties of 12 Ni-5 Cr-3 Mo maraging steel

plate have shown only a small increase in Charpy-V transition temperatures and commensurately small losses in full shear energy absorption values for a neutron fluence of $7 \times 10^{18} \text{ n/cm}^2 > 1 \text{ MeV}$.⁽⁶⁾ Since the conventional (pre-service) properties of the high yield strength (160 to 180 ksi) steels being considered for use as deep submergence vehicle hull materials appear so promising, two 1 in. thick weldments of 12 Ni-5 Cr-3 Mo plate have been prepared by the TIG process at NRL for further evaluation of this type steel. The nominal chemical composition of the filler wire supplied by the International Nickel Company for the prepara-

tion of these welds is given in Table 9.8. The post-weld aging heat treatment, 12 hr at 850 °F, developed 179 ksi YS (0.2% offset) in both weld deposits. The notch ductility behavior of the weld metals before and after irradiation are shown in Figures 9.2 and 9.3. A summary of conventional properties measurements for the pre-irradiation condition is given in Table 9.9.

A comparison of test results for both weld metal compositions reveals that the Charpy-V specimens did not exhibit a difference in transition temperature after irradiation. More important, the postirradiation test

TABLE 9.8. Chemical Composition of Weld Wire for the Fabrication of 12 Ni-5 Cr-3 Mo Maraging Steel Weldments

NRL Code	Nominal Chemical Composition, wt%					
	Ni	Cr	Co	Mo	Al	Ti
M5W	12	5	-	3	0.3	0.3
M6W	17	-	2	3	0.3	0.3

TABLE 9.9. Charpy-V Notch Ductility and Tension Test Properties of 12 Ni-5 Cr-3 Mo Maraging Steel Weldments as Fabricated and Aged at 850 °F

NRL Code	Charpy-V 30 ft-lb Transition Temp., °F	Full Shear Energy Absorption, ft-lb		Hardness Rockwell-C
M5W	-70	47		43
M6W	-175	47		42
NRL Code	Yield Strength, 0.2% offset, ksi	Tensile Strength, ksi	Elongation, 1 in. gage length, %	Reduction of Area, %
M5W	179.0	188.0	16.0	62.5
M6W	178.8	187.5	16.0	64.7

FIGURE 9.2. Charpy-V Notch Ductility Properties of 1 in. Thick 12 Ni-5 Cr-3 Mo Weld Metal Before and After Irradiation at <250 °F to a Fluence of $\approx 6 \times 10^{18}$ n/cm² > 1 MeV.

FIGURE 9.3. Charpy-V Notch Ductility Properties of 1 in. Thick 17 Ni-2 Co-3 Mo Weld Metal Before and After Irradiation at <250 °F to a Fluence of $\approx 6 \times 10^{18}$ n/cm² > 1 MeV.

data did not reveal any loss in full shear energy absorption wherein any reduction in the 47 ft-lb upper shelf energy would be more critical to the application than a moderate increase in transition temperature.

The very satisfactory properties obtained for 550 °F and 650 °F irradiation conditions of 160 to 180 ksi YS 12 Ni-5 Cr-3 Mo maraging steel plates, presented in a preceding section coupled with the above exploratory assessments of companion weld wire compositions, provide an incentive to extend the weld metal irradiation studies to include elevated temperature exposures. With the addition of tensile properties data for both base plate and weld metal for some of these projected irradiation conditions, the full potential of the maraging steels for broad reactor structural component use may be recognized.

RESPONSE OF A350-LF1 (MODIFIED) STEEL TO POSTIRRADIATION ANNEALING AT TEMPERATURES IN THE RANGE OF 550 TO 590 °F

Preliminary investigations of the notch ductility behavior of A350-LF1 (Mod.) steel forming the pressure vessel of the Army SM-1A reactor have been performed to determine its potential for embrittlement relief (Table 9.10).⁽³⁾ These investigations and the present research have been accomplished at the request of the Army in support of the planned, in-service annealing of the SM-1A reactor. Engineering studies of the reactor system have indicated to the Army that the SM-1A could attain a uniform vessel temperature of 585 °F by use of nuclear heat alone. Thus, NRL began studies to determine the effect of this annealing temperature upon A350-LF1 (Mod.) steel after low temperature (<240 °F) irradiation for

TABLE 9.10. Chemical Composition and Heat Treatment of A350-LF1 (Modified) Steel Ring Forging

<u>A. Chemistry, wt%</u>										
NRL	<u>C</u>	<u>Mn</u>	<u>Si</u>	<u>P</u>	<u>S</u>	<u>Ni</u>	<u>Cr</u>	<u>Mo</u>	<u>H</u>	<u>O</u>
Analysis	0.13	0.76	0.25	0.021	0.036	1.64	0.16	0.06	0.1 ^(a)	12 ^(a)

B. Heat Treatment History

Forged in the temperature range of 1700 °F minimum to 2250 °F maximum.
Postforging treatment: normalized at 1600 °F, water quenched, tempered at 1250 °F, stress relieved at 1150 °F.

(a) ppm

reference purposes as well as after 430 °F irradiations to depict realistic conditions.

As shown in Figure 9.4., the transition temperature increase resulting from neutron irradiation of 1.4×10^{19} n/cm² > 1 MeV at <240 °F was found to be 90% reversible by annealing at 585 °F. With these encouraging results, another irradiation experiment was performed involving six groups of A350-LF1 (Mod.) Charpy-V specimens exposed at a temperature more representative of SM-1A vessel service conditions. In this recent study, the individual groups of irradiated specimens were annealed at temperature intervals in the range of 550 to 590 °F to determine the sensitivity of the steel to an annealing temperature other

than the nominal 585 °F. Five of the six groups of irradiated specimens were subjected to individual 168 hr post-irradiation annealing treatments of 550, 560, 570, 580, 590 °F, respectively, while the sixth specimen set served as a control, being used to establish notch impact properties of the steel in the as-irradiated condition. The test results, with the exception of the 550 °F annealing data, are given in Figure 9.5.

The 275 °F radiation-induced increase in the Charpy-V 30 ft-lb transition temperature depicted by the data corresponds well with the projections for the described irradiation exposure. The 168 hr heat treatments at 560, 570, 580, and 590 °F all produced similar degrees of recovery of

FIGURE 9.4. Transition Temperature Behavior of A350-LF1 (Modified) Steel With Cyclic Irradiation and Intermediate 585 °F Annealing.

FIGURE 9.5. Postirradiation Heat Treatment of A350-LF1 (Modified) Steel at 560, 570, 580 and 590 °F Showing Similarity of Notch Ductility Behavior in the Various Annealed Conditions.

the initial Charpy-V 30 ft-lb transition temperature. The two higher heat treatment temperatures (580 and 590 °F), however, were more effective in overall embrittlement relief as suggested by the data points above the 45 ft-lb energy level. By contrast, the 550 °F heat treatment (not illustrated) was measurably less effective when compared to the heat treatments at 560 to 590 °F. With this annealing condition, only 64% of the initial 30 ft-lb transition temperature shift was recovered.

The effective coincidence of post-irradiation anneal data points in Figure 9.5 at and below the Charpy-V

30 ft-lb level marking the transition temperature indicates that the recovery of notch impact properties of A350-LF1 (Mod.) steel after 430 °F irradiation is not critically dependent upon the actual temperature of annealing in the range of 560 to 590 °F. A practical implication of this observation is that the Army SM-1A reactor pressure vessel should receive a satisfactory heat treatment in terms of achieving the anticipated percentage recovery in initial transition temperature, even if the actual temperature during annealing operations deviates (10 °F) from the planned nominal value of 585 °F.

NOTCH DUCTILITY BEHAVIOR OF A350-LF1
(MODIFIED) STEEL WITH CYCLIC 430 °F
IRRADIATION-168 HOUR ANNEALING

Full experimental assessment of the benefits of periodic heat treatment for embrittlement relief of a particular carbon or low alloy steel serving as a nuclear reactor structural component ultimately involves duplication of the irradiation and annealing conditions of the component, including its exposure temperature and the intervals at which the heat treatment will be applied. Such an evaluation has been performed for the Army SM-1A reactor pressure vessel. A sister forging fabricated from the same heat of A350-LF1 (Mod.) steel as the vessel was used for experimental tests (Table 9.10).

Data presented in the foregoing section indicated that the SM-1A steel is not overly sensitive to variations in annealing temperature over the range of 560 to 590 °F. However, comparisons of data presented in Figure 9.5 (430 °F irradiation) with data reported earlier⁽³⁾ for <240 °F irradiation and annealing (Figure 9.4) reveal a significant dependency of annealing response on prior neutron exposure temperature. Specifically, the percentage recovery by annealing was notably higher when the steel was irradiated at the lower temperature. Prior irradiation temperature was again demonstrated to be a significant factor in heat treatment response by the results of the most recent experimental series involving two complete cycles of 430 °F irradiation followed by 585 °F postcycle annealing. In this series, Charpy-V properties were

established at each interval of irradiation and annealing as was necessary for the development of Figure 9.4.

As shown in Figure 9.6, special half-length assemblies for irradiation in tandem were designed for this experimental operation wherein one assembly containing control test specimens could be removed from the reactor (and another inserted) while the other remained in place for a second period of exposure. Each assembly consisted of two specimen sections with individual and independently controlled section heaters. In addition, twin assembly temperature control and recording systems permitted the in-place annealing of one specimen assembly separate from the other as required at the end of the first irradiation period. The planned sequence of assembly irradiation operations together with notations as to the actual performance of the assemblies over their lifetimes are given in Table 9.11.

In view of the long term nature of the main assembly irradiations coupled with the number of major assembly design modifications required for separately removable units, the new assemblies performed surprisingly well. The major operational difficulty proved to be the loss of temperature control of Assembly 3 (second cycle control) after a short period of operation due to a weld failure in the by-pass section of the lead tube. However, data from this unit were not critical to the success of the experimental analysis or to the main objective of the experimental series. Similarly, problems with one

FIGURE 9.6. Specially Designed NRL Half-Length Assemblies for 430 °F Irradiation in Tandem. {Specimens contained in one assembly [lower photograph] were annealed in reactor at 585 °F for a period of 168 hr to represent annealing of Army SM-1A Reactor Vessel Steel, A350-LF1 (modified)}

of two section heaters in each of the remaining assemblies did not reduce or limit planned data acquisition as the specimens from a single assembly (two sections) were selectively intermixed for a statistical evaluation of both exposure temperature conditions. Results of such planned sampling in the test program are shown in Figures 9.7, 9.8, and 9.9.

Referring first to the data presented in Figure 9.7, transition temperature increases of 320 degrees (Section A: 430 °F) and 415 degrees (Section B: 430/350 °F) are indicated for fluences of 2.0×10^{19} and 2.7×10^{19} n/cm² is greater than 1 MeV respectively. From prior data compilations for this steel,⁽⁷⁾ it may be assumed that the difference in transition temperature increase is due to the difference in

fluence levels, rather than to the relatively small difference in irradiation temperatures. (Measured fluence differences will be discussed in greater detail below.) It is significant to this experimental series, however, that postirradiation annealing data for both sections are in close agreement with regard to absolute recovery (Charpy-V 30 ft-lb transition temperature of +5 °F). Thus, a good "fix" on the transition temperature at the end of irradiation-annealing Cycle 1 is provided though it is open to some speculation whether or not the greater percentage recovery found in one case is mostly due to a somewhat lower exposure temperature for the latter part of the in-reactor cycle, or, to the higher fluence level.

TABLE 9.11. Experimental Operations and Assembly Performance

Experimental Unit	Temperature, °F Irradiation Period 1, 12 weeks			Temperature, °F Anneal Period 1	Temperature, °F Irradiation Period 2, 10 weeks			Temperature, °F Anneal Period 2
	Month Number			In-Reactor, 168 hr	Month Number			X-Reactor, 168 hr
	1	2	3		4	5	6	
<u>Assembly Number 1</u>								
Section A	430	430	430	430 ^(a)	(Assembly discharged at end of anneal period 1)			
Section B	430	350 ^(b)	350	350 ^(a)				
<u>Assembly Number 2</u>								
Section A	430	430	430	585	430	380 ^(b)	380	585
Section B	430	430	430	585	430	430	430	585
<u>Assembly Number 3</u>								
(Second Cycle Only)								
Section A	-	-	-	-	200 ^(c)	200	200	585
Section B	-	-	-	-	200 ^(c)	200	200	585

(a) Annealed out-of-reactor.

(b) Heater output limited to less than maximum capacity.

(c) Assembly developed leak after 5 days operation at 430.

FIGURE 9.7. Cyclic 430 °F Irradiation and 585 °F Annealing of Army SM-1A Reactor Vessel Steel, A350-LF1 (Modified), Showing Charpy-V Notch Ductility Behavior at Both Stages of the First Cycle (Assembly 1; Control Unit)

FIGURE 9.8. Cyclic 430 °F Irradiation and 585 °F Annealing of Army SM-1A Reactor Vessel Steel, A350-LF1 (Modified), Showing Charpy-V Notch Ductility Behavior Following Re-irradiation and Re-annealing During the Second Cycle. [Postcycle annealing data for the first cycle are included in this figure (assembly 2)]

FIGURE 9.9. Cyclic 200 °F Irradiation and 585 °F Annealing of Army SM-1A Reactor Vessel Steel, A350-LF1 (Modified), Showing Charpy-V Notch Ductility Behavior at Both Stages of the Second Cycle. [The high percentage recovery depicted by annealing data are in accordance with the low irradiation temperature (assembly 1)]

In Figure 9.8, the results from specimens subjected to two complete irradiation-annealing cycles (Assembly 2) are shown referenced to postcycle annealing data for the first complete cycle (Figure 9.7). For the "as-irradiated, annealed, and re-irradiated" condition, three ductile-brittle transition curves are given and identified by three corresponding fluence levels. The formation of three distinct data point groupings from specimens in Section A and B was generally expected and may be readily traced to the "magnification" at high fluences of small, but measurable, differences in neutron flux over the length of the assembly. Thus, much larger than normal axial and transverse fluence gradients were particularly anticipated in the testing of specimens from Assembly 2. While not the normal case, the necessarily large vertical separation of the two assemblies (Figure 9.6) in the core piece and resultant placement of specimens away from the "flat flux region" about the fuel centerline readily produced this flux or fluence gradient situation.

Before analyzing and evaluating the data for two complete cycles, it may be well to acknowledge data developed from Assembly 3 which served as the second cycle control unit. In this case, measured fluence differences were not as severe ($\sim 1.4 \times 10^{19}$ and $\sim 1.8 \times 10^{19}$ n/cm² > 1 MeV), and the separation of data points was less distinct as shown in Figure 9.9. For subsequent data comparisons, the average transition temperature increase of approximately 320 degrees has been used with a neutron fluence value of 1.6×10^{19} n/cm². From Table 9.11,

however, it is again pointed out that the irradiation temperature was less than 200 °F for this unit which contributed directly to the high amount of transition temperature recovery illustrated by the postcycle annealing data.

On evaluating the data shown in Figure 9.8 on the basis of single cycle irradiation data presented in Figures 9.7, 9.9, and 9.4, and in earlier reports^(3,7), several observations on the annealing response of this steel forging may be made. First, transition temperature recovery for a given heat treatment following 430 °F irradiation is less than for the same heat treatment following <240 °F irradiation ($\sim 73\%$ versus $>90\%$). Secondly, irradiation of A350-LF1 (Mod.) steel followed by intermediate 585 °F annealing and $\sim 73\%$ recovery does not result in re-embrittlement at a more rapid rate than virgin material irradiated at the same temperature to the same neutron fluence (275/325° ΔT: Unit 2 versus 320° ΔT: Units 1 and 3). Thirdly, intermediate 585 °F-168 hour annealing of this steel applied once during 430 °F irradiation service can be quite beneficial toward limiting the total transition temperature increase, or toward maintaining the absolute transition temperature below some preset maximum value. For example, a transition temperature increase of 440° (360 °F absolute), would be projected for a neutron exposure of 3.9×10^{19} n/cm² > 1 MeV whereas intermediate mid-cycle annealing limited the total increase to 360° (280 °F absolute). Fourth, the benefits of periodic annealing would appear greatest with the more frequent annealing schedule

(after the first annealing heat treatment). Specifically, a transition temperature increase of 460 °F (380 °F absolute) would be projected for a fluence of $4.6 \times 10^{19} \text{ n/cm}^2 > 1 \text{ MeV}$ without annealing versus an increase of 410 °F (330 °F absolute) with annealing. Fifth, significant full shear energy absorption is retained by highly irradiated but intermediate-annealed specimens of the forging. Sixth and last, second cycle annealing data, while very limited, indicate that a 585 °F annealing treatment when applied the second time retains its degree of effectiveness in transition temperature recovery. Specifically, specimens re-irradiated to a total neutron fluence of 3.9 and $4.6 \times 10^{19} \text{ n/cm}^2$ is greater than 1 MeV and re-annealed showed 76.5% and 65% recovery of the second cycle transition temperature shift respectively.

The effectiveness of periodic 585 °F-168 hour annealing treatments for embrittlement relief of the Army-SM-1A reactor pressure vessel has been determined experimentally using steel representative of the actual vessel. Irradiation temperature and fluence conditions duplicating those in service prior to initial vessel annealing operations were also investigated. Two complete cycles of irradiation and annealing were examined with transition temperature behavior being established at each stage of cyclic operations. The major significance of the data lies in the illustration of relief of embrittlement for the maximum fluence condition of 3.9×10^{19} and $4.6 \times 10^{19} \text{ n/cm}^2 > 1 \text{ MeV}$ coupled with the observation that embrittlement is not accel-

erated during a second cycle exposure compared to the rate established for virgin material.

PREIRRADIATION MECHANICAL PROPERTIES OF MH-1A PRESSURE VESSEL STEEL

One part of the current effort by NRL for the Army Nuclear Power Program (ANPP) was the design of a suitable pressure vessel surveillance program for the barge-mounted, MH-1A reactor, and the fabrication of the necessary irradiation capsules and associated hardware. In order to make the surveillance program as meaningful as possible, the test specimens were machined from nozzle cutouts of the upper shell course ring forging. This forging was produced by the Standard Steel Division of the Baldwin-Lima-Hamilton Corporation according to specification A-182 F316 S/S, and is identified with Heat No. E-5367. The chemistry of this heat, as determined both by the manufacturer and by NRL, is shown in Table 9.12.

Test specimens for the surveillance capsules were taken from five layers through the thickness of the 3 1/8 in. forging. The properties of the steel at these layers prior to irradiation service are documented here for future reference. The individual specimen layers are separately identified as Inner Radius, Mid-Inner Radius, Middle, Mid-Outer Radius, and Outer Radius.

The Charpy-V specimens representative of the five thickness locations were tested over the temperature range of -250 to 500 °F. In every case, the specimens were bent into a U-shape and absorbed the full impact energy of the pendulum (in excess of 200 ft-lb)

wherein only slight ductile tearing in the vicinity of the V-notch was observed. In the region of maximum strain about the V-notch, the specimen surfaces developed an "orange peel" appearance not unusual for forged stainless steels. Tensile properties at ambient temperature and at 500 °F, depicted by specimens representing four of the five layers in the 3 1/8 in.

forging, are presented in Table 9.13. Again, specimens tested at both temperatures displayed the characteristic "orange peel" surface appearance indicative of material working capability. Both the 500 °F and ambient temperature data show a trend of slight degradation in properties toward the middle of the forging, but the differences are so slight as to be of little

TABLE 9.12. Chemical Composition of MH-1A Reactor Pressure Vessel Surveillance Steel, Standard Steel Division, Baldwin-Lima-Hamilton Corp. Heat No. E-5367, ASTM A-182 F316 S/S

Sample	C	Mn	P	S	Si	Ni	Cr	Mo	Co	Ta
Ladle (BLH)	0.058	1.70	0.018	0.007	0.63	10.90	17.83	2.41	0.02	0.09
Check (BLH)	0.063	1.74	0.017	0.009	0.58	10.87	17.83	2.46	0.02	0.09
NRL	0.060	1.51	0.014	0.010	0.57	10.80	17.60	3.00	0.01	-

TABLE 9.13. Tensile Properties of MH-1A Reactor Pressure Vessel Surveillance Steel, Standard Steel Division, Baldwin-Lima-Hamilton Corp. Heat No. E-5367, ASTM A-182 F316 S/S

Forging, Radius Location	Test Temperature, °F	Yield Strength, 0.2% Offset, ksi	Tensile Strength, ksi	Reduction in Area, %
Mid-Inner	Ambient	40.05	81.2	83.0
Mid-Inner	500	28.0	64.4	66.0
Mid-Inner	500	30.4	66.0	71.7
Middle	500	28.4	70.6	63.6
Middle	500	28.5	68.6	66.9
Mid-Outer	Ambient	37.8	79.5	84.6
Mid-Outer	500	27.4	64.2	73.0
Mid-Outer	500	29.0	65.4	68.7
Outer	Ambient	41.0	81.0	78.8
Outer	500	28.9	64.8	67.9
Outer	500	29.1	70.0	70.4

consequence. Noteworthy for the material are the high reduction of area values.

As specimens from the pressure vessel surveillance program of the MH-1A are withdrawn in the future, the test results will be published and compared with the values reported here for the unirradiated condition.

NOTCH DUCTILITY OF SEVERAL REACTOR STRUCTURAL STEELS AFTER IRRADIATION IN A HEAVY WATER MODERATED REACTOR

Previous research at NRL⁽⁸⁾ has included gathering of notch ductility data on several candidate reactor structural steels after their irradiation in widely different reactor neutron spectra. Two pressure vessel steels, ASTM A212-B and A302-B, which have been irradiated in light water moderated reactors as well as in air-graphite and heavy water moderated reactors, have now been irradiated in a second heavy water moderated reactor together with ASTM A517-E, a new and potentially important structural steel. A plan view of the heavy water moderated CVTR wherein this irradiation took place is shown schematically in Figure 9.10. The CVTR operates on the principle of containment of pressure and temperature within U-shaped pressure tubes, while the heavy water reflector portion of the reactor vessel (where the NRL capsule was located) remains at a temperature well below 240 °F. Although the NRL capsule was almost 24 in. in length, its neutron exposure was quite uniform since the capsule was centered at midplane of the greater than 8 ft high fuel core and was rotated 180 degrees midway through the irradiation.

The chemical composition and heat treatment of the three steels irradiated in the CVTR experiment are presented in Table 9.14. The notch ductility characteristics of these steels before and after irradiation are summarized in Table 9.15, and the nil-ductility transition (NDT) temperature increases, translated along the 30 ft-lb energy level, are shown schematically in Figures 9.11 through 9.13 for A212-B, A302-B, and A517-E steels respectively. The neutron fluence data shown in Figures 9.11 through 9.13 are all based upon the assumption of a fission spectrum distribution of neutrons at the NRL capsule location in the CVTR. A value of 68 mb, averaged over the entire fission spectrum, was used for the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ reaction cross section.

The continuing objective of this study has been the determination of the interrelation of mechanical property data from irradiations of A212-B and A302-B steel in reactors of differing moderating media. The principle feature of the study is the calculation of the distribution of neutrons by energies in the spectrum of each reactor location utilized and the evaluation of the body of data in that light. Accordingly, the distribution of neutrons by energy groups has been calculated for the 10-L location in the CVTR by Dahl and Ulseth at the Battelle-Northwest Laboratory under contract to NRL (Figure 9.14). With this information, Dahl and Ulseth re-determined the activation cross section for the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ reaction in this spectrum to account for neutrons of energies >1 MeV and >0.5 MeV. The spectrum-averaged cross sections, which

CAROLINAS VIRGINIA TUBE REACTOR

FIGURE 9.10. Schematic Representation of the Core Arrangement in the Carolinas-Virginia Tube Reactor Showing the Location of the NRL Irradiation Capsule.

are developed from a fission averaged cross section for the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ reaction of 68 mb, are presented in Table 9.16.

The CVTR data thus may be plotted in concert with the body of data already developed for low temperature irradiation of A212-B and A302-B steels. The A212-B and A302-B steel ΔNDT data have been plotted versus neutron fluence (n/cm^2), in Figures 9.15 and 9.16 respectively, by accounting for neu-

trons of energies >1 MeV in an assumed fission spectrum (open points), and alternately of energies >0.5 MeV in the calculated spectra of all the reactors involved (closed points). Close inspection of the curves is not necessary to determine that the calculated spectrum CVTR data points (closed symbols) do not fit the remainder of the calculated spectra data body.

Several interesting observations can be made concerning the data in

TABLE 9.14. Chemical Composition and Heat Treatment of Three Steels Irradiated in the Carolinas-Virginia Tube Reactor

Type of Steel	Chemical Analysis, wt%				
	C	Mn	Si	P	S
A212-B	0.26	0.76	0.24	0.011	0.031
A302-B	0.20	1.31	0.25	0.012	0.023
A517-E	0.18	0.51	0.28	0.017	0.020

Type of Steel	Chemical Analysis, wt%					
	Ni	Cr	Mo	Al	Cu	Ti
A212-B	0.22	0.20	0.02	-	-	-
A302-B	0.20	0.17	0.47	-	-	-
A517-E	0.08	1.72	0.50	0.036	0.26	0.085

4 in. A212-B -- Austenitized at 1650 °F for 2 hr; water quenched; tempered at 1175 °F for 4 hr; furnace cooled to below 600 °F.

6 in. A302-B -- Austenitized at 1650 °F for 2 hr; water quenched; tempered at 1200 °F for 6 hr; furnace cooled to below 600 °F.

6 in. A517-E -- Austenitized at 1650 °F; water quenched; tempered at 1225 °F for 1 1/2 hr; retempered at 1240 °F for 2 hr.

TABLE 9.15. Charpy-V Notch Ductility Characteristics for Three Steels After 240 °F irradiation in the Carolinas-Virginia Tube Reactor

Type of Steel	CV 30 ft-lb			Full Shear Energy Absorption, ft-lb	
	NDT	Increase, °F	ΔT	Initial Average	Radiation-Induced Loss
A212-B	5	255	250	82	24
A302-B	30	270	240	72	14
A517-E	-45	165	210	115	28

FIGURE 9.11. Charpy-V Notch Ductility Characteristics of ASTM A212-B Plate Before and After Irradiation at 240 °F in the Carolinas-Virginia Tube Reactor.

FIGURE 9.12. Charpy-V Notch Ductility Characteristics of ASTM A302-B Plate Before and After Irradiation at 240 °F in the Carolinas-Virginia Tube Reactor.

FIGURE 9.13. Charpy-V Notch Ductility Characteristics of ASTM A517-E Plate Before and After Irradiation at 240 °F in the Carolinas-Virginia Tube Reactor.

FIGURE 9.14. Schematic Representation of the Neutron Spectrum in the 10-L Position of the Carolinas-Virginia Tube Reactor.

TABLE 9.16. Spectrum-Averaged Cross Sections for ^{54}Mn Activation and Thermal-to-Fast (>1 MeV Fission) Flux Ratios at Midplane of Position 10-L, Carolinas-Virginia Tube Reactor

ϕ 2200 m/sec	$\bar{\sigma}$, barns	(calculated spectrum)
$\phi > 1$ MeV, Fission	$n/\text{cm}^2 > 1$ MeV	$n/\text{cm}^2 > 0.5$ MeV
147 : 1	0.0984	0.0784

FIGURE 9.15. NDT Temperature Increase (Indexed to Charpy-V 30 ft-lb level) for A212-B Steel Irradiated at Temperatures <250 °F Plotted Versus Neutron Fluences for $n/\text{cm}^2 > 1$ MeV in an Assumed Fission Spectrum (Open points), and for $n/\text{cm}^2 > 0.5$ MeV in the Calculated Spectra of the Individual Reactors (Closed points)

FIGURE 9.16. NDT Temperature Increase (Indexed to Charpy-V 30 ft-lb level) for A302-B Steel Irradiated at Temperatures <250 °F Plotted Versus Neutron Fluences for $n/\text{cm}^2 > 1$ MeV in an Assumed Fission Spectrum (Open points), and for $n/\text{cm}^2 > 0.5$ MeV in the Calculated Spectra of the Individual Reactors (Closed points)

Figures 9.15 and 9.16. First, the CVTR data points as well as the data points from the air-graphite and other heavy water reactor irradiations all indicate more radiation damage than do the data points from the light water moderated reactor irradiations. This fact agrees with the thesis of Wechsler⁽⁹⁾ that if the thermal neutron population is great enough, (n, γ) recoil events caused by thermal neutrons can produce significant changes in mechanical properties in reactor structural steels. In this connection, note that the thermal-to-fast ($>1\text{MeV}$) neutron ratio for the CVTR is 147 to 1; by way of comparison, the ratio for the air-graphite reactor is 22.5 to 1, and the applicable ratios for the light water moderated reactor irradiation facilities in this study are on the order of 1 or 2 to 1. Therefore, noticeable mechanical property changes could be expected from (n, γ) recoil events on the basis of the large thermal neutron populations in the non-light water moderated reactor environments.

Another interesting feature of the CVTR data is that it is similar to other data from a heavy water moderated reactor regarding the closeness of the respective fission spectrum and calculated spectrum data points. By comparison, notice that the data points based on a calculated spectrum for the air-graphite and light water moderated reactor irradiations are relatively far to the right of the fission spectrum data points. Obviously, the overall spectral differences in the heavy water moderated reactors are significantly different from those of the other types.

In summary, data from heavy water moderated reactor irradiations apparently cannot be accurately evaluated by comparison with data from light water moderated reactors using the treatment indicated in Figures 9.15 and 9.16. More sophisticated analyses will be required of heavy water irradiation data for comparison with data from other significantly different reactor types if consistent trends in notch ductility property changes are desired for accurate predictions of future material behavior under neutron irradiation.

MODIFICATION, INSTALLATION, AND INITIAL OPERATION OF A REMOTELY OPERATED, TAPE CONTROLLED MILLING MACHINE

The remotely operated machine shop in the High Level Radiation Laboratory (HLRL) was conceived as a multipurpose facility with a capability for a number of machining operations including milling, turning, grinding, sawing, drilling, and broaching as well as a welding capability. The most recent permanent addition to the facility is a Pratt and Whitney tape-controlled milling and drilling machine. In addition to design features, this machine was chosen because it was the only machine found to be in production on a routine basis. Thus, it was considered that the concept had had sufficient exposure to be thoroughly proof tested, a most desirable feature for hot cell equipment.

The machine as built was not remotely operable, however, and many modifications were required. Modifications were made while the milling machine was located in an assumed operating position in the HLRL mockup area (Figure 9.17). The importance of this area

FIGURE 9.17. Hot Cell Mockup Facility in the Nonradiation Area. (A new tape-controlled milling machine for use in the machine shop cell is shown with the remote controls at an assumed operating station.)

and the benefits of equipment mockup were well demonstrated as this work proceeded. Using master-slave manipulators identical to those installed in the hot cell, it was possible, for example, to thoroughly evaluate each modification in progress and to maximize its future effectiveness. Thus, installation within the hot cell could be confidently planned with exact assurance of its abilities as a remotely operable tool.

The first modification requirement was the replacement of the straight-through wiring from control cabinet to milling machine with wiring that had quick-connect electrical plugs at either end, and included the installation of junction boxes on the console and milling machine for termination of the wiring. Control cables (30 ft long) were specially fabricated and installed in the U-tubes under the wall of the machine shop cell (42 in. thick) for operations within the hot cell; additional 10 ft control cables were fabricated for use during the modification period. The additional cables will also be used in event of machine failure that may require its removal from the hot cell for repairs.

Another series of modifications involved the installation of protective coverings to prevent the loss of radioactive chips in inaccessible areas of the milling machine. The first of these was a curtain fabricated on the principle of a window shade which was installed between the frame of the milling machine and the saddle. As the saddle moves in and out, chips are dumped into a remotely removable tray underneath the curtain, or, if any

should remain on the curtain, these are removed by remote vacuuming. A skirt was then installed around the remaining three sides of the table which effectively isolated the area beneath the machine from radioactive chips generated during the machining operations. Finally, extensions were added to the front and sides of the table. In addition to providing a generous work surface, the table with its vertical extensions at the outer perimeter has virtually eliminated the scattering of chips to areas inaccessible for remote housekeeping.

To facilitate the interchanging of holding fixtures and vises to be used on the milling machine, a 4 in. thick plate was mounted to the table. Holes were drilled and tapped for each clamping fixture, then plugged with a threaded rod to protect the threads and to avoid entrapment of radioactive contaminants. The plate eliminates the difficult task of decontaminating the T-slots found in standard milling machine tables, yet provides the flexibility required to meet special hot cell requirements.

Unlike conventional milling machines, the table on this machine is limited to horizontal (x and y) movement. The required (Z-axis) flexibility is provided through a broad range of head and spindle adjustment. The head movement (approximately 14 in.) is motorized and is controlled at the remote operating station.

Vertical positioning of the head is monitored by a special long range dial gage indicator. Then, to assure accurate machining, the head is locked into position by the action of solenoid

operated air cylinders on four cam-type locks. Final positioning of the milling cutter is performed manually using the master-slave manipulators to adjust the vertical position of the spindle. The spindle movement is also monitored by a dial gage and, upon achieving the desired setting, it too is securely locked before the actual cutting operation is begun. Precision measuring equipment utilizing numerical readouts will be available in the

near future and will be used to expedite the vertical positioning of the milling cutter. In addition, this equipment will serve to enhance final quality control of the remote machining operations.

The remaining modifications to the mill can be seen in Figure 9.18 and include the installation of the following: handles compatible with the gripping fingers of the master-slave manipulators, dial gages suitable for

FIGURE 9.18. The Remotely Operated Milling Machine is Shown Viewed Through the Cell Window Illustrating the Following Features: (1) New Table Surface Plate, (2) Vacuum Fixture, (3) Pneumatically Operated Head Locks, (4) Automatic Spindle Feed Actuator, (5) Spray Mist Coolant System, (6) Table Extensions, (7) Dial Gage Indicators for Monitoring Spindle and Head Movement, and (8) A Special Fixture for Machining Gripping Grooves in the Shoulders of Radioactive Tensile Specimens

monitoring head and spindle movement, a mist type coolant system with a remotely refillable 1 gal capacity reservoir, and a spotlight for improved illumination of the work piece.

After all modifications had been accomplished, the machine was transferred to and operated by the production machining group of NRL to determine and correct any abnormalities that are generally associated with new equipment. After four months of production-type operation, the machine was installed in the machine shop hot cell and was placed in service the following day. At the present time, this machine receives more continuous usage than any other hot cell tool but has operated without fault or further need of modification for five months. It should be noted that machining tolerances have not been relaxed in any case because of the handicap imposed by the remote operational requirements.

REMOTELY OPERATED PRECISION MEASURING EQUIPMENT

The Reactor Materials Branch's technique for remotely positioning irradiated Charpy-V impact specimens on the test anvil utilizes four solenoid actuated air cylinders; two cylinders center the notch while the other two force the specimen firmly against the anvil face. The system depends upon close tolerances on the specimen length (± 0.002 in.) as well as on the placement of the notch at the exact center of the specimen length.

The special device shown in Figure 9.19 was designed to permit routine determinations of deviations, if any, from the NRL standard specimen dimensions or notch placement. The design and fabrication of this device was especially prompted by the frequent receipt of highly radioactive specimens belonging to other agencies for testing in the HLRL which did not contain

FIGURE 9.19. Charpy-V Notch Placement Measuring Fixture. (Evaluating the specimen in place, the dial gages indicate that the notch is centered but the specimen is short by 0.002 in.)

centered notches. The device utilizes two opposed dial gage indicators and a centering point which precisely mates with the specimen V-notch. When a perfectly machined specimen is in position, both dial gages register zero. Any deviation from this value represents the adjustment required for the anvil alignment mechanism to properly center a specimen having an off-center notch for testing.

The notch-locating device has been coupled with another measuring device, as shown in Figure 9.20, for post-test lateral expansion measurements of high strength steel (above 100 ksi YS) specimens which do not feature a well defined fracture appearance. The device is also used for those materials such as stainless steel which do not exhibit a brittle-ductile transition. The philosophy of simplicity, when

possible, for hot cell equipment has been followed in the design of this device. A single switch engages two solenoids which retract the dial gage pointers. The specimen is then mechanically secured in a fixture and, upon releasing the dial gage pointers, the total lateral expansion is indicated by the combined deviation from zero on the dial gages.

As evidenced by the photographs, these instruments are simple in design and economical to fabricate. Experience with the hot cell unit thus far has shown it to be a very useful tool in remote evaluations of high strength materials. Its most important function, however, is its significant reduction of the number of man-hours required in the remote measurements of nonstandard radioactive test specimens from outside laboratories.

FIGURE 9.20. The Notch Placement Device is Shown Coupled with the Lateral Expansion Measuring Fixture. (A high strength steel specimen has been locked in position and lateral expansion is shown as the deviation from zero on the dial gages)

EFFECT OF RADIATION ON REFRACTORY METALS AND HEAT RESISTANT ALLOYS

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REFRACTORY METALS AND ALLOYS

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

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

EXPERIMENTAL PROGRAM

Tungsten Creep Rupture: Fluence Dependency - J. Moteff and F. D. Kingsbury

An investigation to determine the influence of thermal and fast neutron fluence on the creep rupture properties of tungsten at 1100 °C was conducted. The specimens were irradiated in either the ORR or ETR at reactor ambient temperature to fast fluence levels of 8.5×10^{17} to 1.5×10^{21} neutrons/cm² ($E_n \geq 1$ MeV). The data from these tests, all conducted at a

stress level of 18.28 kg/mm², are presented in Table 10.1 and Figures 10.1 and 10.2.

Creep rate or rupture life versus fast ($E \geq 1$ MeV) neutron fluence plots (Figure 10.1a and b) show that increasing the fluence results in a significant decrease in creep rate and an increase in the rupture life. The creep rate, for instance, was reduced by a factor of 500; the rupture life was increased by a factor of about 40 and the elongation at fracture was reduced from an average value of 30% down to about 3.8%.

The effect of the different reactor neutron spectra may be noted in these plots. The distinct discontinuities in the curves for the ORR or ETR irradiations are principally due to the differences in the ratio of the respective thermal to fast neutron flux densities. This effect can be especially seen in the ORR-irradiated specimen where the ratio of thermal to fast neutron flux was deliberately altered by shielding the specimen with 0.5-mm-thick cadmium cover.* Here, the specimen, with its lower ratio of thermal to fast neutron fluence, exhibited a higher creep rate and shorter rupture life than would be predicted on the basis of fast neutron fluence level. Since

* This resulted in a factor of 10 fewer thermal neutrons for the same fast neutron fluence.

TABLE 10.1. Summary of Creep-Rupture Tests on Tungsten Specimens at 1100 °C and 18.28 kg/mm²

Specimen No. ^a	Capsule	Fluence, nvt		Linear Creep Rate, ε, (sec ⁻¹)	Rupture Life, t _r , (hr)	Elongation, ^b %	Reduction in Area, %
		Thermal	Fast (E _n ≥ 1 Mev)				
Rod P Specimens							
1336	Control			4.46 × 10 ⁻⁶	7.99	—	—
1351	ORM-31 ^c	3.9 × 10 ¹⁹	4.0 × 10 ¹⁸	3.87 × 10 ⁻⁶	12.65	35.7	90.0
1359	ORM-33	4.1 × 10 ¹⁹	5.9 × 10 ¹⁸	2.86 × 10 ⁻⁶	15.22	29.8	94.9
1362	ORM-34	2.3 × 10 ²⁰	3.8 × 10 ¹⁹	1.20 × 10 ⁻⁶	43.98	24.4	92.9
1377	ORM-37 ^d	2.5 × 10 ¹⁹	3.9 × 10 ¹⁹	5.0 × 10 ⁻⁷	21.04	14.0	93.5
1373	ORM-36	4.9 × 10 ²⁰	7.9 × 10 ¹⁹	1.23 × 10 ⁻⁷	58.38	12.2	89.0
1369	ORM-35 ^e	7.7 × 10 ²⁰	1.2 × 10 ²⁰	2.66 × 10 ⁻⁸	352.1	28.9	87.4
Rod Q Specimens							
1390	Control			5.13 × 10 ⁻⁶	7.82	31.0	98.3
1391	Control			6.39 × 10 ⁻⁶	6.23	30.3	98.7
1393	Control			4.52 × 10 ⁻⁶	9.10	32.4	70.2
1384	ORM-32	6.8 × 10 ¹⁸	8.5 × 10 ¹⁷	6.26 × 10 ⁻⁶	6.87	33.9	95.9
1383	ORM-31	3.9 × 10 ¹⁹	4.0 × 10 ¹⁸	3.60 × 10 ⁻⁶	12.61	28.0	90.0
1385	ORM-33	4.1 × 10 ¹⁹	5.9 × 10 ¹⁸	3.4 × 10 ⁻⁶	15.92	32.2	95.1
1395	ORM-34	2.3 × 10 ²⁰	3.8 × 10 ¹⁹	1.08 × 10 ⁻⁶	41.92	26.3	92.3
1394	ORM-37 ^d	2.5 × 10 ¹⁹	3.9 × 10 ¹⁹	3.0 × 10 ⁻⁶	14.64	28.0	90.0
1388	ORM-35	7.7 × 10 ²⁰	1.4 × 10 ²⁰	2.98 × 10 ⁻⁷	194.9	33.5	92.4
1397	2-233 ^e	3.0 × 10 ²⁰	1.4 × 10 ²⁰	1.30 × 10 ⁻⁶	64.16	32.3	92.9
1400	2-233	3.0 × 10 ²⁰	1.4 × 10 ²⁰	9.6 × 10 ⁻⁷	42.47	22.9	54.6
1386	2-232	5.6 × 10 ²⁰	2.3 × 10 ²⁰	4.1 × 10 ⁻⁷	64.86	12.7	57.3
1403	2-234	2.7 × 10 ²¹	1.5 × 10 ²¹	1.08 × 10 ⁻⁸	315.41	3.79	32.5
1405	2-234 ^f	2.7 × 10 ²¹	1.5 × 10 ²¹	2.87 × 10 ⁻⁸	644.23	10.2	43.9

^aSpecimens from 0.508-cm-diameter tungsten rod (powder metallurgy) post-grind annealed at 1750°C for 1 hour in hydrogen.

^bPercent in 4.54-cm gage length.

^cORM-31 through -37 represent numbers of capsules irradiated at reactor ambient temperatures in ORR-A2 facility.

^dSpecimens shielded with 0.51-mm cadmium.

^e2-232 through 2-234 represent identifications of capsules irradiated in the ETR F-13 and J-14 facilities at reactor ambient temperature.

^fAnnealed at 1700°C for 30 hours in hydrogen prior to creep-rupture testing.

the fast neutron fluence of the corresponding unshielded specimen was essentially the same as that of the shielded specimen, the main difference between the two is the rhenium content caused by transmutation from the (n, γ) reaction with tungsten.

As shown in Figure 10.2a and b, a much better ordering of experimental data is obtained in these thermal neutron fluence plots; i.e., they do not show discontinuities in the curves due to the different reactor spectra. The cadmium-shielded, ORR-irradiated specimen also shows a reasonable fit to both the creep-rate and rupture-life curves. At this test temperature (1100 °C), where most of the neutron-induced displacement defects are believed to be completely annealed out, the remaining radiation-induced defects

would be the transmutation products rhenium and osmium; e.g., it was calculated⁽¹⁾ that 2.5% rhenium atoms are produced from tungsten at a thermal neutron fluence of 2.7×10^{21} neutrons/cm². The decrease in the effective flow stress for a fixed applied stress due to the added strengthening of the tungsten by rhenium atoms would account for the observed decrease in creep rate. Preliminary calculations, assuming that solution hardening is proportional to the square root of the rhenium atom concentration, shows good agreement with the experimental data.

Molybdenum Creep Rupture - J. Moteff, F. D. Kingsbury and R. L. Treinen

Tests were conducted on arc-cast molybdenum to investigate the effect

a. Minimum creep rate

b. Rupture life

FIGURE 10.1. Minimum Creep Rate and Rupture Life of Irradiated Polycrystalline Tungsten, Rod Q at 1100 °C and an Applied Stress of 18.28 kg/mm^2 as a Function of Fast Neutron Fluence ($E \geq 1$ MeV)

a. Minimum creep rate

b. Rupture life

FIGURE 10.2. Minimum Creep Rate and Rupture Life of Irradiated Polycrystalline Tungsten at 1100 °C and an Applied Stress of 18.28 kg/mm² as a Function of Thermal Neutron Fluence

of test temperature on the creep rupture properties and on the ductility at fracture and also to further investigate any accelerated creep rates or reductions in the time to rupture in the material irradiated at the lower neutron fluences. Accelerated creep rates had been exhibited previously⁽²⁾ in the 0.29 to 0.32 T_m range (560 to 650 °C) in specimens irradiated 2 to 5 x 10¹⁹ neutrons/cm² ($E_n \geq 1$ MeV).

Specimens fabricated from two heats of material were tested. One heat was a commercial heat containing 290 ppm carbon and 10 ppm oxygen. The other heat contained 30 ppm carbon and approximately 26 ppm oxygen and was vacuum stress-relieved by the vendor. The higher carbon content material possessed greater strength and hence, was not directly comparable to the other heat.

The test results in the temperature range from 580 to 980 °C are presented in Table 10.2. It can be seen that specimens irradiated to a fast neutron fluence of 1.1 x 10²⁰ neutrons/cm² had longer rupture lives and lower creep rates than that of the corresponding control specimens whereas the specimens irradiated at the lower fast neutron fluences of 3.0 x 10¹⁹ and 4.8 x 10¹⁸ neutrons/cm² possessed shorter rupture lives and higher creep rates.

The irradiated (1.2 x 10²⁰ neutrons/cm²) specimen tested at 700 °C exhibited about a 30 hr incubation period in which the creep was abnormally low (Figure 10.3). Although this phenomenon had been noted previously⁽³⁾ in irradiated (ambient temperature) tungsten specimens; it

had not been observed in molybdenum specimens. Specimens irradiated to the same fast neutron fluence and tested at higher temperatures (850 and 980 °C) did not show any pronounced incubation period.⁽⁴⁾

The 580 and 600 °C tests involved specimens from the low carbon heat of molybdenum. The preliminary results (Figure 10.3) from this series of tests show accelerated creep at 580 °C for the lower neutron fluence (4.8 x 10¹⁸ neutrons/cm²) irradiated specimens. This accelerated creep was exhibited at two stress levels (Table 10.2). One specimen irradiated to 3 x 10¹⁹ neutrons/cm² showed negligible creep for 259 hr at 580 °C and the test was terminated.

The irradiated specimen (3 x 10¹⁹ neutrons/cm²) tested at 600 °C exhibited (Figure 10.3) a relatively long incubation period of approximately 165 hr in which the creep rate was extremely low. This stage was followed by an abrupt accelerated flow rate terminating in fracture. It appears that some defect which caused the hardening required a long period of time at the relatively low temperature to be removed sufficiently to permit dislocation mobility.

RESISTIVITY STUDIES - L. K. Keys, J. P. Smith and J. Moteff

The previously reported studies⁽⁵⁾ of the isochronal resistivity recovery of defects, introduced into several pre-annealed W, Mo, Mo-TZM (Mo-0.5Ti-0.08Zr), and W-25Re specimens by irradiation with fast ($E_n \geq 1$ MeV) neutrons at ambient (70 to 80 °C) reactor temperatures, were completed.

The results of these studies indicate that the high-temperature recovery mechanisms are very complex, and are greatly modified by the presence of impurities and because of the production of impurity elements by neutron-activated transmutations. Studies of these effects are needed to gain better knowledge of the influence which point defects and cluster defects as well as impurities may have on the mechanical properties of these alloys. The specimens were polycrystalline and were annealed in a flowing argon

atmosphere for 1 hr; the tungsten and W-25Re specimens were given a 1900 °C anneal while molybdenum and Mo-TZM specimens were annealed at about 1250 °C. All recovery studies were carried out under the same experimental conditions. The resistivity measurements were made at -196 °C using a previously described resistivity bridge.⁽⁵⁾

TUNGSTEN

A number of recrystallized, polycrystalline tungsten specimens were

TABLE 10.2. Summary of Molybdenum Creep-Rupture Data

Specimen No. ^a	Heat No. ^b	Condition ^c	Test Temperature, °C	Stress, kg/mm ²	Rupture Life, hr	Linear Creep Rate, sec ⁻¹	Elongation, ^d %
2115	C-6605	ORM-38	980	10.55	13.54	7.44×10^{-6}	77.0
2120	C-6605	Control	980	10.55	>10.6	7.19×10^{-6}	(>56.8) ^e
2123	C-6605	Control	850	14.00	57.52	1.56×10^{-6}	73.0
2116	C-6605	ORM-38	850	13.82	80.12	1.17×10^{-6}	68.8
2118	C-6605	ORM-38	850	13.30	107.44	9.94×10^{-7}	63.5
2124	C-6605	Control	700	19.00	288.04	$\sim 3 \times 10^{-7}$	60.2
2103	C-6605	Control	700	20.00	118.91	6.09×10^{-7}	49.2
2122	C-6605	ORM-38	700	20.00	180.48	4.11×10^{-7}	46.8
2664	C-7720	Control	600	19.40	59.1	6.22×10^{-7}	44.3
2685	C-7720	ORM-43	600	19.40	206.98	7.15×10^{-9}	32.7
2656	C-7720	Control	580	19.40	221.15	1.87×10^{-7}	45.2
2684	C-7720	ORM-43	580	19.40	(259) ^f	(<1 x 10 ⁻⁹)	—
2679	C-7720	ORM-44	580	19.40	98.45	5.43×10^{-7}	50.1
2670	C-7720	Control	580	20.00	81.4	3.75×10^{-7}	40.1
2696	C-7720	ORM-44	580	20.00	132.35	4.04×10^{-7}	51.4
2711	C-7720	Control	580	20.00	(246) ^f	2.02×10^{-7}	—

^aSpecimens from 0.5-mm-thick arc-cast molybdenum sheet post-grind annealed at 1200 °C in vacuum for 1 hour.

^bHeat C-6605 is commercial molybdenum with 290-ppm carbon content and 10-ppm O₂ content. Heat C-7720 is low-carbon molybdenum with 30-ppm carbon content and 26-ppm O₂ content.

^cIrradiations at reactor ambient conditions to fast neutron fluence ($E_n \geq 1$ Mev):

ORM-38, 1.1×10^{20}

ORM-43, 3.0×10^{19}

ORM-44, 4.8×10^{18}

^dPercent in 2.54-cm gage length

^eLever arm accidentally unloaded against stop while in third-stage creep.

^fTest discontinued at time indicated.

FIGURE 10.3. Elongation Versus Time Curves for Unirradiated Molybdenum Specimens Tested at Various Temperatures and Stresses

irradiated in various reactor facilities to fast ($E_n \geq 1$ MeV) neutron fluences ranging from 8.5×10^{17} to 1.5×10^{21} neutrons/cm². The isochronal resistivity recovery increment ($\Delta\rho$), from 0° to 1500 °C, was used as a measure of the radiation-induced damage in these specimens; this resistivity increment is plotted in Figure 10.4 versus the annealing temperature for several specimens. The derivative $\left[\frac{d}{dT} \left(\frac{\Delta\rho}{\Delta\rho_0} \right) \right]$ of the normalized resistivity recovery increment $\frac{\Delta\rho}{\Delta\rho_0}$ with respect to

temperature is presented in Figure 10.5. It is evident that there are several recovery regions or peaks present as indicated by the numerous maxima observed in this plot. Partic-

ular significance is attached to recovery regions as they relate to the melting temperature, T_m (3410 °C). These important regions are: $0.15 T_m$, $0.22 T_m$, $0.31 T_m$, and $0.35 T_m$; these have been described as representing recovery of specific types of defects (although which temperature refers to what type of defect is apparently still a controversy). These recovery regions have been called recovery stages and are referred to by numbers. In this section, the $0.15 T_m$ peak is Stage III and the $0.31 T_m$ peak Stage IV. The other peaks will not be referred to as specific stages at the present time.

It has been previously considered in this laboratory⁽⁵⁾ that $0.15 T_m$ corresponds to interstitial migration

FIGURE 10.4. Isochronal Resistivity Recovery of Neutron-Irradiated, Recrystallized Tungsten as a Function of Annealing Temperature

FIGURE 10.5. Derivative of the Normalized Isochronal Resistivity Recovery of Neutron-Irradiated, Recrystallized Tungsten as a Function of Annealing Temperature

or recovery; $0.22 T_m$ corresponds to di-vacancy migration; $0.31 T_m$ corresponds to vacancy migration; and $0.35 T_m$ corresponds to polygonization. These may change as more experimental data is generated and evaluated. The exact temperature of recovery, listed according to its relative melting temperature fraction, for all the described recovery regions as well as the irradiation conditions employed are presented in Table 10.3. The

temperature given for a particular recovery refers to the temperature at which the maximum in the recovery peaks in Figure 10.5 occurs. The stages in which a temperature is not presented result from lack of observation of a definite maximum in the recovery peaks.

The recovery observed at $0.35 T_m$ appears to vary randomly with the fluence and gives an average value of about 1027°C which agrees very well with the predicted $0.35 T_m$ of 1016°C .

TABLE 10.3. Defect Recovery Stages in Neutron Irradiated Tungsten
Determined for Isochronal Resistivity Studies

Sample No.	Rod	Capsule	Irradiation Conditions ^a		Maximum Rate of Recovery Temperature, °C ^b , and Area Under Respective Recovery Peak, micro-ohm-cm								Residual Resistivity, micro-ohm-cm ± 0.005
			Fluence, nvt										
			Fast (E _n ≥ 1 Mev)	Thermal	0.15 T _m	Area	0.22 T _m	Area	0.13 T _m	Area	0.35 T _m	Area	
354	A	ORM-32	8.5 x 10 ¹⁷	6.8 x 10 ¹⁸	362	0.196	568	0.021	867		992	—	—
101	D	ORM-7	5.4 x 10 ¹⁸	2.6 x 10 ¹⁹	345	0.604	550	0.077	—	—	—	—	—
355	A	ORM-33	5.9 x 10 ¹⁸	4.1 x 10 ¹⁹	333	0.816	519	0.132	855	0.103	1022	—	0.024
175	E	ORM-14	1.1 x 10 ¹⁹	9.3 x 10 ¹⁹	325	1.224	556	0.153	853	0.209	—	—	0.046
104	D	ORM-11	3.3 x 10 ¹⁹	1.2 x 10 ²⁰	819	1.783	541	0.294	884	0.390	—	—	0.108
356	A	ORM-34	3.8 x 10 ¹⁹	2.3 x 10 ²⁰	316	2.014	520	0.368	875	0.523	1021	—	0.205
359	A	ORM-37	3.9 x 10 ¹⁹	2.5 x 10 ¹⁹	320	1.722	484	0.291	873	0.452	1049	—	0.016
337	A	ORM-19	7.3 x 10 ¹⁹	3.2 x 10 ²⁰	313	2.388	540	0.263	877	0.781	1042	—	0.332
348	A	MT-138	9.8 x 10 ¹⁹	3.7 x 10 ²⁰	312	3.161	532	0.431	873	0.788	1052	—	0.369
2181	J	ORM-38	1.1 x 10 ²⁰	5.2 x 10 ²⁰	307	3.384	534	0.434	875	1.025	1033	—	0.521
2189	J	MT 2-234	1.5 x 10 ²¹	2.6 x 10 ²¹	270	7.4463	516	1.1073	874	2.1462	—	—	2.843

^aThe specimens were irradiated at reactor ambient temperatures in a water-moderated reactor.

^bThe maximum rate of recovery temperature refers to the temperature at which the maximum recovery occurs for a specific recovery stage over a temperature region. This stage is related to the melting temperature, T_m, as shown.

The $0.31 T_m$ recovery peak behaves in a manner similar to the $0.35 T_m$ peak with an average temperature of 862°C which agrees favorably with the value of 869°C assumed for this peak. For the $0.22 T_m$ peak, the assumed value of 537°C is in excellent agreement with the average observed recovery temperature for this stage of 536°C . There does appear to be a possible shift of this peak to lower temperatures at the highest fluence, but a more quantitative discussion will require further study. The recovery observed for the $0.15 T_m$ stage has been more quantitatively investigated and the corresponding temperature to this stage is 280°C . As can be seen, only a few of the higher fluence specimens come close to this value while a much higher temperature is obtained at the lower fluences. A preliminary study of this

effect was previously described⁽⁶⁾ as well as variation of the total resistivity recovery (area under the $0.15 T_m$ recovery peak) with fluence. The changes in the $0.15 T_m$ peak recovery temperature with the fast neutron fluence are shown in Figure 10.6 for all measured tungsten samples.

The initial high peak temperature required to recover this peak is believed to result from the presence of a defect trap or sink. Until a temperature of around 360°C is reached, the defect recovering at this stage is not freed from the defect trap. This trap is currently believed to be either rhenium or osmium produced by a transmutation resulting from thermal neutron (n, γ) reactions with tungsten, in addition to the impurity atoms present in the material. At intermediate and high neutron fluences, the defect sink

FIGURE 10.6. Variation of Stage III Peak Recovery Temperature as a Function of Fast Neutron Fluence

is most likely rhenium since it is directly formed from tungsten, whereas osmium forms secondarily from the resulting rhenium. As fast fluence increases, the temperature of this recovery peak shifts to a lower value of 312 °C at a fast fluence of 9.8×10^{19} neutrons/cm² and finally to 270 °C at a fast neutron fluence of 1.5×10^{21} neutrons/cm². It is tentatively believed that the peak shift in temperature is facilitated by other competing recovery mechanisms, arising from increased concentration of defects generated at the higher fast neutron fluences.⁽⁶⁾ In general, the total resistivity is seen to show evidence of saturation in a manner apparently related to the shift of the recovery peak temperature. The reasons for this saturation are quite complex and further investigation is necessary before a detailed explanation can be given. The saturation effect undoubtedly arises in this complex system from a number of competing recovery mechanisms. A few of the most likely are:

- Annihilation of neighboring defects of opposite sign (Frenkel pairs)
- Annihilation of distant defects of opposite sign after migration
- Migration of point defects to clusters of the same or opposite sign
- Freeing of point defects from point-defect traps or sinks such as impurities, grain boundaries, dislocations, etc.
- General increase in the fast fluence to thermal fluence ratio, indicating a larger defect-to-defect trap ratio with increasing fast fluence.

Many of these mechanisms will be affected by the magnitude of the neutron fluence since relative concentration of the various defects and some of the traps involved are similarly affected by this fluence. As indicated by Figure 10.4, the residual resistivity increment (that fraction which does not apparently anneal out), increases with the fast, or related thermal, neutron fluence. This is believed due to the increased concentration of rhenium (and osmium) produced by the higher thermal fluences. Residual resistivity as well as thermal neutron fluence can be used to calculate the rhenium concentration from previously established relations^(7,8) with the results shown in Table 10.4. Comparison between these two different measurements of the atom fraction of rhenium is good considering:

- Relative errors in thermal neutron fluence measurements
- Small residual resistivities involved for many of the specimens
- The assumption that the resistivity increases uniformly with increasing rhenium content from 0.00 to 0.04 atom fraction
- The fact that the concentration of osmium is not significant below a thermal fluence of about 10^{21} neutrons/cm². The resistivity measurement is believed to give more accurate values than those calculated from thermal fluence data.

In addition to the "expected" recovery peaks given in Table 10.3, several other recovery peaks appeared at various fluences. These peaks appeared in the following samples at

TABLE 10.4. Atom Fractions of Rhenium Obtained from Residual Resistivity and Thermal Fluence Measurements

Specimen No.	Experimental		Calculated	
	Residual Resistivity, micro-ohm-cm	Atom Fraction Re	Thermal Fluence, nvt	Atom Fraction Re
175	0.0459	0.0007	9.3×10^{19}	0.0009
152	0.0961	0.0014	1.1×10^{20}	0.0012
104	0.1082	0.0016	1.2×10^{20}	0.0013
356	0.2048	0.0031	2.3×10^{20}	0.0024
337	0.3321	0.0049	3.2×10^{20}	0.0033
358	0.3445	0.0051	4.9×10^{20}	0.0051
348	0.3692	0.0055	3.7×10^{20}	0.0039
2181	0.5210	0.0074	5.2×10^{20}	0.0054
357	0.6357	0.0094	7×10^{20}	0.0075
2189	2.8428	0.0420	2.6×10^{21}	0.0250

the reported temperatures: Sample 337 at 685 °C and 493 °C; Sample 348 at 686 °C and 473 °C; Sample 2181 at 698 °C and 470 °C; and Sample 2189 at 663 °C. The recovery occurring at these two different temperature regions is not understood at this time; however, it appears that the recovery at around 480 °C is related to that at $0.22 T_m$ as these two peaks appear to coalesce in Sample 2189 giving a resultant recovery at about 516 °C.

The dependence of total resistivity increment ($\Delta\rho$), which is a measure of the total radiation effects, on fast neutron fluence for tungsten is given in Figure 10.7. The log of resistivity is seen to increase linearly with the log of fast fluence at low fluence levels. The slope obtained for this straight line is about 0.70. At around 10^{20} neutrons/cm² the line shows considerable bending which is evidence of saturation (although it is still in doubt as to what saturation really is). Two possible causes of saturation are:

- A greater degree of local point defect annealing at higher fluence

levels (defect density by thermal spikes)

- Formation of clusters from point defects as well as formation of fewer large clusters from many small clusters.

It was stated above that many of the tungsten specimens did not show complete recovery of the radiation-induced resistivity. Specimens irradiated at low fluence levels were found to completely recover the induced resistivity; however, as the fluence increased, residual resistivity after high-temperature anneals also increased. The total resistivity recovery found for the various stages ($0.15 T_m$, $0.22 T_m$, and $0.31 T_m$) as well as the observed residual resistivities are shown as a function of fast neutron fluence in Figure 10.8. Although all the recovery stages show some saturation effects, residual resistivity continues to increase.

As previously noted, thermal neutrons can bring about a transmutation of ^{187}W to ^{188}W by the thermal neutron

FIGURE 10.7. Fractional Increase in Resistivity of Recrystallized Tungsten as a Function of Fast Neutron Fluence

FIGURE 10.8. Total Radiation-Induced Resistivity, $\Delta\rho_r$, $0.15T_m$, $0.22T_m$, $0.31T_m$ and Residual Increments as a Function of Fast Neutron Fluence

(n, γ) reaction.⁽⁹⁾ The ^{188}W then decays to ^{188}Re by β decay. There are other sources of transmutations to rhenium and possibly to osmium.

Quantitative measurements of residual resistivity are presented versus measured thermal neutron fluence in Figure 10.9. The error in the residual resistivity measurement is shown by the vertical line through the respective point. This thermal neutron fluence dependence demonstrates the effect of transmutation products.

Transmutation Effects

To resolve the effects of tungsten transmutation reactions, further study was initiated which investigated the resistivity recovery of two tungsten specimens irradiated at about the same fast neutron fluence but different

thermal neutron fluences. One specimen was in a cadmium shield to screen out some of the thermal neutrons. The unshielded specimen (ORM-34) was irradiated to a fast neutron fluence of 3.8×10^{19} neutrons/cm² and a thermal neutron fluence of 2.3×10^{20} neutrons/cm². The shielded specimen (ORM-37) was irradiated to a fast fluence of 3.9×10^{19} neutrons/cm² and an effective thermal neutron fluence of only 2.5×10^{19} neutrons/cm² due to the cadmium shield. The observed radiation-induced fractional resistivity ($\Delta\rho/\rho_0$) of the nonshielded specimen was 4.34135, while that for the shielded specimen was about 20% smaller--3.45910. After completion of the anneals, the residual resistivity fractions differed by only about 6%.

FIGURE 10.9. Average Residual Resistivity of Recrystallized Tungsten as a Function of Thermal Neutron Fluence

Results of this study are shown in Figure 10.10 in which the resistivity recovery is plotted versus annealing temperature for the two specimens; the difference between resistivity recovery of the two is plotted in Figure 10.11. The cadmium shielded specimen recovers almost all its resistivity, whereas

the nonshielded specimen recovers only about 94% of its resistivity. The actual contribution of transmutation products (foreign atoms) to the resistivity amounts to only about 6% of the resistivity measured. However, the effect of these foreign atoms as defect traps is about twice this value

FIGURE 10.10. Isochronal Resistivity Recovery for Neutron-Irradiated, Cadmium-Shielded and Unshielded, Recrystallized Tungsten as a Function of Annealing Temperature

FIGURE 10.11. Difference Between the Isochronal Resistivity Recovery of Neutron-Irradiated, Unshielded, Recrystallized Tungsten as a Function of Annealing Temperature

as shown by the initial resistivity difference of about 20%. This defect trapping ability is also evident from Table 10.5 which shows that the area under all the peaks considered is greater for the unshielded specimen than for the shielded specimen. Thus, the transmutation-produced foreign atoms apparently alter the recovery of many defects during irradiation. This trapping ability of the rhenium atom is believed to be a major cause of higher saturation fluence in tungsten.

W-25Re

Resistivity studies, similar to those described above for tungsten, were initiated to gain better understanding of the contribution of radiation-induced point defects to the mechanical properties of irradiated W-25Re. This study was also undertaken to provide further information on the effects of rhenium on defect recovery in tungsten as well as the effects of any subsequent tungsten

TABLE 10.5. Comparison of the Area and Temperature of Recovery Peaks for Cadmium-Shielded and Unshielded Tungsten Specimens

Specimen Condition	Stage	Temperature of Recovery Peak, °C	Area, micro-ohm-cm ^a
<u>Unshielded</u>	0.15 T _m	316	2.3319
	0.22 T _m	521	0.3929
	0.31 T _m	875	0.5360
	0.35 T _m	1021	—
<u>Shielded</u>	0.15 T _m	320	1.7223
	0.22 T _m	484	0.2910
	0.31 T _m	873	0.452
	0.35 T _m	1049	—

^aThe area under the recovery peak represents the total resistivity recovery for that particular defect recovery stage.

or rhenium transmutation reactions resulting from thermal neutron irradiation. The results are presented in Figure 10.12 and Table 10.6.

The recovery peaks presented in Table 10.6 refer only to related (by the previously defined melting or solidus) temperature fractions. In this analysis, a solidus temperature (T_s) of 3000 °C was used for the W-25Re. The theoretical temperature expected for the 0.15 T_s , 0.22 T_s , 0.31 T_s , and 0.35 T_s recoveries are 218, 447, 742, and 873 °C, respectively. Both the residual resistivity and the increase of the low-temperature recovery peaks seem to indicate that the specimen irradiated at the higher fluence is exhibiting a considerable transmutation effect. This effect could presumably arise from additional rhenium formation; however, because of its definite difference from the other specimens and the already high rhenium

concentration, it appears to support increased osmium formation and its resulting defect trapping effects. Calculations have been made which yield the relative concentration of tungsten, rhenium, and osmium as a function of thermal neutron fluence in W-25Re.⁽¹⁰⁾ The osmium concentration increases from about 0.001 atom fraction to almost 0.01 atom fraction over the range of thermal neutron fluence of this study. This 0.01 atom fraction of osmium would be expected to represent a sufficient concentration to give a detectable contribution to the resistivity. Since W-25Re alloy could have an ordered W_3Re arrangement of atoms, this large increase in the apparent residual resistivity could be due to radiation-induced atom disorder. It is possible that the slightly higher temperature of 212 °C for the 0.15 T_m peak observed in specimen 764, compared with the 202 °C temperature observed

in specimen 700, represents an early indication of the growing importance in the effect of increased osmium formation.

MOLYBDENUM

The recovery by annealing of defects produced by neutron irradiation of polycrystalline molybdenum specimens has been observed in a study of isochronal recovery of the radiation-induced resistivity increment. The various samples used and the pertinent irradiation conditions are shown in Table 10.6. The recovery of the radiation-induced resistivity increment for two of these specimens is presented in Figure 10.13. The recovery peaks are given in Figure 10.14.

The temperature of the $0.15 T_m$ recovery peak is relatively constant over this neutron fluence region and agrees well with the resistivity studies by Peacock and Johnson,⁽¹¹⁾ which were performed at much lower neutron fluences with a peak temperature of about 160 °C. This peak is also in good agreement with earlier studies by Kinchin and Thompson⁽¹²⁾ of irradiation damage and recovery in molybdenum by resistivity and stored energy measurements. A temperature of about 150 °C was obtained for this recovery stage.

The $0.31 T_m$ and $0.35 T_m$ recovery peaks also remain relatively constant and are quite close to the expected values of 624 and 740 °C, respectively.

FIGURE 10.12. Isochronal Resistivity Recovery of Neutron-Irradiated, Recrystallized W-25Re as a Function of Annealing Temperature

TABLE 10.6. Defect Recovery Stages in Neutron Irradiated W-25Re, Mo, and Mo-TZM, Determined from Isochronal Resistivity Studies

Sample No.	Rod	Capsule	Irradiation Conditions ^a		Maximum Rate of Recovery			
			Fluence, nvt		Temperature, °C ^b			
			Fast ($E_n \geq 1$ Mev)	Thermal	0.15 T_m	0.22 T_m	0.31 T_m	0.35 T_m
W - 25Re^c								
700	A	ORM-14	1.1×10^{19}	9.3×10^{19}	202	—	787	—
764	C	ORM-18	2.3×10^{19}	1.1×10^{20}	212	414	788	900
765	C	ORM-19	7.3×10^{19}	3.2×10^{20}	—	450	776	880
Molybdenum								
520	C	ORM-14	1.1×10^{19}	9.3×10^{19}	153	377	655	—
527	C	ORM-18	2.3×10^{19}	1.1×10^{20}	162	334	620	732
909	C	MT-138	9.8×10^{19}	3.7×10^{20}	162	294	619	743
923	C	ORM-38	1.1×10^{20}	5.2×10^{20}	155	280	617	738
Mo-TZM								
966	S	ORM-18	2.3×10^{19}	1.1×10^{20}	169	398 (284) ^d	615	773
983	S	MT-138	9.8×10^{19}	3.7×10^{20}	160	(267) ^d	617	779

^aThe specimens were irradiated at reactor ambient temperatures in a water-moderated reactor.

^bThe maximum rate of recovery temperature refers to the temperature at which the maximum recovery occurs for a specific recovery stage over a temperature region. This stage is related to the melting or solidus temperature, T_m or T_s , as shown.

^cTemperature fractions shown are based on solidus temperature (3000°C).

^dBelieved to be related to the 0.22 T_m peak. See text.

However, the 0.22 T_m (364 °C) recovery peak appears to shift to lower temperatures, possibly as a result of saturation effects. Several other recovery peaks are also evident in Figure 10.14. One peak occurs in the range of 250 to about 290 °C and agrees with a peak reported by Peacock and Johnson⁽²¹⁾ at around 280 °C. There is also an additional peak at about 430 to 460 °C which has not been previously observed. These two peaks are believed at the present to arise as substages of the previously described recovery stages.

Kissinger, et al.,⁽¹³⁾ indicated that interstitial point defects saturate in molybdenum at a fast neutron fluence of approximately 10^{19} neutrons/cm² and above this, they

either form clusters or are annihilated at vacancies. The vacancies, however, seem to remain isolated and do not give evidence of saturation until a neutron fluence of about 10^{20} neutrons/cm² is reached. These limited studies by resistivity of the effects of irradiation on molybdenum appear at first to be conflicting. If the interstitials, considered to migrate at about 160 °C at high fluences, did saturate, this peak might then be expected to shift to a lower temperature as a first approximation. However, the temperature for this peak does not change. The lattice parameter measurements⁽¹³⁾ indicate that the interstitial recovery goes through a maximum at about a fast fluence of 10^{19} neutrons/cm² and then decreases to a

FIGURE 10.13. Isochronal Resistivity Recovery for Neutron-Irradiated, Recrystallized Molybdenum as a Function of Annealing Temperature

value below the preirradiation value. This is interpreted to support the migration of interstitials to form large clusters. The combination of these two facts seems to indicate that the interstitials, at least in the range of neutron fluence of 10^{18} to 10^{20} neutrons/cm² do not saturate but either migrate to vacancies or, more importantly, migrate to form inter-

stitial clusters over this total range of fluence. Increasing the fluence from 10^{18} to 10^{19} neutrons/cm² increases the number of interstitials capable of migrating at about 160 °C to form clusters, but does not appear to make it easier for clusters to form. Therefore, an interstitial saturation condition apparently does not arise in this fluence region.

FIGURE 10.14. Derivative of the Normalized Isochronal Resistivity Recovery of Neutron-Irradiated, Recrystallized Molybdenum as a Function of Annealing Temperature

As shown in Figure 10.13, some small residual resistivity is evident in molybdenum at high fast fluence levels. Although this is not well understood at the present, two possible explanations are: (1) some type of a transmutation reaction is occurring, but in a very minor manner as compared to tungsten; and (2) some higher temperature defect is occurring which is observable only at high concentrations (or high fast fluence levels).

The occurrence of saturation of radiation-induced defects has been described, with regard to specific defects, in numerous instances. However, measurements of the saturation by re-

sistance studies of the total damage produced have been very limited. This fact is shown by the linearity of the plots of the logarithm of the total resistivity change (or its normalized counterpart) versus the logarithm of the fast or epithermal radiation fluence obtained in most experiments. This observation of saturation of the defects has been previously shown for tungsten but is more clearly demonstrated by molybdenum in Figure 10.15. In molybdenum, this saturation is seen to occur at a fast neutron fluence of about 10^{20} neutrons/cm². The resistivity change then decreases with increasing fluence. The tungsten

FIGURE 10.15. Fractional Increase in Resistivity of Recrystallized Molybdenum as a Function of Fast Neutron Fluence

specimens appear to reach saturation at about 10^{21} neutrons/cm², but studies of higher fluences will be necessary to bear out this observation. The slope obtained for the linear portion of this plot is 0.58 which is in very good agreement with a slope of 0.54 (at lower fast neutron fluences) found by Peacock and Johnson.⁽¹¹⁾

MOTZM (Mo-0.5 Ti-0.08 Zr)

To further understand the effect of solute atoms on the isochronal recovery of defects in molybdenum and to generate defect recovery spectra of a class of structural alloys, two specimens of Mo-TZM were investigated after fast neutron irradiation to respective fluences of 2.3×10^{19} and 9.8×10^{19} neutrons/cm². The isochronal resistivity recovery increment is plotted versus the annealing temperature in Figure 10.16, and the derivation of the normalized resistivity recovery increment is given in Figure 10.17.

There appears to be a spreading and merging of the recovery peaks at the higher fluence. The recovery temperatures are comparable to appropriate fractional recovery temperatures found for molybdenum. The Mo-TZM specimen irradiated at 2.3×10^{19} neutrons/cm² shows two additional peaks at 529 and 920 °C which are not yet understood. With the exception of the 160 °C peak, all the recovery peaks are smaller for molybdenum than for Mo-TZM. Evidently, the solute atoms (titanium and zirconium) are more capable of trapping the defects recovering at higher temperatures than those at 160 °C; in fact, it appears that fewer defects recover in the Mo-TZM at 160 °C. The increased recovery at higher temperature for the Mo-TZM could be due to the freeing of the additional low-temperature (160 °C) point defects from foreign atom traps at higher temperatures.

FIGURE 10.16. Isochronal Resistivity Recovery of Neutron-Irradiated, Recrystallized Mo-TZM as a Function of Annealing Temperature

FIGURE 10.17. Derivative of the Normalized Isochronal Resistivity Recovery of Neutron-Irradiated Mo-TZM as a Function of Annealing Temperature

STRUCTURE STUDIES

R. C. Rau and R. L. Ladd

Structural studies using transmission electron microscopy are being conducted on neutron-irradiated tungsten, tensile and creep rupture specimens to supplement the physical and mechanical property data obtained. The objective of these studies is to aid in the interpretation of the observed irradiation effects and to elucidate the basic mechanism of radiation damage in refractory metals.

Three groups of irradiated polycrystalline tungsten specimens were studied by transmission electron microscopy. These include: Group I specimens, which were irradiated to 1.6×10^{19} neutrons/cm² ($E_n \geq 1$ MeV) and annealed at various temperatures prior to creep rupture testing at 900 °C; Group II specimens, irradiated to various neutron fluences and tensile tested at 400 °C; and Group III specimens, irradiated to various neutron fluences and creep rupture tested at 1100 °C. All irradiations were carried out at reactor ambient temperature (~70 °C).

Examinations were made in a JEM-6A electron microscope operating at 100 KV and equipped with a tilting and rotating stage. To minimize contamination rate and thereby increase the useful lifetime of a thin foil in the intense electron beam, a liquid-nitrogen cooling device was used in the specimen chamber. For all specimens, thin foils were studied from both the unstressed button head and from the reduced section near the fracture.

Group I Specimens

Although both irradiation and subsequent annealing treatments produced changes in the mechanical properties of Group I specimens, no significant changes were evident by electron microscopy. These specimens are listed in Table 10.7. The microstructures of the as-irradiated as well as the various irradiated and annealed specimens remained similar to the unirradiated microstructures.⁽¹⁴⁾ However, this series of samples provided vivid evidence of the effect of stress on the microstructure.

Examination of the button heads of these and other specimens showed that the microstructure initially consisted of large tungsten grains divided into many small subgrains by elaborate geometric dislocation networks. As shown in Figure 10.18a, several subgrain boundary dislocation networks were inclined at various angles to the foil surface and terminated on a grain boundary running diagonally across the photograph. Strain during testing causes a degradation of the dislocation networks into randomly tangled arrays as shown in Figure 10.18b. This photomicrograph shows dislocation tangles typical of the reduced sections of tested specimens.

Group II Specimens

The second group of specimens was irradiated to fast neutron fluences ranging from 4.0×10^{18} to 1.2×10^{20} neutrons/cm² and tensile tested at 400 °C ($0.18 T_m$). During testing, the specimens were held at this temperature for about 1 hr.

This series of specimens, listed in Table 10.8, presented dramatic evidence of the buildup of defect clusters with increasing neutron fluence. The buildup is illustrated in Figures 10.19 and 10.20, which respectively show button heads and reduced sections. Figures 10.19 and 10.20 show typical unirradiated microstructures consisting of dislocations networks (Figure 10.19) and tangles (Figure 10.20) superimposed on a structureless matrix.

Specimens from the two lowest fluence levels, 4.0 and 5.9×10^{18} neutrons/cm², showed microstructures basically similar to the unirradiated

specimens; however, a population of very fine black dots was present in the matrix of the button heads. Figures 10.19 and 10.20 show typical low fluence specimens. The tiny black dots present in the button heads were estimated to be approximately 25 to 50 Å in average size and were present in concentrations of about 1.4×10^{16} /cm³.

The four specimens from this group irradiated to fluence levels of 3.8×10^{19} neutrons/cm² or greater showed appreciable buildup of defect clusters throughout the matrix. As shown in Figures 10.19 and 10.20, both button

TABLE 10.7. Postirradiation Annealed Tungsten Specimens for Electron Microscopy Studies

Specimen ^a	Capsule ^b	Annealing ^c Temperature, °C	Creep Rate, sec ⁻¹	Rupture Life, hr	Elongation in 2.54 cm, %	Reduction in area, %
1265	Control	—	2.32×10^{-6}	12.45	36.2	96.6
1268	ORM-17	as-irradiated	5.46×10^{-7}	47.48	26.8	96.5
1263	ORM-17	1050	5.08×10^{-7}	77.04	32.5	96.8
1271	ORM-17	1200	1.96×10^{-7}	124.60	30.0	96.6
1266	ORM-17	1400	1.01×10^{-6}	32.20	35.3	93.7
1267	ORM-17	1700	1.26×10^{-6}	24.12	33.9	94.8

^aCreep-rupture tested at 900°C and 22.64 kg/mm².

^bIrradiated at reactor ambient temperature (~70°C) to 1.6×10^{19} nvt ($E_n \geq 1$ Mev).

^cPost-irradiation annealed 1 hour in H₂ atmosphere.

TABLE 10.8. Tungsten Tensile Specimens Tested at 400 °C

Specimen	Capsule	Fluence, ^a nvt		Strength, kg/mm ²		Elongation, %
		Thermal	Fast ($E_n \geq 1$ Mev)	Yield	Ultimate	
1332	Control	—	—	21.9	39.8	45.6
1325	ORM-31	3.9×10^{19}	4.0×10^{18}	32.4	42.4	39.6
1329	ORM-33	4.1×10^{19}	5.9×10^{18}	35.9	39.8	37.2
1331	ORM-34	2.3×10^{20}	3.8×10^{19}	58.5	58.5	4.4
1343	ORM-37	2.5×10^{19}	3.9×10^{19}	54.8	54.8	0.3
1340	ORM-36	4.9×10^{20}	7.9×10^{19}	72.0	72.0	Nil
1334	ORM-35	7.0×10^{20}	1.4×10^{20}	82.7	82.7	Nil

^aIrradiated at reactor ambient temperature (~70°C).

^bSpecimen shielded with 0.63-mm-thick cadmium.

Neg J-1233

a. Unstressed Button Head Area

Neg J-1238

b. Strained Area Near Fracture

FIGURE 10.18. Transmission Electron Micrographs of Unirradiated Tungsten

Neg J-1552

a. Unirradiated

Neg J-1527

b. Irradiated to 5.9×10^{18} nvt
($E_n \geq 1$ MeV)

Neg J-1449

c. Irradiated to 1.2×10^{20} nvt
($E_n \geq 1$ MeV)

FIGURE 10.19. Transmission Electron Micrographs of Button Heads of Tungsten Specimens Tensile Tested at 400 °C

Neg J-1588

a. Unirradiated

Neg J-1605

b. Irradiated to 5.9×10^{18} nvt
($E_n \geq 1$ MeV)

Neg J-1659

c. Irradiated to 1.2×10^{20} nvt
($E_n \geq 1$ MeV)

FIGURE 10.20. Transmission Electron Micrographs of Reduced Areas Near Fracture of Tungsten Specimens Tensile Tested at 400 °C

heads and reduced sections contained large numbers of enlarged dot clusters, probably about 100 Å in diameter, and also occasionally contained resolvable loops of 200 to 300 Å in diameter. This coarsening of the clusters was more noticeable the higher the neutron fluence. Cluster densities for all four specimens were estimated to be approximately 2 to $3 \times 10^{16}/\text{cm}^3$, but these values are probably inaccurate and represent apparent saturation values arising from counting errors introduced by image overlap.⁽¹⁵⁾ The observation of broadening of diffraction spots and Kikuchi lines in the electron diffraction patterns as fluence level increased indicates that cluster densities probably also continued to increase.

As indicated in Table 10.8, this group of samples contained a pair which received nearly identical fast neutron fluences (3.8 and 3.9×10^{19} neutrons/cm²), but which received thermal fluences differing by an order of magnitude. Although the decreased thermal fluence in the cadmium-shielded specimen resulted in greater embrittlement, no microstructural changes could be seen in the electron microscope. The photomicrographs in Figure 10.21 illustrate the similarity of the two specimens.

In addition to the foregoing effects, the subgrain boundary dislocation networks were much less evident in the higher fluence specimens. However, this effect is ascribed to overlap by the large number of cluster images rather than to an actual depletion of the networks, since tilting showed approximately the same subgrain dis-

tribution as in unirradiated or low fluence samples. No evidence of denudation of clusters near subgrain or grain boundaries was seen.

An initially surprising result of this study was the lack of observation of cleared pathways or channels through the defect clusters in specimens from the reduced areas near the fracture. Such channeling has been observed in molybdenum^(16,17) and occurs when glide dislocations move through the lattice and sweep up obstacles. However, the present lack can be explained by assuming that the high fluence specimens were apparently hardened by their high cluster content, and therefore failed by brittle fracture with little or no dislocation movement. This conclusion is substantiated by the two photomicrographs in Figure 10.21. Both of these show segments of dislocation networks which are unusually well preserved in comparison with the usual tangled arrays present after stressing.

Comparison of clustering in the present study with earlier work⁽¹⁸⁾ indicates that heating irradiated tungsten to 400 °C ($0.18 T_m$) produces observable effects in the microstructure. In particular, the presence of dot clusters in the lower dose specimens and loops in the higher dose specimens can be ascribed to this heating. Although the nature of the clusters and loops was not determined, comparison with work on neutron irradiated molybdenum⁽¹⁹⁻²¹⁾ suggests that they are probably interstitial. Future work will be directed toward an identification in tungsten.

Neg J-1617

- a. Unshielded; Fast Fluence 3.8×10^{19} nvt;
Thermal Fluence 2.3×10^{20} nvt

Neg J-1637

- b. Shielded with 0.51 mm Cadmium; Fast Fluence 3.9×10^{19} nvt;
Thermal Fluence 2.5×10^{20} nvt

FIGURE 10.21. Transmission Electron Micrographs of Reduced Sections of Shielded and Unshielded Irradiated Tungsten Specimens Tensile Tested at 400 °C

Group III Specimens

The specimens in Group III were irradiated to fast ($E \geq 1$ MeV) neutron fluences ranging from 8.5×10^{17} to 1.5×10^{21} neutrons/cm² and then creep rupture tested at 1100 °C ($0.37 T_m$). Data for this group are present in Table 10.9. Because of the heating during testing, this series of specimens basically illustrates the effects of high temperature annealing on the radiation produced microstructure. However, the results are probably also related to heating time since each specimen was held at the test temperature for a different period of time, depending upon its rupture life.

Typical microstructures are shown in Figures 10.22 and 10.23, which show button heads and reduced sections, respectively. Unirradiated and low fluence level (5.9×10^{18} neutrons/cm² and below) specimens had similar microstructures, consisting of dislocation networks in the button heads (Figure 10.22a) and tangles in the reduced sections (Figure 10.23a). No evidence was seen of dot clusters in the matrix as occurred in the button heads of the 400 °C tensile specimens.

Specimens irradiated to fluences of 3.8×10^{19} neutrons/cm² or greater prior to testing showed clusters and resolvable dislocation loops in the matrix, but in quantities considerably below those of the 400 °C tensile specimens irradiated to similar fluences. Button heads contained appreciably greater numbers of the defects than the reduced sections, as can be seen by comparing Figures 10.22b and 10.22c with Figures 10.23b and 10.23c. Defect concentrations in the button heads of the specimens irradi-

ated in the 10^{19} and 10^{20} neutrons/cm² fluence range were estimated to be approximately 10^{14} /cm³. The reduced sections and button head of the 10^{21} sample contained concentrations at least an order of magnitude lower.

The average size of the defects increased with neutron fluence. In the specimens irradiated to 3.8×10^{19} neutrons/cm², most of the defects appeared as dot clusters approximately 100 Å in diameter, with some resolvable loops approaching 300 Å diameter. In the specimen irradiated to 1.2×10^{20} neutrons/cm², most of the defects appeared as loops averaging 200 Å in diameter, with large loops approaching 500 Å. Finally, in the highest fluence specimen, 1.5×10^{21} neutrons/cm², the average and maximum loop sizes were 1000 Å and 2000 Å, respectively.

Like the previous group, this group of specimens also contained a pair of cadmium-shielded/unshielded specimens which received similar fast neutron fluences but widely differing thermal fluences. Electron microscopy again failed to show any significant changes in microstructure, although the creep properties were different.

A noteworthy observation for all of the Group III samples was the rather high degree of retention of some relatively intact dislocation subgrain boundary networks in the reduced sections. Some of these retained networks appear to contain higher dislocation densities than networks in the button heads (see Figure 10.24). This is indicative of a rearrangement of the subgrains into orientations of greater lattice mismatch during high temperature testing.

TABLE 10.9. Tungsten Creep-Rupture Specimens Tested at 1100 °C and 18.28 kg/mm²

Specimen	Capsule	Fluence, ^a nvt		Creep Rate, sec ⁻¹	Rupture Life, hr	Elongation, %	Reduction, %
		Thermal	Fast (E _n ≥ 1 Mev)				
1391	Control	—	—	6.39 x 10 ⁻⁶	6.23	30.3	98.7
1384	ORM-32	6.8 x 10 ¹⁸	8.5 x 10 ¹⁷	6.26 x 10 ⁻⁶	6.87	33.9	95.9
1383	ORM-31	3.9 x 10 ¹⁹	4.0 x 10 ¹⁸	3.60 x 10 ⁻⁶	12.61	28.0	90.0
1385	ORM-33	4.1 x 10 ¹⁹	5.9 x 10 ¹⁸	3.00 x 10 ⁻⁶	15.92	32.2	95.1
1395	ORM-34	2.3 x 10 ²⁰	3.8 x 10 ¹⁹	1.08 x 10 ⁻⁶	41.92	26.3	92.3
1394 ^b	ORM-37	2.5 x 10 ¹⁹	3.9 x 10 ¹⁹	3.58 x 10 ⁻⁶	14.64	28.0	90.0
1388	ORM-35	7.0 x 10 ²⁰	1.4 x 10 ²⁰	2.93 x 10 ⁻⁷	194.9	33.5	92.4
1403	GEFP2-234	2.7 x 10 ²¹	1.5 x 10 ²¹	1.08 x 10 ⁻⁸	315.41	6.6	32.5

^aIrradiated at reactor ambient temperature (~ 70°C).

^bShielded with 0.51-mm cadmium.

The observations of annealing of radiation-induced defects in these specimens are in good agreement with previous results on irradiated, single crystal tungsten annealed at 1090 °C.⁽¹⁸⁾ The lower loop densities in the reduced sections in comparison with the unstressed button heads can probably be ascribed to slightly higher temperatures in the gage lengths during creep rupture or tensile tests, or to interaction of the defects with moving dislocations during reorientation of the subgrains. Although diffraction contrast analysis of the large loops found in the present study has not yet been carried out, their similarity to large interstitial loops formed on annealing irradiated molybdenum^(19,21) suggests that these loops may be interstitial. Tilting experiments are planned to determine the nature and crystallography of loops in tungsten.

HEAT RESISTANT METALS AND ALLOYS -

J. Moteff and J. P. Smith

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

Materials to be investigated in this program include A-286, Hastelloy X, Hastelloy N (INOR-8), Hastelloy R-235, Fe-Cr-Al-Y alloys, AISI 304 SS, ASTM-A302B and A350-LF3 pressure vessel steels, and various Inconel and Incoloy alloys.

Transmission Microscopy (D. A. Woodford,* J. P. Smith, J. Moteff)

Transmission electron microscopy work initiated last year was extended

* General Electric Co. Power Generation Division, Materials and Processes Laboratory.

Neg J-1700

- a. Irradiated to 4.0×10^{18} nvt
($E_n \geq 1$ MeV)

Neg J-1766

- b. Irradiated to 1.2×10^{20} nvt
($E_n \geq 1$ MeV)

Neg J-1794

- c. Irradiated to 1.5×10^{21} nvt
($E_n \geq 1$ MeV)

FIGURE 10.22. Transmission Electron Micrographs of Button Heads of Tungsten Specimens Creep-Rupture Tested at 1100 °C

Neg J-1836

- a. Irradiated to 4.0×10^{18} nvt
($E_n \geq 1$ MeV)

Neg J-1906

- b. Irradiated to 1.2×10^{20} nvt
($E_n \geq 1$ MeV)

Neg J-1921

- c. Irradiated to 1.5×10^{21} nvt
($E_n \geq 1$ MeV)

FIGURE 10.23. Transmission Electron Micrographs of Reduced Areas Near Fracture of Tungsten Specimens Creep-Rupture Tested at 1100 °C

Neg J-1826

- a. Irradiated to 8.5×10^{17} nvt
($E_n \geq 1$ MeV)

Neg J-1870

- b. Irradiated to 3.8×10^{19} nvt
($E_n \geq 1$ MeV)

FIGURE 10.24. Transmission Electron Micrographs Showing Retained Dislocation Networks in Reduced Sections of Tungsten Specimens Creep-Rupture Tested at 1100 °C

to more fully determine the role of helium in the above discussed A-286 specimens. Specimens cut from the unstressed buttonhead portion of irradiated samples of the low- and high-boron heat were aged at 760, 845, and 955 °C for 3 hr to grow helium bubbles large enough to be readily studied in the electron microscope. All six specimens showed the presence of well-defined helium bubbles whose average diameter increased as the annealing temperature was increased. Also, the high-boron specimen yielded a larger bubble size than the corresponding low boron specimen for a given heat treatment. The histograms in Figure 10.25 summarize these results. No bubbles were observed in unirradiated control specimens given heat treatments simultaneously with the irradiated specimens.

A study of numerous photomicrographs of the annealed specimens yielded these conclusions:

- The helium bubbles tend to form preferentially on dislocations rather than at the gamma prime (γ') particle--matrix interfaces. This is illustrated in Figures 10.26a and b. Figure 10.26a shows that virtually all the bubbles lie on dislocation lines (γ' particles, being out of contrast in this picture, are not visible). Figure 10.26b shows the presence of γ' strain fields as well as dislocations (the γ' itself is too small to be visible). Note that, although some small bubbles are present at the γ' strain fields, most of them (and virtually all the largest ones) lie on the dislocation lines.

- The preference of dislocation sites for bubble formation is not surprising since the effective cross section exposed to a recoiling helium atom traveling through the matrix is much larger for a dislocation line than for a γ' particle. Also, a helium atom has a much better diffusion path when attached to a dislocation line. Figure 10.26b confirms this condition, showing that, in general, the bubbles associated with γ' particles are much smaller than the bubbles on the dislocations.
- Another significant conclusion is that the helium does not tend to migrate to the grain boundaries, possibly because the elevated-temperature heat treatment was performed on unstressed samples. Heat treatment of samples from stressed specimens is currently underway. This is illustrated in Figure 10.26c, which shows a high density of bubbles near the boundary but only a few on the boundary itself. Note again that all the bubbles are associated with dislocation lines.

The results discussed above tend to indicate that radiation-induced embrittlement may be caused by bubbles pinning dislocation, especially in areas adjacent to grain boundaries. This prevents the dislocations from moving to the boundaries--a necessary condition for grain boundary sliding.

Another interesting observation was made on the high-boron specimen annealed at 955 °C, illustrated in Figure 10.27, which shows an impurity particle (TiC) surrounded by a "shell" of bubbles. The radius of this shell

FIGURE 10.25. Variation of Helium Bubble Diameter with Boron Concentration and Postirradiation Annealing Temperature in Unstressed Irradiated A-286

a. Bubbles on Dislocations

b. Bubbles on Dislocations and Gamma Prime Particles

c. Bubbles on Dislocations and Grain Boundaries

FIGURE 10.26. Transmission Electron Micrographs Showing Precipitation of Helium Bubbles in Irradiated A-286

FIGURE 10.27. Shell of Helium Bubbles Surrounding an Impurity Particle in Irradiated A-286. (Shell radius ~ 2.5 microns)

and of others that have been observed is approximately 2 to 3 μ . The normal range of an α particle in an iron matrix is approximately 2 μ . It appears that a fair amount of boron may be soluble in the TiC particles or at the particle-matrix interface, and that these particles may be surrounded by a sphere of helium atoms.

Another possible explanation for this shell may be that a strain field exists around the impurity particle in such a manner that dislocations present in or about this field are effective sinks for mobile helium atoms.

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RADIATION EFFECTS IN FUEL CLADDING MATERIALS

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IN-REACTOR AND POSTIRRADIATION STRESS-RUPTURE PROPERTIES OF TYPE 304 STAINLESS STEEL

E. E. Bloom and J. R. Weir

The creep-rupture properties of type 304 SS have been determined in the unirradiated condition and for irradiated specimens tested both during and after irradiation. Tests were performed at 650 °C at stresses between 15,000 and 35,000 psi. The composition of the experimental alloy was as follows (by weight): 0.057% C, 0.76% Mn, 0.020% P, 0.016% S, 0.48% Si, 19.0% Cr, 8.9% Ni, and 2.2 ppm B. The final heat treatment prior to testing was 1 hr anneal at 1038 °C which developed a grain size of ASTM 6. In-reactor, stress rupture

tests were conducted at a poolside position of the Oak Ridge Research reactor. The thermal flux was 6×10^{13} neutrons $\text{cm}^{-2} \text{sec}^{-1}$ and the fast ($E > 2.9 \text{ MeV}$) flux 5×10^{12} neutrons $\text{cm}^{-2} \text{sec}^{-1}$. Postirradiation tests were performed on specimens irradiated at 650 °C to a fluence of 2×10^{20} neutrons/ cm^2 (thermal) and 2×10^{19} neutrons/ cm^2 ($E > 2.9 \text{ MeV}$).

Minimum creep rate and rupture life values for each test are listed in Table 11.1. The creep rates for both in-reactor and postirradiation tests are a factor of 2 to 5 higher than those of the unirradiated alloy. It is important to note, however, that the irradiated specimens failed at times approximately equal to the time

TABLE 11.1. Minimum Creep Rate and Rupture Life of Type 304 SS Tested at 650 °C.

Stress, psi	Minimum Creep Rate, %hr, and Rupture Life, hr ^(a)		
	Unirradiated	In-Reactor	Postirradiation
35,000	2.26 (7.7)	- -	- -
30,000	0.92 (9.0)	- -	0.50 (12.0)
27,500	0.33 (44.1)	- -	- -
25,000	0.048 (122.8)	0.138 (17.1)	0.139 (24.4)
22,250	0.042 (157.7)	- -	- -
20,750	- -	- -	0.014 (123.4)
20,000	0.0073 (366.6)	0.016 (98.0)	- -
17,000	0.0028 (1876.0)	0.0035 (423.0)	0.006 (314.4)
15,000	0.00032	0.0014	0.0018 (956.9)

(a) Rupture life is listed in parenthesis.

required for the establishment of secondary creep processes in the unirradiated alloy. Creep curves for irradiated and unirradiated specimens tested at a stress of 25,000 psi are shown in Figure 11.1. Results of optical and electron metallography indicate that the primary effect of irradiation is an enhancement of the intergranular fracture process and that irradiation has little effect upon the deformation processes occurring during creep testing. It thus appears that the higher creep rates observed for the irradiated alloy were a result of fracture of the specimen while still in the primary stage of creep.

Ductility, as measured by elongation or reduction in area, was significantly reduced for both in-reactor and post-irradiation tests. Figure 11.2 is a plot of ductility as a function of rupture life. For the stress levels used in this investigation, the loss of ductility became more severe as the stress (or strain rate) was reduced.

MECHANICAL PROPERTIES OF TYPE 304 STAINLESS STEEL CONTAINING CYCLOTRON-INJECTED HELIUM

R. T. King and J. R. Weir

In a previous report⁽¹⁾ it was demonstrated that the presence of 0.1 ppm of cyclotron-injected helium caused significant reduction of the postbombardment tensile ductility of 304 SS. The loss of ductility occurred in samples tested at 700, 800, and 900 °C at a strain rate of 0.026 min^{-1} .

Samples of the same material were prepared from 50% cold rolled 0.010 in. thick sheet and annealed for 1 hr in argon at 925 and 1038 °C. They were

bombarded in a scanning device to contain 1 ppm of helium, calculated from the measured beam current and known α -particle losses. These samples and control samples of the same material that were not bombarded have been tested in tension at 0.026 min^{-1} at 500, 600, 700, 800, and 900 °C. The results of these tests are given in Table 11.2. Because no significant changes in yield stress, engineering ultimate stress, or uniform elongation was observed, only the total percent elongation to failure is reported.

The results indicate the 1 ppm of uniformly dispersed helium does not affect the tensile ductility of 304 SS at 500 or 600 °C. The ductility of bombarded samples is a monotonically decreasing function of test temperature range, the ductility of the control sheet samples annealed for 1 hr at 1038 °C. The ductility of 304 SS containing 1 ppm of helium is higher for samples annealed for 1 hr at 925 °C than for those annealed for 1 hr at 1038 °C. In fact, the ratios of the ductility of bombarded samples to the ductility of control samples at a given temperature are the same for either an annealing treatment to within ± 0.11 .

Further investigation of the microstructure of this material and comparisons with the results of neutron irradiations are in progress.

MECHANICAL PROPERTIES OF 0.2 wt% TITANIUM-MODIFIED TYPE 304L SS CONTAINING CYCLOTRON-INJECTED HELIUM

R. T. King and J. R. Weir

The mechanical properties of a type 304L SS modified with 0.2 wt%

Neg Y-78791

FIGURE 11.1. Creep-Rupture Curves for 304 SS Tested at 650 °C and 25,000 psi

Neg Y-78792

FIGURE 11.2. Stress-Rupture Ductility of 304 SS at 650 °C

titanium have been investigated previously⁽¹⁾ after the injection of 0.1 ppm of helium using the Oak Ridge Isochronous Cyclotron. At this level of helium content, the tensile properties of the titanium-modified steel were not affected.

Samples of this material were prepared as before, except that intermediate anneals at 1200 °C in argon were used to eliminate a second magnetic phase, presumably ferrite, from the microstructure. The material was finally cold rolled 50% to 0.010 in. thickness, samples were cut and then annealed for 1 hr at 925 and 1038 °C in argon.

Control samples and samples bombarded in the scanning device to contain 1 ppm of helium were then tensile tested at 0.026 min⁻¹ strain rate at 500, 600, 700, 800, and 900 °C. As

was the case for the 304 SS samples, the yield stress, engineering ultimate stress, and uniform elongations were not altered appreciably by the bombardment. The tensile ductilities of these samples are presented in Table 11.3.

It is significant that 1 ppm of helium, uniformly dispersed in the titanium-modified steel annealed at 925 °C, did not cause a decrease in the tensile ductility. However, 1 ppm of helium caused the ductilities of the samples which were annealed at 1038 °C to decrease. The fractional loss of ductility of the titanium-modified 304L SS is lower at all test temperatures than that of 304 SS. These observations are in agreement with earlier data from neutron irradiated materials.

There are several possible causes for the differences in behavior caused

TABLE 11.2. Ductility of Type 304 SS After Cyclotron Bombardment

Type 304 SS	Engineering Percent Elongation at Test Temperature, °C				
	500	600	700	800	900
Annealed 1 hr at 1038 °C, control	41.6	34.7	48.1	47.2	41.9
Annealed 1 hr at 1038 °C, 1 ppm helium	45.9	35.7	32.8	25.0	12.0
Annealed 1 hr at 925 °C, control	42.1	44.3	69.6	77.2	68.6
Annealed 1 hr at 925 °C, 1 ppm helium	41.6	42.3	55.3	32.8	27.7

TABLE 11.3. Ductility of 0.2 Titanium-Modified Type 304L After Cyclotron Bombardment

Material Condition	Engineering Percent Elongation at Test Temperature, °C				
	500	600	700	800	900
Annealed 1 hr at 1038 °C, control	41.1	42.7	77.3	66.3	52.6
Annealed 1 hr at 1038 °C, 1 ppm He	42.7	39.2	60.8	45.1	36.6
Annealed 1 hr at 925 °C, control	36.3	38.7	72.4	74.7	78.1
Annealed 1 hr at 925 °C, 1 ppm He	34.6	35.2	70.1	75.5	79.2

by different annealing treatments of this material, and for the increased resistance to helium damage in this material in contrast to 304 SS.⁽²⁾ Investigation of these differences is being pursued.

ELECTRON MICROSCOPY OF IRRADIATED STAINLESS STEELS

E. E. Bloom, J. O. Stiegler, and J. R. Weir

Studies are continuing to determine the location and behavior of helium in irradiated stainless steels. Modified type 304 SS containing 0.015 wt% C, 0.14 wt% Ti and 0.001 to 0.0015 wt% B has been irradiated for three cycles in the B-8 position of the ORR (1.3×10^{21} neutrons/cm² thermal, 1.0×10^{21} neutrons/cm² E > 1 MeV) at temperatures of 650 and 800 °C. The calculated helium concentration in the irradiated alloy is 15×10^{-6} atom fraction. To date we have examined samples in the as-irradiated condition for the 800 °C irradiation temperature. The irradiation temperature was above the solubility limit for carbon in the alloy and thus only a few precipitates were present. Helium bubbles as shown in Figure 11.3 were found in several isolated areas of the irradiated samples. In the areas in which bubbles were found several important features were noted:

- Bubbles, present in the grain boundary, are several times larger than those present within the matrix.
- A necklace of bubbles is present along the triple grain junction located in the lower left-hand corner of the figure. Since this

boundary extends from the top of the foil to the bottom, the light circular regions must be bubbles which lie within the foil and cannot be a polishing effect on the surface.

- Clusters of bubbles are attached to the surface of precipitates present within the matrix.
- In several instances the bubbles have a polyhedral shape which suggests that equilibrium between surface tension and internal gas pressure is being approached. This is suggested by contrast variations in the large bubbles lying in the grain boundary of Figure 11.3.

MSRE SURVEILLANCE PROGRAM - HASTELLOY N

H. E. McCoy

Several stringers of Hastelloy-N test specimens were located in an axial position near the centerline of the MSRE for surveillance purposes. These specimens were from Heats 5085 (cylindrical vessel) and 5081 (miscellaneous parts). They were removed after 4800 hr at 645 ± 10 °C, during which the reactor had operated 7612 MW-hr. The peak thermal dose was 1.3×10^{20} nvt and the peak fast dose (>1.2 MeV) was 3×10^{19} nvt. The peak-to-minimum thermal flux over the length of the test specimen array was a factor of 5, but this was not found to be a significant variable in this evaluation. The surveillance control specimens were exposed to MSRE-type fuel salt and duplicated the thermal history of the in-reactor specimens.

Neg Y-79894

24,000X

FIGURE 11.3. Type 304L SS Containing 0.14 wt% Titanium,
Irradiated at 800 °C, Containing 15×10^{-6} Atom Fraction
Helium

Samples of both reactor and control specimens were examined metallographically. No evidence of attack or deposition was found within the gage lengths of any of the samples. However, all test specimens from Heat 5085 (both irradiated and control) were found to have a surface layer on the 1/4 in. diam portion of the samples on the sides in contact or near contact with the graphite. Such layers were not found on the smaller 1/8 in. diam gage length of the specimens which were separated from the graphite by 1/16 in. Figure 11.4 shows the nature of the surface layer on these samples. Electron microprobe studies showed that the carbon content near the edge of the sample was from 0.3 to 1.2 wt% higher than the interior of the sample.

Both tensile and creep-rupture tests have been conducted on the surveillance specimens. The creep-rupture testing is not yet complete, so only the results of the tensile tests will be reported. The tensile results are given in Table 11.4.

The total elongation at fracture when deformed at a strain rate of 0.05 min^{-1} is shown as a function of temperature in Figure 11.5. Both heats show some reduction in ductility in the irradiated condition, with a greater reduction being observed for Heat 5085. At temperatures above 500°C , the ductility of the irradiated and the control material decreases with increasing temperature, with the irradiated material showing a greater loss in ductility. At temperatures above 650 to

Neg Y-78266

FIGURE 11.4. Edge of Hastelloy N (Heat 5085) Specimen Exposed to Molten Salt for 4800 hr at 650°C . Surface in Intimate Contact with Graphite. Etchant: Glyceria Regia

TABLE 11.4. Postirradiation Tensile Properties of Several Heats of MSRE Hastelloy N After Irradiation in a Helium Environment

Heat No.	Condition	Boron Level (ppm)	Experiment No.	Irradiation Temp (°C)	nvt, Thermal Dose $\times 10^{20}$	Test Temp (°C)	$\dot{\epsilon}$	Yield Stress (psi)	Ultimate Stress (psi)	Uniform Elongation (%)	Total Elongation (%)	Reduction in Area (%)	Specimen No.
5065	A.R.	20	ORR-149	43	8.5	25	0.05	102,900	135,100	31.5	35.5	52.0	570
5065	A.R.	20	ORR-149	43	8.5	200	0.05	82,300	119,600	36.1	39.2	54.8	571
5065	A.R.	20	ORR-149	43	8.5	650	0.05	44,600	76,600	26.3	27.3	24.5	572
5065	A.R.	20	ORR-149	43	8.5	650	0.002	40,800	57,400	12.2	13.1	21.9	574
5065	A.R.	20	ORR-149	43	8.5	871	0.05	33,500	36,400	1.7	1.8	3.25	573
5065	A.R.	20	ORR-149	43	8.5	871	0.002	23,200	23,400	0.9	1.8	6.07	575
5065	8 hr at 871°C	20	ORR-149	43	8.5	25	0.05	101,500	132,200	30.0	33.6	55.2	581
5065		20	ORR-149	43	8.5	200	0.05	78,600	118,300	37.3	39.8	45.9	582
5065		20	ORR-149	43	8.5	650	0.05	35,100	77,700	25.3	25.8	22.6	583
5065		20	ORR-149	43	8.5	650	0.002	38,100	66,200	13.7	14.3	17.3	585
5065		20	ORR-149	43	8.5	871	0.05	37,100	38,500	1.7	1.9	2.94	584
5065		20	ORR-149	43	8.5	871	0.002	25,900	25,900	0.8	1.6	5.26	586
5067	A.R.	20	ORR-155	500-700	1.4	25	0.05	59,000	123,700	49.4	51.2	45.4	2289
5067	A.R.	20	ORR-155	500-700	1.4	650	0.05	38,400	66,800	14.0	14.0	20.5	2290
5067	A.R.	20	ORR-155	500-700	1.4	650	0.002	36,400	59,900	9.6	9.6	16.0	2291
5085	A.R.	38	ORR-155	500-700	1.4	25	0.05	46,300	110,600	41.1	41.2	40.3	2285
5085	A.R.	38	ORR-155	500-700	1.4	650	0.05	30,800	64,400	18.0	21.2	19.1	2286
5085	A.R.	38	ORR-155	500-700	1.4	650	0.002	30,200	51,800	9.3	9.9	15.4	2287
5065	A.R.	20	ORR-155	500-700	1.4	25	0.05	50,100	117,000	54.4	56.1	53.2	1857
5065	A.R.	20	ORR-155	500-700	1.4	650	0.05	37,400	62,100	12.3	12.4	17.2	1858
5065	A.R.	20	ORR-155	500-700	1.4	650	0.002	34,800	49,100	6.4	6.5	15.3	1859
5065	A.R.	20	ETR-41-31	600 $\pm$ 100	3.5	550	0.002	49,100	68,600	9.1	9.4		1273
5065	A.R.	20	ETR-41-31	600 $\pm$ 100	3.5	600	0.002	42,200	56,300	8.2	8.5		1276
5065	A.R.	20	ETR-41-31	600 $\pm$ 100	3.5	650	0.05	41,200	59,000	10.8	11.3		1270
5065	A.R.	20	ETR-41-31	600 $\pm$ 100	3.5	650	0.002	42,600	51,800	5.9	6.1		1271
5065	A.R.	20	ETR-41-31	600 $\pm$ 100	3.5	760	0.002	41,900	46,000	2.8	2.8		1274
5065	A.R.	20	ETR-41-30	<150	5	650	0.05	46,700	76,200	21.6	22.2	28.8	383
5065	A.R.	20	ETR-41-30	<150	5	650	0.002	37,700	53,300	9.3	10.0	10.1	380
5065	A.R.	20	ETR-41-30	<150	5	650	0.002	40,000	57,500	11.4	11.6	17.5	384

11.8

BNWL-455

Neg ORNL-Dwg 67-2452

FIGURE 11.5. Comparative Tensile Ductilities of MSRE Surveillance Specimens and Their Controls at a Strain Rate of 0.05 min^{-1}

700 °C, the control material exhibits improved ductility; whereas, the ductility of the irradiated material continues to decrease.

Figure 11.6 compares the properties of the irradiated and control specimens at a lower strain rate, 0.002 min^{-1} . Qualitatively, the behavior is very similar to that noted at a strain rate of 0.05 min^{-1} . However, the loss in ductility is magnified at the lower strain rate. The variations in the properties of the two heats of material are minimal.

Figures 11.7 and 11.8 show how the ratio of the irradiated to the unirradiated tensile property varies for Heats 5081 and 5085, respectively, as

a function of temperature. For both heats, the yield strength is unchanged by irradiation. The ultimate tensile stress is reduced about 8% up to a temperature of about 500 °C where the reduction becomes considerably greater. The reduction in ductility at room temperature and at elevated temperatures is also clearly demonstrated.

We have compared the irradiated and control surveillance specimens. Let us now look at how the properties of the control specimens have changed during their 4800 hr of thermal exposure to molten salt. Table 11.5 shows representative properties of Heat 5085 after several different heat treatments. The first group of tests was run with

Neg ORNL 67-2453

FIGURE 11.6. Comparative Tensile Ductilities of MSRE Surveillance Specimens and their Controls at a Strain Rate of 0.002 min^{-1}

Neg ORNL 67-2458

FIGURE 11.7. Comparative Tensile Properties of Irradiated and Unirradiated MSRE Surveillance Specimens, Heat 5081

Neg ORNL 67-2459

FIGURE 11.8. Comparative Tensile Properties of Irradiated and Unirradiated MSRE Surveillance Specimens, Heat 5085

the material in the as-received condition (mill annealed 1 hr at 1177 °C). Annealing for 600 hr at 650 °C had no appreciable effects on the properties. The MSRE surveillance specimens were given a 2 hr anneal at 900 °C before insertion into the reactor and the properties of the material in this state are also indicated in Table 11.5. The properties at 25 °C were unchanged and slight reductions in tensile strength and ductility were noted at a test temperature of 650 °C. After exposure to salt for 4800 hr at 650 °C, there is a further reduction of 10% in the tensile strength and the ductility. Table 11.6 shows that Heat 5081 underwent similar changes at elevated tem-

peratures, but did not suffer the reduction in ductility at 25 °C noted for Heat 5085.

Some of the test specimens have been examined metallographically. Figure 11.9 shows the microstructure of control specimen No. AC-8 from Heat 5081 that was tested at 25 °C. This specimen exhibited good ductility and this is reflected in the intragranular, shear-type fracture and the lack of intergranular cracking. Figure 11.10 shows the irradiated specimen from the same heat that was tested at 25 °C. The fracture is largely intergranular, and intergranular cracks are numerous. The control and irradiated specimens

TABLE 11.5. Tensile Properties of
Hastelloy N, Heat 5085.

Specimen No.	Heat Treatment	Test Temp (°C)	Strain Rate (min ⁻¹)	Yield Stress (psi)	Ultimate Stress (psi)	Uniform Elongation (%)	Total Elongation (%)	Reduction in Area (%)
76	As received	25	0.05	52,200	116,400	51.3	52.5	56.3
77	As received	427	0.05	30,700	102,900	57.6	59.4	49.9
284	As received	600	0.02	32,200	85,100	46.6	47.6	40.4
78	As received	650	0.05	28,700	80,700	35.5	36.7	33.2
285	As received	650	0.02	31,900	65,000	25.8	31.8	27.2
283	As received	650	0.002	30,500	64,300	22.6	24.1	28.8
79	As received	760	0.05	32,100	61,500	24.7	27.0	31.9
80	As received	871	0.05	30,700	42,300	9.0	31.8	33.4
81	As received	982	0.05	23,100	23,100	1.8	40.2	43.9
1339	A.R. + 600 hr at 650°C	25	0.05	41,100	115,700	52.5	52.8	41.7
1340	A.R. + 600 hr at 650°C	650	0.05	33,800	76,000	36.8	37.5	36.4
4295	A.R. + 2 hr at 900°C	25	0.05	41,500	120,800	52.3	53.1	42.2
4298	A.R. + 2 hr at 900°C	500	0.05	32,600	94,800	51.2	54.1	40.9
4299	A.R. + 2 hr at 900°C	500	0.002	33,500	100,200	52.0	53.3	41.7
4296	A.R. + 2 hr at 900°C	650	0.05	29,600	75,800	31.7	33.7	34.6
FC-3	A.R. + 2 hr at 900°C + 4800 hr in MSRE salt at 650°C	25	0.05	45,500	111,200	46.8	46.8	31.5
DC-19		500	0.05	33,600	94,300	48.8	49.3	41.5
DC-26		500	0.002	33,400	91,700	43.3	44.3	37.8
DC-14		650	0.05	31,800	70,100	25.8	26.8	30.0
DC-25		650	0.002	31,500	62,500	22.8	24.3	27.2

TABLE 11.6. Tensile Properties of
Hastelloy N, Heat 5081.

Specimen No.	Heat Treatment	Test Temp (°C)	Strain Rate (min ⁻¹)	Yield Stress (psi)	Ultimate Stress (psi)	Uniform Elongation (%)	Total Elongation (%)	Reduction in Area (%)
4300	A.R. + 2 hr at 900°C	25	0.05	52,600	125,300	56.7	59.5	50.5
4303	A.R. + 2 hr at 900°C	500	0.05	32,000	100,300	57.8	60.7	44.4
4301	A.R. + 2 hr at 900°C	650	0.05	32,200	81,800	31.7	33.9	29.9
4302	A.R. + 2 hr at 900°C	650	0.002	32,900	74,900	29.0	29.5	31.8
AC-8	A.R. + 2 hr at 900°C + 4800 hr in MSRE salt at 650°C	25	0.05	47,700	118,700	55.9	57.6	48.8
AC-19		500	0.05	35,800	97,800	53.6	56.6	46.2
BC-9		500	0.002	36,200	95,300	46.2	47.0	38.1
AC-27		650	0.05	32,400	68,400	23.8	24.6	23.1
AC-17		650	0.002	33,600	66,700	22.8	23.2	21.6

Neg Y-78243

80X

a. Fracture

Neg Y-78244

80X

b. Edge about 1/2 in. from fracture

FIGURE 11.9. Microstructure of Hastelloy N, (Heat 5081), Exposed to Salt for 4800 hr at 650 °C. Tested at 25 °C, at a Strain Rate of 0.05 min^{-1} . [(Specimen No. D-16), Etchant: Glyceria Regia].

Neg R-34227

80X

a. Fracture

Neg R-34229

80X

b. Edge about 1/2 in. from fracture

FIGURE 11.10. Microstructure of Hastelloy N, (Heat 5081), Exposed for 4800 hr in the MSRE at 650 °C. Tested at 25 °C, at a Strain Rate of 0.05 min^{-1} . [(Specimen No. D-16), Etchant: Glyceria Regia].

from Heat 5085 tested at 25 °C exhibited the same general characteristics as those illustrated in Figure 11.10. The other specimens examined were tested at 650 °C. The failures were all intergranular with the irradiated specimens exhibiting the typical characteristics of a high temperature, low ductility, intergranular fracture.

We compared the ductilities of the surveillance specimens with those for specimens irradiated in other experiments without salt present. Heat 5081 had been irradiated previously in the ORR.⁽³⁾ The ORR experiment was run at 700 °C to a thermal dose of 9×10^{20} nvt, with the material in the as-received condition. The MSRE surveillance specimens were run at 650 °C to a thermal dose of 1.3×10^{20} nvt; the preirradiation anneal was different. However, none of these differences are

thought to be particularly significant, and the results compare rather well. Figure 11.11 shows that the post irradiation ductilities of Heat 5081 after both experiments are very similar.

MEASUREMENT OF THE NUCLEAR HEATING RATE IN EBR-II

E. E. Bloom, A. F. Zulliger, and J. R. Weir

An experiment has been designed and constructed to measure the axial distribution of nuclear heating in non-fissionable materials in Row 2 and Row 7 positions of the EBR-II. These positions are representative of those to be used in subsequent ORNL materials irradiations. Nuclear heat susceptors (Figure 11.12) will be placed in the central pin of Mark B-7 sub-assemblies. The gas composition, gap

Neg ORNL 67-3531

FIGURE 11.11. Comparative Effects of Irradiation in MSRE and ORR on the Ductility of Hastelloy N, Heat 5081.

FIGURE 11.12. Schematic of Gamma Heat Susceptor

size, and susceptor material were selected on the basis of heat transfer calculations performed on an IBM 360 Computer. The code for determining susceptor temperature includes radial heat transfer from the susceptor by conduction, and radiation and accounts for axial conduction losses through the specimen and support pins. Thermal conductivity, susceptor dimensions, and emissivity values as a function of temperature are incorporated into the program. Susceptors were designed on the basis of published heat rates to produce susceptor centerline temperatures between 500 and 800 °C. Melt wires having melting points both above and below the design temperature (to cover the range of $\pm 50\%$ of the published heating rate) were placed in each susceptor (Figure 11.13). The irradiation will consist of bringing the reactor to the desired power level, followed by an irradiation time of approximately 24 hours. Postirradiation examination will place the actual operating temperature between a lower value (determined by the melt wire of highest melting point which has melted) and an upper value (determined

by the melt wire of lowest melting point which has not melted). It will then be possible, through calculations based on temperature and susceptor dimensions, to calculate the nuclear heating rate to $\pm 10\%$. The results of this experiment will allow subsequent materials irradiations to be conducted at ± 25 °C of the desired irradiation temperature when a susceptor of identical design but containing mechanical properties test specimens is used. Figure 11.14 is a schematic diagram of the specimen holder.

A second objective of this experiment is to measure the axial neutron flux distribution in the Row 2 and Row 7 subassemblies. One tube of the Row 2 and one tube of the Row 7 subassembly is being used for this purpose. Titanium, iron, nickel, and cobalt flux monitors are being used.

A third objective is to determine the combined effects of fast neutron flux and irradiation temperature upon the dynamic recovery of lattice displacement damage. Type 304 SS and high-purity nickel will be irradiated at temperatures between ambient sodium and 650 °C at neutron fluxes between 3 x

Neg 87200

FIGURE 11.13. Photograph of Nuclear Heating Susceptor.

FIGURE 11.14. Schematic of Specimen Holder

10^{15} n/cm²-sec (axial midplane Row 2) and 2×10^{13} n/cm²-sec (axial extreme Row 7). Postirradiation examination will consist of hardness measurements

and electron microscopy studies to determine the effect of irradiation temperature upon the recovery of irradiation-induced displacement damage.

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IRRADIATION DAMAGE TO REACTOR METALS

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A. L. Bement

REACTOR METALS RESEARCHALLOY SELECTION

K. R. Wheeler

The purpose of this program is to study, on a limited basis, materials which are potentially useful for reactor application. From such cursory examination, the more promising alloys can be selected for study in depth. In addition, program alloys are procured, warehoused at BNWL, and disbursed to the various sites participating in the Irradiation Effects on Reactor Structural Materials Program.

Production of a new program material—Incoloy Alloy 800, Heat No. HH7531A—is completed. The heat, fabricated at Huntington Alloy Products Division of INCO, has the following chemistry:

Element	% Composition	Element	% Composition
C	0.03	Mn	1.05
Fe	46.48	S	0.007
Si	0.26	Cu	0.16
Ni	32.44	Cr	19.25

Element	% Composition	Element	% Composition
Al	0.10	Ti	0.20
Co	0.11	Mo	0.12
P	0.011	B	0.0002

All material passed the acidified copper sulphate test for intergranular attack according to the procedures of ASTM-A-393. Each individual piece was visually surface checked and ultrasonically inspected. The tubing was subjected to a 1000 psi internal hydrostatic test, and according to various methods used for inspection, the material was of good quality. Physical properties reported by Huntington Alloy are given in Table 12.1.

Five additional sheets of Incoloy 800 of this program heat are being cold rolled to 0.083 thickness. After final annealing, this material will be shipped to Battelle-Northwest to complete the order. Material from Heat No. HH7531A was distributed as follows:

Shape	Size (In.)	Quantity (Pcs.)	Location
Sheet	0.109 x 12 x 48	4	Battelle-Northwest
Sheet	0.109 x 12 x 48	1	Atomics International
Plate	1 x 24 x 48	3	Battelle-Northwest
Plate	2.5 x 12 x 48	5	Battelle-Northwest
Rod	0.625 x 60	200	Battelle-Northwest
Tube Blanks	1.5 ODx 0.125 wall	12 (376 1b)	Atomics International
Tube Blanks	1.5 ODx 0.125 wall	6 (187 1b)	Superior Tube

TABLE 12.1. As Fabricated Mechanical Properties of Incoloy 800, Heat No. HH7531A

Shape (Thickness or Diam)	Condition	ASTM Grain Size	0.2 YS psi x 10 ³	UTS psi x 10 ³	% Elong.
Sheet (0.109)	Solution Annealed	5.5	40.0	76.5	43.0
Plate (1.00)	Hot Rolled, Annealed	4.5	35.5	75.5	48.0
Plate (2.50)	Hot Rolled, Annealed	5	29.0 ^(a)	73.5 ^(a)	50.0
Rod (0.625 ϕ)	Hot Fin, Annealed	7	33.5	80.0	57.0
Tube Blanks (1.50 OD x 0.125 wall)	Cold Drawn Annealed	4.5	36.0	79.0	44.0

(a) Specified strength values are 30,000 and 75,000 psi for 0.2% YS and UTS, respectively. The low readings for the 2 1/2 in. plate are attributed by Huntington Alloy to the low (0.3% combined Al + Ti) content of this heat of material. Standard material combined (Al + Ti) content is ~0.6%.

In keeping with the major function of Alloy Selection, specimens of 10 alloys residual from the GE-San Jose superheat fuel-clad-materials investigations are undergoing tensile properties evaluations at BNW. The following alloys and numbers of specimens are involved: (a) (irradiated in the GETR) 16 Incoloy 825, 16 Incoloy 801, 6 In 102, 10 Inconel 718, 4 Incoloy 804, 10 Hastelloy F, and 4 Rene-62; (b) (irradiated in the EBR II) 2 Incoloy 800, 4 Inconel 625, and 4 Hastelloy-X. A similar group of specimens are being creep tested at GE-NMPO.

Tensile results of specimens irradiated in the EBR-II for 200 hours and tested at 1300 °F (704 °C) at a strain rate of 0.02 in/min are given in Table 12.2.

At the level of fast fluence reported by GE for the Incoloy 800, the uniform elongation appears not to be affected by the fast neutron

radiation. Necking strain, however, has been reduced approximately 70% by the irradiation. Much greater reductions in total elongation are observed⁽¹⁾ at fluence levels of 2×10^{21} n/cm², with the elongation being about 7%.

Inconel 625 and Hastelloy X are known⁽²⁾ to possess a degree of microstructural instability when aged at elevated temperature. The results of this series of tests conducted at BNW confirm the observations by GE investigators⁽²⁾ that fast neutron spectra cause large reductions in ductility at relatively low fast fluence. The Inconel 625 seems particularly sensitive to elevated temperature aging and to radiation damage. One would expect this, however, from its higher alloying content and the increased number of competing reactions occurring within its structure.

TABLE 12.2. Effect of a Fast Neutron Spectra^(a) ($E > 0.05$ MeV) 200 hr Irradiation on the 1300 °F (704 °C) Tensile Properties of Three Superalloys

Specimen Number	Material	Fluence, n/cm ²	Irradiation Temperature, °F	0.2% YS psi x 1000	UTS psi x 1000	% U.E. ^(b)	% T.E. ^(c)	% R.A. ^(d)
--	Incoloy 800	Control	--	26.7	35.3	11.1	45.5	--
L-6	Incoloy 800	0.17×10^{20}	<1000	24.6	34.3	12.5	22.6	33.3
L-45	Incoloy 800	0.17×10^{20}	<1000	22.5	34.5	12.3	26.5*	35.4
--	Inconel 625	Control	1000	58.3	85.0	16.4	20.8	--
V-18	Inconel 625	2.7×10^{20}	1000	91.9	103.8	3.1	3.6*	19.35
V-29	Inconel 625	2.9×10^{20}	1000	96.6	105	1.7	2.4*	20.00
--	Hastelloy X	Control	1000	26.3	60	26.4	34.9	--
X-2	Hastelloy X	2.7×10^{20}	1000	39.3	60.6	10.5	11.7	26.6
X-17	Hastelloy X	2.9×10^{20}	1000	52.7	69.0	8.34	10.88*	20.0

(a) EBR II, thermal neutron flux negligible

(b) Uniform elongation

(c) Total elongation

(d) Reduction of area

* Fractured at gage length marker

IRRADIATION FACILITIES OPERATION

R. E. Robbins and R. W. Barker

The purpose of this phase of the program is to provide for the timely accomplishment of irradiation, testing, and subsequent processing of the data obtained from 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 unirradiated specimens, will be developed.

Facilities Operation

The experiments discharged from the various Engineering Test Reactor (ETR) core positions for Cycles 86 and 87 are given in Table 12.3 along with the estimated neutron fluxes and exposures. Several ambient temperature capsules were discharged from core positions in

the ETR at the completion of Cycles 86 and 87.

The operating histories of the ETR and G-7 hot water loop for Cycles 85 and 86 are given in Table 12.4. Loop operation was normal for these cycles. The history for the out-of-reactor loop is given in Table 12.5.

An inert (helium) atmosphere, out-of-reactor capsule has been assembled and charged into its initial thermal cycle. This capsule contains annealed AISI 304 and 316 SS tensile specimens. It will duplicate the thermal history of sodium capsule GEH-22-3, which, having received an exposure of 1.3×10^{21} fast fluence ($E > 1$ MeV) at a specimen temperature of 1100 °F (593 °C), was discharged at the conclusion of ETR Cycle 86. Temperature of the sodium capsule was monitored at four locations with four Chromel-Alumel thermocouples. The inert atmosphere capsule will be aged

TABLE 12.3. Summary of Experiments Discharged at the Conclusion of ETR Cycles 86 and 87

Experiment Designation	Number of Specimens	Type of Specimens	Estimated Flux, $n_v^{(a)}$	Estimated fluence, n/cm^2 (a)
<u>CYCLE 86</u>				
GEH-20-251 ^(b)	6	304 SS Miniature Tensile	6.0×10^{13}	2.0×10^{20}
252 ^(b)	6	304 SS Miniature Tensile	10.6×10^{13}	3.8×10^{20}
253 ^(c)	2	A302-B Steel DCB	7.5×10^{13}	2.7×10^{20}
254 ^(c)	2	A302-B Steel DCB	5.5×10^{13}	2.0×10^{20}
257 ^(b)	6	348 SS Miniature Tensile	10.4×10^{13}	1.9×10^{20}
258 ^(b)	6	348 SS Miniature Tensile	7.5×10^{13}	1.4×10^{20}
GEH-14-677	34	Mo, Ni, Re Crystals and Foil	2.7×10^{12}	2.3×10^{19}
679	15	Fe Crystals and Wire	2.7×10^{12}	2.3×10^{19}
686	4	Fe Crystals and 6 Fe Foil	10.4×10^{13}	2.9×10^{20}
<u>CYCLE 87</u>				
GEH-20-243	36	Zr-2 Corrosion Blanks	10.0×10^{13}	7.2×10^{20}
248	6	Hastelloy-X Tensile	10.0×10^{13}	5.7×10^{20}
250	6	304 SS Tensile	10.0×10^{13}	5.7×10^{20}
260	18	25-20 SS Tensile	25.0×10^{13} (d)	5.0×10^{20} (d)
261 ^(b)	6	304 SS Miniature Tensile	10.6×10^{13}	1.9×10^{20}
262 ^(b)	6	304 SS Miniature Tensile	6.0×10^{13}	1.1×10^{20}
263	6	Inconel-X Tensile	7.5×10^{13}	1.4×10^{20}
274 ^(c)	2	A542 Steel DCB	7.5×10^{13}	1.3×10^{20}
275 ^(c)	2	A542 Steel DCB	5.5×10^{13}	1.0×10^{20}
GEH-14-684	36	Mo and Re Crystals and Foil	3.6×10^{13}	2.0×10^{20}

(a) Neutrons with energies greater than 1 MeV.

(b) Indicates elevated temperature capsule experiment.

(c) Indicates double length quadrant.

(d) Indicates thermal neutron flux.

NOTE: Total specimens discharged, Cycles 86 and 87 = 205

Total specimens discharged, ETR G-7 Loop, Cycles 86 and 87 = 116

Total specimens discharged, ETR G-7 Loop, to date = 3,124

at 1100 °F (593 °C) through four simulated ETR cycles totaling 99.9 days, as outlined in the following schedule.

ETR Cycle	Effective Days	Number of Mid-Cycle Shutdowns	Total Days
83	26.7	0	26.7
84	22.3	0	49.0
85	27.0	4	69.0
86	30.9	0	99.9

The capsule and specimen array are shown in Figure 12.1.

A series of irradiated Tempil melt pellets has been examined and tested to determine the effect of irradiation on their melting points. The previously observed⁽³⁾ structural instability of the pellets has been attributed to irradiation effects, since pellets exposed to only the thermal environment for long times exhibited no porosity. Examination of pellets irradiated at temperatures

TABLE 12.4. Summary of Operating History for ETR and G-7 Hot Water Loop - ETR Cycles 85 and 86.

Operating History		ETR Cycle Number	
		85	86
1.	<u>Reactor History</u>		
	Start up ^(a)	11-17-66	12-26-66
	End of Cycle	12-15-66	1-30-67
	Megawatt Days	3,500	5,403
	Effective Days at 175 MW	20.0	30.9
	Number of Scrams ^(b)	N.A.	4
	Number of Shutdowns	4	1
2.	<u>G-7 Hot Water Loop History</u>		
	Maximum Temperature	520 °F	540 °F
	Effective Days above 200 °F	20.8	31.7
	Effective Days at Oper. Temp.	18.7	29.8
	Operating Efficiency ^(c)	104%	102.5%
	Temperature Efficiency ^(d)	89.7%	93.9%

(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).

TABLE 12.5. Summary of Operating History and Discharge Schedule for Out-of-Reactor Loop, ETR Cycles 85 and 86

Loop History		Cycle Number	
		85	86
1.	<u>Time at Temperature, hr</u>		
	200 °C (392 °F)	32.1	21.0
	218 °C (424 °F)	4.0	8.3
	246 °C (475 °F)	15.6	17.3
	282 °C (540 °F)	448.3	714.2
	Total Time, hr	500.0	760.8

2. Specimen Discharge

(None after Cycle 85 or 86)

FIGURE 12.1. View of Capsule and Contents of GEH-22-3 Inert Atmosphere Out-of-Reactor Capsule

above their melting points is hindered due to pellet decomposition. It is extremely difficult to determine whether or not a pellet has melted while in the reactor.

The results of the melting point tests, outlined in Table 7.6, illustrate the variations in melting points between the irradiated and unirradiated pellets. It should be noted that the observed differences are not uniform but depend upon the particular temperature being considered.

The above results indicate that Tempil melt pellets are not suitable as thermal monitors in neutron environments.

Data Processing

Two hundred and twenty-four tensile data reports have been added to the

Radiation Effects on Metals (REM) computer program during the quarter, raising the accumulative total to 5009. Conversion of the program language from FORTRAN II to FORTRAN IV for use with the UNIVAC 1107 is still in progress.

IRRADIATION DAMAGE TO STAINLESS STEELS

R. E. Robbins, J. J. Holmes,
J. L. Brimhall, B. Mastel

The purpose of this program is to determine the combined effects of neutron irradiation and environment on stainless steels. Irradiation-induced mechanical property changes will be determined as a function of fast and thermal neutron fluence, temperature and environment of irradiation and postirradiation test temperatures and environments.

TABLE 12.6. Melting Points of Several Irradiated and Unirradiated Tempil Melt Pellets

Melting Points (°F)		
<u>Unirradiated</u>	<u>Irradiated</u>	<u>Vendors</u>
750	877	750
810	877	800
920	990	900
960	960	950
980	1010	977

Emphasis will be placed on tensile and creep investigations and on studies of the mechanisms of irradiation-induced property changes.

Annealed AISI 304 SS tensile specimens have been irradiated at 650 °C in a NaK environment to an exposure of 3.5×10^{20} fast fluence and tested at temperatures to 750 °C. As shown in Figure 12.2, the effect of increasing test temperature is to reduce the strength and ductility of 304 SS to values below the original room temperature values. At a test temperature of 750 °C, the strengths of the irradiated material approach those of the unirradiated, as-fabricated material; but the irradiated ductilities remain below the unirradiated values. Transmission electron microscopy, shown in Figures 12.3 to 12.6, has been performed on unstressed sections of tensile specimens tested at room temperature, 300, 500, and 750 °C in order to observe any microstructural changes at these temperatures. A fine carbide precipitate, probably an $M_{23}C_6$ type, was observed in the matrix and grain boundaries of specimens tested at room temperature and 300 °C. At 500

and 750 °C test temperatures, carbides about 270 and 700 Å, respectively, in size were found in grain boundaries and associated with dislocations in the matrix. It appears that carbide precipitation could be partly responsible for the loss of ductility at the higher test temperatures. No helium agglomeration could be observed.

The question arises as to why carbides greater than about 300 Å but less than 700 Å in size were not observed in the 304 SS specimen tested at room temperature due to the 650 °C irradiation temperature. It appears that under the conditions described neutron irradiation may be inhibiting carbide precipitation in these specimens. Similar observations have been made by Meakin and Greenfield⁽⁴⁾ for neutron irradiated molybdenum. However, to clarify this point, additional investigations will be made.

The effect of test temperature on the strength and ductility of annealed AISI 304 SS irradiated at 290 °C is shown in Figure 12.7. These tensile specimens received an exposure of 6.0×10^{21} fast fluence, which is the highest obtained to date in the ETR G-7

FIGURE 12.2. The Effect of Test Temperature on the Strength and Ductility of AISI 304 SS Irradiated in the ETR. (Irradiated at 650 °C to 3.51×10^{20} nvt)

*FIGURE 12.3. AISI 304 SS Irradiated
at 650 °C to 3.5×10^{20} Fast Fluence;
Room Temperature Test. (Note: Distance
between markers is 1 μ .)*

FIGURE 12.4. AISI 304 SS Irradiated at 650 °C to 3.5×10^{20} Fast Fluence; 300 °C Test. (Note: Distance between markers is 1 μ .)

*FIGURE 12.5. AISI 304 SS Irradiated
at 650 °C to 3.5×10^{20} Fast Fluence;
500 °C Test. (Note: Distance between
markers is 1 μ .)*

FIGURE 12.6. AISI 304 SS Irradiated at 650 °C to 3.5×10^{20} Fast Fluence; 750 °C Test. (Note: Distance between markers is 1 μ .)

FIGURE 12.7. The Effect of Test Temperature on the Strength and Ductility of Irradiated AISI 304 SS. (Irradiated at 290 °C to 6.0×10^{21} Fast Fluence)

hot water loop for 304 SS. The tensile data plotted in Figure 12.7 are of significant importance for several reasons. Although the control data are not shown, radiation hardening of the yield stress was observed at test temperatures above one-half of the melting point. Also a direct comparison of these data with that obtained by extrapolation from lower exposures can be made to test the reliability of analytical models of irradiation effects on tensile properties. The data should provide an important link for the comparison of fast-to-thermal irradiations because

exposures of about 6.0×10^{21} fast fluence represent relatively short-term irradiations in a fast reactor. A better understanding of the mechanisms of radiation-induced property changes in 304 SS will be obtained by comparing the nature of displacement type damage produced by the 6.0×10^{21} fast fluence exposure with that produced at lower exposures.

One of the significant features about the tensile data obtained for 304 SS at an exposure of 6.0×10^{21} fast fluence is illustrated in Figure 12.8. The uniform ductility follows a

FIGURE 12.8. The Effect of Test Temperature on the Ductility of AISI 304 SS. (Irradiated at 290 °C in the annealed condition, all transverse specimens)

general trend in the development of a recovery peak as the exposure increases to 1.0×10^{21} fast fluence. The ductility recovery peak is probably caused by the annealing out of displacement type damage. The ductility behavior at 6.0×10^{21} fast fluence is different from what might be predicted from ductility values at 1.0×10^{21} fast fluence. Probably the most important difference is the removal of the recovery peak in the 400 to 700 °C test temperature range. The behavior of the ductility at this exposure indicates the presence of some type displacement defect, probably different from that responsible for the ductility behavior up to 1.0×10^{21} fast fluence, which is stable at test temperatures above one-half the melting point. This explanation is substantiated by the fact that the yield strength of 304 SS continues to increase with exposure at all temperatures. For example, at 650 °C the yield strengths at 1.0 and 6.0×10^{21} fast fluence are 20,000 and 38,000 psi, respectively. However, to observe the damage state that exists at the higher exposure, transmission electron microscopy will be performed.

It is interesting to note that the strength and ductility of annealed AISI SS converges at an exposure of 6.0×10^{21} fast fluence. (Figures 12.9 and 12.10). These plots were constructed from irradiation data obtained in the ETR and EBR-II for a variety of irradiation temperatures and illustrate the way in which data at 6.0×10^{21} fast fluence can be used to relate fast-to-thermal irradiation studies. It can be seen

that, independent of irradiation temperature, the yield and tensile strengths continue to increase; but the ductility changes very little beyond the exposure of 6.0×10^{21} fast fluence. The increase in strength is probably caused by irradiation hardening. This suggests the presence of some type of defect, other than the normal defect clusters observed at low exposures and irradiation temperatures below half the melting point, which is stable at the 750 °C test temperature. For exposures less than 6.0×10^{21} fast fluence, the strength of 304 SS is a function only of exposure as long as the irradiation temperature is 290 °C or above. This does not appear to be the case for uniform ductility since only a scatter band could be delineated.

It is worth mentioning that, at test temperatures below 750 °C, the tensile data do not converge at a particular exposure. The reason for this behavior is not clear, but it may be that the unstable form of the displacement damage is not fully annealed out at the lower test temperatures.

IRRADIATION DAMAGE TO NICKEL-BASE ALLOYS

I. S. Levy

The requirements for corrosion resistance and high temperature strength place nickel-base alloys among the more promising materials for advanced nuclear reactor applications. However, when these alloys are irradiated at service temperatures, they generally suffer embrittlement and reduction in rupture life. The purpose of this program is to determine the effect of modified microstructures upon the irradiation

FIGURE 12.9. Strength of Annealed AISI 304 SS at 750 °C.

FIGURE 12.10. Ductility of Annealed AISI 304 SS at 750 °C

stability of nickel-base alloys. These structural modifications are made by preirradiation thermal or thermo-mechanical treatments and are evaluated by tensile and stress-rupture tests and microstructural examinations.

Solid-Solution Hardened Alloys

Hastelloy X-280

Tensile and stress-rupture tests upon specimens irradiated at 540 °F (280 °C) to 1×10^{20} nvt (>1 MeV) have shown that certain experimental pre-irradiation treatments can improve post-irradiation mechanical properties.^(5,6) To elucidate the mechanisms for these improvements, extensive micro-structural examinations have been undertaken using optical, replica, and transmission microscopy.^(7,8,9,10,3)

In previous reports, it was shown that tensile strain at 1350 °F (732 °C) produces a uniform dispersion of a very fine (≤ 400 Å) precipitate within the matrix of unirradiated standard solution-treated material.⁽⁹⁾

Irradiation of this solution-treated material at 540 °F (280 °C) to 1×10^{20} nvt (>1 MeV) produced small point defect clusters 60-120 Å in diameter, with a density of $\sim 2 \times 10^{16}/\text{cm}^3$.⁽¹⁰⁾ These defects annealed out of the matrix during the pretest soak at 1350 °F (732 °C);⁽⁸⁾ but tensile strain produced a change in the morphology and positioning of the strain-induced precipitate (relative to that observed in the unirradiated material) in that it was now larger, had a different etching character, and was concentrated within 1 to 2 μ of grain boundaries.⁽¹⁰⁾

Ductility of the irradiated material was about one fifth of that of the unirradiated material.

In this standard-treated material, annealing studies were initiated to investigate the mechanisms, and to link cluster defect breakup at high temperature to the resultant heavy precipitation around grain boundaries during tensile testing. Annealing at 935 °F (500 °C) for 2 hr of specimens irradiated at 540 °F (280 °C) produced only a very slight growth of the cluster defects with no change in density. As in the as-irradiated condition, the defects were uniformly dispersed throughout the matrix and around precipitates, twins, and grain boundaries. This quarter, annealing was performed at 1113 °F (600 °C) for 2 hr. An appreciable decrease in cluster density was observed; cluster size increased only slightly. But more significantly, denuding of the defects occurred within 1 to 2 μ of precipitate particles in the matrix or in grain boundaries, while denuding occurred at twin boundaries (Figure 12.11).

Since stress fields associated with noncoherency are short range, it is not felt that this is the driving force for the denuding around matrix and grain boundary precipitates. Instead, the driving force is considered to be a chemical gradient caused by the difference between the composition of the precipitates and that of the surrounding matrix and defects. As the annealing temperature is raised sufficiently to allow breakup of the cluster defects, this driving force

Neg 4667A

15,000X

FIGURE 12.11. Cluster Defect Denuding Around Grain Boundary Precipitates in Irradiated Hastelloy X-280 after 600 C/2 hr anneal.

results in preferential diffusion of the constituent interstitials to these particles. Therefore, it is now felt that the precipitate found around grain boundaries of irradiated standard-treated material strained at 1350 °F (736 °C) can be explained by:

- This driving force for diffusion of irradiation-induced interstitials towards grain boundary precipitates
- The addition of excess vacancies due to the breakup of the irradiation defects
- Pipe diffusion towards grain boundaries during the tensile straining.

This precipitation, nucleating as it does on dislocations piled up at grain boundaries, results in a hardening of the grain boundary area, and causes the observed loss of ductility.

Inconel 600

It was previously reported⁽¹⁰⁾ that, for material irradiated at 540 °F (280 °C) to 1×10^{20} nvt (>1 MeV) and tested at 1350 °F (732 °C), improvements in postirradiation tensile ductility could be achieved with no reductions in strength through the use of experimental preirradiation aging treatments. Microstructural examination of unirradiated specimens had suggested a possible mechanism for these improvements; namely, that carbides formed at the grain boundaries during the pre-irradiation treatment depleted the area adjacent to the boundaries of solid solution hardeners—such as chromium—and made that area more ductile and better able to resist crack propagation. Unirradiated ductility data appeared to support this suggestion in that non-uniform elongation increases of up to 300% were observed for some treat-

ments, an effect considered to be a manifestation of a more ductile grain boundary area.⁽³⁾ Phase analyses conducted during the past quarter add further weight to this hypothesis, for example, while the standard treated material had the M_7C_3 phase (a chromium-rich phase) in only rare amount, the experimental treatments had it in abundant amounts.

Stress-rupture tests at 1350 °F (732 °C) and the 1000 hr rupture stress of 9800 psi were completed during the past quarter on specimens given preirradiation treatments and irradiated at 540 °F (180 °C) to 1×10^{20} nvt (>1 MeV). Preliminary examination of the data indicates that some treatments can improve postirradiation rupture life. The tests on the unirradiated control specimens are in progress.

Precipitation-Hardened Alloys

Inconel X-750

The ductility improvement found for certain experimental treatments after irradiation at 540 °F (280 °C) to 1×10^{20} nvt (>1 MeV) was correlated with microstructural observations using replica microscopy.⁽¹⁰⁾ The correlation was that treatments that had the best postirradiation (and unirradiated) ductility had retained more grain boundary carbides and a larger denuded zone around the grain boundary after tensile testing at 1350 °F (732 °C). Phase analysis was instituted during the past quarter to illuminate this area of investigation.

Stress-rupture tests at 1350 °F (732 °C) and the 1000 hr rupture stress of 37,500 psi were initiated

on specimens irradiated at 540 °F to 1×10^{20} nvt (>1 MeV) and their controls during the past quarter.

IRRADIATION DAMAGE TO SINTERED ALUMINUM PRODUCT

A. J. Lovell

A program to determine the effect of irradiation on Sintered Aluminum Products (SAP) is being initiated. The objective of the program is to provide sufficient irradiation effects data to allow an adequate and safe design using SAP in an irradiation environment. The few irradiation program efforts in the past on SAP material have shown, in general, little effect of displacement damage or mechanical properties at relatively low exposures. Some low temperature irradiations have shown slight increases in strength and reduction in ductility over a range of test temperature to 400 °C. Emphasis should now be placed on higher neutron fluences and higher irradiation and test temperatures.

The program has been outlined to provide high temperature irradiations (300 to 450 °C) of SAP to fluence levels of 1×10^{22} ($E \geq 1$ MeV). The primary emphasis is being placed on postirradiation creep-rupture data.

The irradiations will be carried out in the G-7 loop in ETR and in instrumented capsules placed in the ETR core. The SAP specimens irradiated in the

G-7 loop will be placed in helium-filled sheath capsules to protect the SAP from the loop water. The gas gap between the sheath container and the SAP specimen will be made such that the specimens will be heated by gamma heating to 300 and 400 °C during irradiation.

The instrumented capsules being designed by Atomics International will be operated at 455 °C. The temperature will be controlled by the binary gas method.

The irradiations will be conducted under existing irradiation agreements between Battelle-Northwest and Idaho Nuclear Corporation. The shielded capsule will be under GEH-20 and the AI designed instrumented capsules under GEH-14. The irradiations should be initiated by June or July 1967.

The SAP 895 (10.5% Al_2O_3) and SAP 930 (7% Al_2O_3) material to be used on the program is on hand. Characterization of the 0.4 in. diam extruded rod stock is in progress. Included in the characterization will be chemistry, metallography and mechanical properties.

The chemical compositions of the starting powders as reported by Montecalini, are given in Table 12.7. The chemical analysis for billets sent to AI from lot 1 g and 3 g are shown in Table 12.8. Tensile test data on tests, ran on billets from lot 1 g and 3 g at Montecalini, are presented in Tables 12.9 and 12.10 for room temperature and 480 °C (900 °F) tests, respectively.

TABLE 12.7. Chemical Composition of Starting Powders

Lot	Powders		Al_2O_3 , %	Fe, %	Si, %	Zn, %	C, %
	Batch	Drum					
1-1g	N114	6	10.6	0.046	0.046	0.0053	0.223
3g	N128	26	6.40	0.054	0.04	0.012	0.12

TABLE 12.8. Chemical Analysis of Specimens Sent to AI

Lot	Al ₂ O ₃ , %	Extracted Gas, cm ³ /100 g	N ₂ , ppm	Fe, %	Si, %	Cu, %	Zn, %	Ti, %	C, %
1g	10.70	1.5	340	0.06	0.04	n.d.	0.005	n.d.	0.12
3g	6.45	1.2	200	0.06	0.04	0.001	0.012	0.001	0.10

TABLE 12.9. Mechanical Properties at Room Temperature

Lot	Billet Number	Al ₂ O ₃ , Content	UTS, psi	YS, psi	UE (A ₄) %	Mechanical Properties
3g	603 (rear)	7%	33,140	21,905	20.0	Longitudinal
1g	510 (front)	10%	41,820	30,580	6.4	Transverse

TABLE 12.10. Mechanical Properties at 480 °C

Lot	Billet Number	Al ₂ O ₃ , Content	UTS, psi	YS, psi	UE(A ₄) %	Mechanical Properties
1g	510 (front)	10%	9,100	8,250	3.2	Transverse
3g	603 (rear)	7%	8,820	8,250	3.2	Longitudinal

The chemical and mechanical data from AI for the 2 in. billets will be included in a later report when both SAP 895 and SAP 930 data are available.

IN-REACTOR MEASUREMENTS OF MECHANICAL PROPERTIES

E. R. Gilbert

The in-reactor measurements program is intended to:

- Determine the effects of irradiation on the mechanical properties of reactor structural materials
- Establish property data suitable for design purposes
- Consider mechanisms controlling deformation using irradiation as a tool in identifying mechanisms

through its effects on macroscopic deformation.

The program currently involves conducting in-reactor creep measurements on annealed 304 SS, Hastelloy X, heat-treated zirconium 2.5 wt% niobium, and Zircaloy-2. Capsule development is being pursued so that program capabilities may be extended.

304 Stainless Steel

In-reactor creep tests and corresponding control tests are being continued. The results have indicated little effect of irradiation on minimum creep rate but have revealed large effects on rupture life and total elongation. The reduction of rupture

life due to irradiation appears to be more severe above 700 °C than below it.

Hastelloy X

The first of a series of in-reactor creep tests to be conducted on Hastelloy X-280 was tested to rupture at 1000 °C. The total elongation was greater than 11% as measured by the in-reactor extensometer system, and the rupture life was less than that predicted from unirradiated data for this alloy.

Zr-2.5 wt% Nb

In-reactor and unirradiated control creep tests are being conducted on heat-treated Zr-2.5 wt% Nb in the temperature range of 300 to 400 °C. The heat treatment consists of a 15 min anneal into the alpha and beta region at 870 °C followed by a quench and a 24 hr aging treatment at 500 °C. In order to study the effect of irradiation under stress on ductility, specimens precrept in the reactor at 350 and 400 °C were tested in a universal tensile machine. The unirradiated control specimens and the precrept specimens had essentially the same total elongation indicating no appreciable loss of ductility.

DAMAGE MECHANISMS

F. A. Smidt, Jr.

The objective of this program is to determine how irradiation produced defects modify the mechanical properties of metals by interacting with moving dislocations.

Radiation Damage in Alpha Iron

Among the more prominent features of radiation damage in metals is the change in mechanical properties. The understanding of these changes

is both an important economic problem and an important scientific problem.

This study was directed toward an investigation of the basic dislocation-defect interactions which give rise to the changes in mechanical properties. At the time of initiation of the study, evidence of the presence of defects in irradiated material was available from direct observations by transmission electron microscopy⁽¹¹⁾, Figure 12.12. It was not apparent from this evidence however, just how the defects were modifying the mechanical properties or indeed whether those visible were actually the source of the observed mechanical property changes since iron contains a very low density of visible defects, Figure 12.13.

The techniques of thermally activated flow⁽¹²⁾ showed promise of correlating the observed microscopic defects with the dislocation motion. Studies of irradiated Cu^(13,14) have shown that marked changes occur in the temperature dependence and strain rate dependence of the flow stress when defects are introduced into the metal. Although there is general agreement as to the role of the defects in modifying the properties there is as yet no consensus as to the specific mechanism involved in the interaction.

The first results on a bcc metal⁽¹⁵⁾ showed no such pronounced changes as those occurring in fcc metals, with only minor changes in activation volume noted around room temperature and negligible changes at lower temperatures.

Materials and Test Procedures

The specimens tested in these experiments were prepared from two lots

Neg 2207B

75,000X

FIGURE 12.12. Electron Transmission Micrograph of Molybdenum Irradiated to a Fluence of 1×10^{19} ($E > 1$ MeV) and Subsequently Deformed. (Dark areas are high density of clustered defects, light areas correspond to slip systems in molybdenum.)

Neg 4596B

150,000X

FIGURE 12.13. Electron Transmission Micrograph of Iron Irradiated to a Fluence of 9×10^{19} ($E > \text{MeV}$) at 60 °C. (Johnson-Matthey high purity iron, C content unknown but believed to be low. Small black dots are defect clusters.)

TABLE 12.11. Compositions of Materials Studies (Impurity concentrations in ppm)

	Ferrovac E, Fe	BMI Zone- Refined Fe
C	50	3.3
N	1	<1
O	200	2.6
Si	200	10
Ni	20	10
Al	--	15

of iron whose composition is listed in Table 12.11. Ferrovac E is a vacuum melted electrolytic iron and the zone-refined iron is an iron prepared by a floating zone induction heating operation by Battelle-Columbus. The tensile specimens were machined from 1/8 in. rod with a reduced section of 1 in. and a gage diam of 0.090 in., electropolished, recrystallized at 750 °C for 1 hr, and furnace cooled.

Two types of tests were used to investigate the thermally activated component of the flow stress, a strain-rate change test and a temperature change test. In the strain-rate change tests, the stress increment accompanying a ten-fold increase in strain rate was measured and used in turn to calculate an activation volume. The base strain-rate was $3.33 \times 10^{-5} \text{ sec}^{-1}$ at each temperature tests were performed. Samples tested at cryogenic temperatures were prestrained at room temperature to inhibit twinning at lower temperatures and to start the tests with the same dislocation density and substructure arrangement.

The temperature change tests were used to determine the temp-

erature at which the effective stress became zero (T_c) and the temperature dependence of the flow stress $(\Delta\tau/\Delta T)_\dot{\epsilon}$. The latter quantity is used in determining experimental activation enthalpies. In these tests the temperature is decreased in steps of about 10 °C, and the increase in flow stress accompanying this change is measured. A cryostat using a spray of liquid nitrogen as coolant was used in these tests. The techniques and apparatus are described in detail elsewhere.⁽¹⁶⁾

Results - Thermally Activated Flow

A change in the mechanism controlling thermally activated flow can be detected by measuring the activation volume at a fixed temperature, $V^* = kT \left[(\partial \ln \dot{\gamma} / \partial \ln \dot{\gamma}_0) / \partial \tau^* \right]_T$. A change in V^* would be indicative of a change in mechanism.

The results of such experiments at 295 °K on unirradiated Ferrovac E, samples irradiated to a fluence of $1.4 \times 10^{18} \text{ n/cm}^2$ and $8.3 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$), and a sample irradiated to 8.3×10^{19} and given a postirradiation anneal at 350 °C for 2 hr are shown in Figure 12.14 as a function of true strain. It should be noted that the values at low strains for the irradiated samples are significantly lower than those for the control sample and that complete recovery results from the anneal at 350 °C. The reproducibility of these measurements of activation volumes is about $\pm 1.0 \times 10^{-22}$ at $30.0 \times 10^{-22} \text{ cm}^3$ and $\pm 0.5 \times 10^{-22}$ at $15.0 \times 10^{-22} \text{ cm}^3$. Various quenching and aging treatments have been performed on the furnace cooled material to ascertain the influence

FIGURE 12.14. Activation Volume Measurements at 295 °K as a Function of Strain for Ferrovac E Fe for the Following Condition. [Control sample, after irradiation to a neutron fluence of 1.4×10^{18} ($E > 1$ MeV), after irradiation to 8.3×10^{19} , and after irradiation to 8.3×10^{19} and a postirradiation anneal at 350 °C for 2 hr.]

of carbon in solution or in the form of precipitates on the activation volume. A water quench from 800 °C displaced the curve for the control sample upward by 1.4×10^{-22} cm³ and an aging treatment of 24 hrs at 300 °C reduced it by 7.2×10^{-22} cm³, while a 2 hr anneal at 350 °C had no effect on the activation volume. The changes in activation volume produced by irradiation are thus greater than those produced by a redistribution of carbon in the samples and are considered significant

although (as will be shown later) they are not large enough to indicate a change in the rate controlling mechanism. Measurements at 200 °K show even less change in the activation volume.

Another feature of these results which should be noted is the increase in activation volume toward the values for the control as the sample is deformed. In molybdenum the deformation has been shown to be highly localized (Figure 12.12), with the first dislocations moving through

the lattice and clearing a defect free "channel" which gradually widens as the deformation proceeds. If the initial decrease in activation volume is due to the defects introduced by the irradiation, then their removal by interaction with moving dislocations should produce an increase in activation volume, toward the control, as deformation proceeds.

The same trends are seen in activation volume measurements on Battelle zone-refined iron

(Figure 12.15). The change in activation volume of the control sample with strain is believed to be due to a change in the density of mobile dislocations during the deformation. Similar trends are seen in other materials that do not display a drop-in-load yield point such as high purity Mo.⁽¹⁴⁾ Quenching the BMI iron from 800 °C displaced the curve upward by $0.8 \times 10^{-22} \text{ cm}^3$ and aging for 24 hr at 300 °C did not result in any change from

FIGURE 12.15. Activation Volume Measurements at 295 °K as a Function of Strain for BMI Zone-Refined Fe for the Following Condition. [Control sample, after irradiation to a neutron fluence of 1.4×10^{18} ($E > 1 \text{ MeV}$), after irradiation to 8.3×10^{19} , and after irradiation to 8.3×10^{19} and a postirradiation anneal at 350 °C for 2 hr.]

the control sample. Irradiation in this case also reduced the activation volume slightly; a post-irradiation anneal at 350 °C for 2 hr resulted in recovery. However, in this case, the 350 °C anneal and strains of 7-10% gave V^* values above the control. It is not evident why recovery should occur.

Another indication of a change in mechanism can be gained from temperature change tests. These tests are used to determine the temperature dependence of the effective stress and accurately determine T_C , the temperature at which thermally activated flow becomes so rapid that it no longer controls the deformation.

In Ferrovac E (Figure 12.16), irradiation decreases the slope slightly and increases T_C about 15°. Because the 8.3×10^{19} fluence samples failed to work harden at temperatures below 0 °C, no useful data was obtained from them.

The temperature change data for BMI iron is shown in Figure 12.17 for the control sample and for the 8.3×10^{19} fluence samples. Previous tests of BMI iron have shown that the deviation of the $\Delta\tau/\Delta T$ versus T plot from a linearity the solid line in Figure 12.16 at about 225 °K is due to a decrease in τ_i during the temperature change test. The decrease in τ_i is attributed to a rearrangement of the substructure

FIGURE 12.16. $(\Delta\tau/\Delta T)_\epsilon$ Data from Temperature Change Tests of Ferrovac E Fe Control Samples and Samples Irradiated to a Fluence of 1.4×10^{18} n/cm² ($E > 1$ MeV)

FIGURE 12.17. $(\Delta\tau/\Delta T)_\epsilon$ Data from Temperature Change Tests on BMI Zone-Refined Fe Control Samples and Samples Irradiated to a Fluence of 8.3×10^{19} n/cm² ($E > 1$ MeV)

under increasing stresses at low temperatures.⁽¹⁶⁾ It should be noted that irradiation to a higher fluence level not only increases T_C about 5 °K and decreases the slope but it also prevents this substructure rearrangement. Tests of 1.4×10^{18} n/cm² samples gave the same temperature dependence and T_C but did not inhibit the substructure re-arrangement. The increases in T_C found in these tests are consistent with the decreases observed in activation volume measurements (V^* is inversely proportional to $\tau^*^{1/2}$).

In a previous paper⁽¹⁶⁾ it was shown that thermally activated deformation in iron could be accurately described by the empirical relation $H/H_0 = T/T_C = (1 - \sqrt{\tau^*/\tau_p})$, where H_0 is the activation enthalpy at T_C , and τ_p is the effective stress at 0 °K. This latter quantity can be evaluated from the temperature dependence of the flow stress and is found to be 57 Kg/mm² in the irradiated samples as compared with 63 Kb/mm² in the control samples. An estimate of H_0 in the irradiated samples was made assuming the slope, dH/dT , was the same as for the control samples and with T_C equal to the observed value in the irradiated samples. H_0 for Ferrovac E increased from 0.93 eV to 0.97 eV and for BMI from 1.00 eV to 1.01 eV. A plot of V^* versus τ^* representative of unirradiated Ferrovac E and BMI iron is shown in Figure 12.18; it can be seen that the observed changes in V^* in the irradiated samples actually represent very small changes in τ^* for the thermally activated process. Thus

FIGURE 12.18. Activation Volume-Effective Stress Relationship for Ferrovac E and BMI Zone-Refined Iron as Deduced from Empirical Equations for Thermally Activated Flow.

it is concluded that the changes in these parameters of thermally activated flow are all quite small and represent only a minor modification in the mechanism of dislocation motion. Other investigations of bcc metals vanadium⁽¹⁸⁾, iron⁽¹⁹⁾, and niobium,⁽²⁰⁾ confirm the conclusion that these changes are of a minor nature.

Results - Athermal Stress

The athermal component of the flow stress is a measure of the internal stresses in a metal arising from stress fields with a longer range than those which can be overcome by thermal fluctuations. Among the factors contributing to τ_i are precipitate particles, dislocation networks produced by cold work, and solute atoms. In these experiments

the lower yield stress at 295 °K was used as an index of τ_i . If absolute values of the yield stress were to be quoted for these experiments a τ^* of about 0.2 to 0.5 kg/mm² should be subtracted from the lower yield stress to give τ_i . As mentioned above irradiation produced a negligible change in the thermally activated flow curves, so the increase in lower yield stress is used here as a measure of the increase in τ_i with exposure. The strengthening produced in Ferrovac E and Battelle zone-refined iron at fluences of 1.4×10^{18} and 8.3×10^{19} are shown in Figure 12.19.

The most prominent feature of these results is the much greater hardening at equivalent neutron fluences in the Ferrovac E iron and therefore a greater rate of strengthening with fluence. Because this material has the higher carbon content, it would appear that carbon is playing a role in the strengthening. The yield stress ($\tau = 6/2$) at a fluence of 8.3×10^{19} is double the yield stress in the control samples while for BMI iron it is about 1.5 times the control sample. Grain size does not influence the accumulation of damage at high fluences although at lower fluences large grained material hardens more

FIGURE 12.19. Increase in Lower Yield Stress as a Function of Square Root of Fluence for Ferrovac E and BMI Zone-Refined Irons

than small grained material.^(21,22) In the experiments reported here, the Ferrovac E had a smaller grain size ($\sim 25 \mu$) than the BMI ($\sim 50 \mu$) so this factor is not influencing the trends observed.

Studies of microhardness recovery in iron⁽²³⁾ show that the damage anneals over the range 300-500 °C. An investigation of the microstructural changes accompanying these changes in hardness⁽²⁴⁾ showed that small spot defects 25-50 Å in diameter grew and formed prismatic loops at about 350 °C, and these loops were stable to at least 500 °C. These loops have been identified as being interstitial in nature.⁽²³⁾

The results of a study of recovery in the samples irradiated in these experiments are summarized in Table 12.12. The first column shows the percentage recovery after a 2 hr anneal at 350 °C in the lower yield stress, and the second column shows the percentage recovery in the $\Delta\tau$ used for activation volume measurements. Examining first the recovery in lower yield stress, it is noted that both materials showed greater recovery at the lower

fluence; the BMI iron showed greater recovery at both fluence values.

It is difficult to compare the rate sensitive stress for the two materials because the BMI iron did not return to the value for the control sample after annealing. It should be noted, however, that both Ferrovac E and BMI showed greater recovery in τ^* at the higher fluence which is just the reverse of the behavior of τ_i .

Discussion

A satisfactory explanation of the influence of radiation damage on the mechanical properties of iron must consider:

- The role of carbon
- A strengthening mechanism taking into account the defects visible by electron microscopy
- The observed changes in the thermally activated flow stress.

The mobilities of interstitial atoms, vacancies, and carbon atoms were considered, and their relative importance during irradiation and annealing is summarized in Table 12.13. The jump frequencies for interstitials and vacancies were estimated from the equation $\Gamma = z \nu \exp(-E_m/kT)$, where

TABLE 12.12. Recovery of Irradiation Produced Changes by a 2 hr Anneal at 350 °C

<u>Material</u>	<u>$\Delta\tau$(LYS)</u>	<u>$\Delta\tau$($\dot{\epsilon}$ Change)</u>
Ferrovac E - 1.4×10^{18} nvt	92%	80%
Zone-Refined - 1.4×10^{18} nvt	100%	130%
Ferrovac E - 8.3×10^{19} nvt	18%	100%
Zone-Refined - 8.3×10^{19} nvt	47%	175%

TABLE 12.13. Defect Migration in Iron

Defect	Irradiation		Defect Spacing (Å)	2 hr Anneal 350 °C	
	$\Gamma_{60\text{ °C}}$ (sec ⁻¹)	$\bar{X}$ (Å)		$\Gamma_{350\text{ °C}}$ (sec ⁻¹)	$\bar{X}$ (Å)
Interstitial	7.2×10^8	4900	1000 Å	1.7×10^{12}	2.8×10^8
Vacancy	7.5×10^3	16	1000 Å	2.7×10^8	3.4×10^6
Carbon Atom	10		40-80	1.7×10^7	8.9×10^3

Assume: $E_{m,i} = 0.33$ eV, $E_{m,v} = 0.68$ eV, $E_{m,c} = 0.87$ eV.

$$\phi = 3 \times 10^{13} \text{ cm}^{-2} \text{ sec}^{-1}, c = 5-50 \text{ ppm}$$

z is the coordination number, ν is the atomic vibration frequency ($\sim 10^{13}$), and E_m is the migration energy. The values of 0.33 eV and 0.68 eV for migration energies were taken from the work of R. A. Johnson.⁽²⁵⁾ The results for carbon were obtained from $\Gamma = 6D/b^2$, where D was given by $D = 0.020 \exp(-0.87/kT)$ ⁽²⁶⁾ and b is the nearest neighbor distance.

An estimate of the annihilation distances during irradiation was made by using the results of Wechsler's calculations⁽²⁷⁾ for displacement production in iron (assumed thermal flux of 3×10^{13} n/cm²/sec) and fusing the calculated jump frequencies. An equilibrium concentration during irradiation of about 10^{-8} is obtained for vacancies and interstitials. This obviously is an oversimplification, which neglects trapping or losses to sinks other than annihilation, but should be adequate for the intended plausibility arguments.

Since the interstitial has the highest jump rate, it will control the

mean lifetime of both vacancies and interstitials. If the concentration of vacancies is c and the interstitial jump rate is Γ_i , then the mean lifetime of either defect will be about $1/2(c \Gamma_i)^{-1}$. The random walk migration distance is approximately $\bar{X} = \sqrt{6 D t}$. Substituting $D \approx 1/6 \Gamma b^2$ and the mean lifetime into the above relationship gives $\bar{X} \approx 0.2 b c^{-1/2}$ for the annihilation distance for interstitials and $\bar{X}_v \approx 0.2 b c^{-1/2} (\Gamma_v/\Gamma_i)$ for vacancies.

The annihilation distances of 4900 Å and 16 Å for the interstitial and vacancy should be compared with the 40-80 Å separation of carbon atoms (for 5 and 50 ppm C, all in solution.) During irradiation at 60 °C, it is obvious that the interstitial is much more likely to reach a carbon atom than a vacancy. The binding energy of a C atom to a split-interstitial is ~ 0.5 eV and to a vacancy 0.4 eV⁽²⁸⁾, so the interstitial should be the species trapped. Carbon motion can be ignored since equilibrium in the

annihilation process is reached in a few seconds.

The last two columns of Table 12.13 show the jump frequencies and migration distances for a 2 hr anneal at 350 °C. It is readily apparent that neither free interstitials or free vacancies would be present in the sample at these temperatures. Accordingly, any recovery taking place at this temperature must be due to the breakup of more complex defects. Some clustered defects undoubtedly form by the spike overlap process observed by Beeler in computer experiments,⁽²⁹⁾ but if this were the only process for complex cluster formation, it would result in equal hardening in both materials. This is not the case.

On the basis of the kinetics of defect migration and the observation of interstitial loops, it is concluded that interstitial clusters are nucleated at C atoms and grow during the irradiation. This differs from the scheme set forth by Damask and coworkers⁽³⁰⁾ in their study of radiation damage in high carbon iron. They proposed that the major reaction was the trapping of C atoms by vacancies. At the time of their work, the existence of interstitial loops stable to 500 °C was unknown, and the presence of these defects was not considered.

The identification of the defects or defect complexes causing hardening is essential to the formulation of a hardening mechanism. At this time the identification has not been satisfactorily completed. Some pertinent observations are that

hardening is observed at low fluences before defects become visible in the electron microscope ($d < 25 \text{ \AA}$) the lack of recovery below 300 °C, and the presence of prismatic loops at 500 °C after recovery of the hardness is essentially complete.

Defects smaller than $25 \text{ \AA}$ could include point defects, clusters, and loops. Extensive quenching experiments on the fcc metals have shown that individual vacancies do not contribute to the hardening.⁽³¹⁾ Recent experiments by Ono⁽³²⁾ show that free interstitials can contribute to the hardening. As shown previously, their high mobility in iron at the irradiation temperature and the absence of recovery below 300 °C should exclude them as a source of hardening. The recovery in hardness prior to the disappearance of large loops also excludes these species as major sources of hardening. Thus, clusters or small loops—most likely of interstitials—are the most likely source of hardening.

Kimura and Maddin^(31,33) have reviewed possible mechanisms for hardening from loops in quenched fcc metals. Kroupa and Hirsch⁽³⁴⁾, Friedel⁽³⁵⁾, and Fleischer⁽³⁶⁾ have proposed mechanisms that could account for hardening by loops. In the case of clustered defects, one would expect a mechanism similar to precipitation hardening⁽³⁷⁾ would be applicable. Recent computer experiments by Foreman and Makin⁽³⁸⁾ have treated random distributions of point obstacles of various strengths.

The hardening, observed in the present experiments, appears to be caused by either small clusters or unresolvable loops and is consistent with the hardening that would be produced by these defects.

One problem is that all the above mechanisms also predict a temperature dependent component to the hardening; this has been observed in radiation damage experiments on Cu.^(13,14) The rate controlling mechanism in the unirradiated iron is generally accepted as the Peierls mechanism at temperatures up to about 200 °K.⁽³⁹⁾ From 200-300 °K, another mechanism may be operative in iron but it has not been conclusively identified. If another mechanism is operative in this region, it has many properties similar to the Peierls mechanism. In fact, the whole range of thermally activated flow can be accurately described empirically with the same activation energy and Peierls stress parameters.⁽¹⁶⁾ Where two mechanisms are competitive, the one with the strongest short range stress field is controlling. Arsenault⁽¹⁸⁾ has considered the possibility of observing the Fleischer hardening mechanism in metals where the Peierls mechanism is operative and has concluded that it would be observed only in the neighborhood of T_c or above. In these experiments it was shown that T_c shifted only slightly, and this shift was not dependent on fluence. Therefore, it must be concluded that the Fleischer mechanism treated in this manner is not applicable.

A modification of the basic rate controlling process could result in

small changes in parameters, such as those produced in these experiments by radiation damage and in those produced by the quenching and aging treatments. Guyot and Dorn⁽³⁹⁾ have considered two such modifications which might occur in the Peierls mechanism, a change in the Peierls stress and a reduction of the free length of dislocation segment. As shown previously there appears to be a slight reduction in Peierls stress as determined by the empirical method but the data is limited and close to the range of experimental error. A change in the length of free dislocation segment in which a double kink could be nucleated does not appear to become important until the pinning points are closer than 20-30 b.⁽³⁹⁾ This would result in a decrease in activation volume as observed, but the uncertainty in defect concentration in the submicroscopic range makes it difficult to evaluate this possibility.

Arsenault⁽¹⁸⁾ has suggested that the changes observed in activation volume with strain may be due to the highly localized deformation in these materials. Schwink and Neuhauser⁽⁴⁰⁾ have studied this phenomena in irradiated copper and have concluded that such localized deformation leads to drastic discrepancies between the true strain rate and that calculated from cross-head velocities. This analysis seems to ignore the fact that slip is always an inhomogeneous process and that the fundamental quantities are mobile dislocation density and dislocation velocity ($\dot{\gamma} = \rho \bar{v}$). If the density

of moving dislocations remains constant, it should make no difference in strain rate whether the deformation is highly localized or uniform. This cannot be measured experimentally, but it is generally considered to remain constant after the initial multiplication at the yield point,⁽⁴¹⁾ especially in cases where there is a drop-in-load yield point.

It is difficult to distinguish between the case where the change in activation volume with strain is due to removal of defects from the slip plane by interaction and the case where the change is due to an increase in the density of dislocations as the deformation becomes less localized with strain. In the second case, one would expect the decreases in activation volume to correspond closely to the degree of localization or the coarseness of slip traces. Following this reasoning then, an increase in fluence, a decrease in deformation temperature, or the difference in hardening in the two materials should all produce a change in the activation volume as a function of strain. However, none is observed. This concept is also incapable of explaining the increase in activation volume observed in molybdenum after irradiation⁽¹⁷⁾ where the localized nature of the deformation is well documented⁽⁴²⁾ and where, because of the high density of defects, probably more pronounced than in iron.

Because the mechanism is unknown, the defect interaction hypothesis is not an entirely satisfying one either.

It does offer the advantage that only a single size of defect might influence the activation volume; this need not change with conditions that would otherwise alter the localized deformation. The annealing experiments suggest this is a small defect since the changes in activation volume anneal out before the hardness fully recovers.

From the results of these experiments and other evidence cited, the following hypothesis is offered as a model for radiation damage in iron: Hardening is produced by interstitial clusters nucleated at carbon atoms; hence, the greater hardening in Ferrovac E. Additional clusters are formed by spike overlap and will be of equal concentration in both samples. Because both types of clusters grow during irradiation, the hardness increases with fluence. There will be a spectrum of sizes produced, and the changes in activation volume are attributed to interaction with the smaller clusters. During annealing, the smallest clusters dissociate first; thus, the greater recovery in τ^* sensitive to the small clusters—and the greater recovery in τ_i at the lower fluences. Interstitials from the breakup of small clusters migrate to sinks or to larger clusters which grow during annealing. After reaching a critical size, they collapse into prismatic loops which do not contribute appreciably to the hardening. Clusters are sheared by moving dislocations and destroyed, thus leaving "channels" free of visible defects and softer than the

surrounding lattice. This leads to localized deformation which is similar in many respects to the easy glide stage of deformation in single crystals.

Summary

This investigation of the influence of neutron radiation damage on two purities of alpha iron has shown that:

- The major effect is an increase in internal stress which is attributed to small defect clusters.
- Carbon impurity atoms serve as traps for interstitials and nucleate the majority of the clusters.
- Defects in irradiated iron produce a small change in activation volume that recovers upon annealing at 350 °C. This is a modification of the basic rate controlling process rather than a new rate process.
- Activation volumes recover with strain, reflecting the interaction between defects and dislocations and the localized nature of the deformation.

ELECTRICAL RESISTIVITY STUDIES

E. R. Bradley

Before a complete understanding of the interaction between irradiation produced defects and moving dislocations can be obtained, a knowledge of the form and distribution of the defects is necessary. Electrical resistivity is a physical property of materials which is affected by point defects. An insight into the form and distribution of defects may be obtained by following the resistivity recovery

during annealing. Isochronal and isothermal recovery curves can be used to determine the temperature ranges for the thermally activated recovery processes and also the kinetics of these processes. With this information, along with mechanical properties, microscopy, and length change-lattice parameter measurements, a better understanding of the defect state and the interactions of dislocations with irradiation induced defects will be obtained.

A system for measuring electrical resistivity by the 4-point potentiometric method has been set up and is presently being checked out and evaluated, (Figure 12.20). A Guildline Instruments Ltd. potentiometer (Type 9180) is employed with a Keithley millimicrovoltmeter (Model 149) as a null-detector. The potentiometer is equipped with four ranges to measure potentials from 1×10^{-8} to 2.1010 V. Thermal compensators are used in both the sample and null-detector circuits to oppose the effect of thermal emf's in the system and to allow the more sensitive ranges of the null detector to be used. Measurements with current flow in forward and reverse directions are taken, and the average of the two measurements is used to eliminate the effect of any thermal emf's remaining in the system. Shielded wiring with a common floating ground is used throughout the system. The components are housed in a metal cabinet to reduce thermal fluctuations and to shield the components from stray magnetic fields.

Two 6 V wet cell batteries supply current for the potentiometer and sample circuits. These are heavy

FIGURE 12.20. Photograph of Electrical Resistivity Measuring System

duty automobile batteries that are enclosed in insulated compartments to minimize temperature fluctuations and improve the stability of the current supply. A fixed current of from 20 to 100 mA, depending on sample geometry, is used in the sample circuit. This current is monitored by measuring the potential drop across a 0.01Ω standard resistor.

The current has been found to vary less than 0.01% during the course of a normal measurement. The reproducibility of the system is about $\pm 0.1\%$ of measured value. Possible causes for these variations are presently being examined and hopefully will be reduced or eliminated in the near future.

HIGH PRESSURE STUDIES

G. L. Kulcinski

The purpose of this phase of the program is to study the effects of ultra high pressures (1,000 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.

Calibration of In-Reactor Diamond High Pressure Cell

The design of this cell was covered in a previous report⁽⁴³⁾ and will not be repeated here. Due to reasons

explained in that report it was necessary to increase the thickness of the sample for satisfactory X-ray analysis at high pressures. This was accomplished by placing the sample in a metallic molybdenum washer. The immediate question then arose as to the relationship between the load applied and the pressure experienced by the sample.

General Considerations

In the simplest case for Bridgman anvils without washers, one could estimate the pressure on a sample by dividing the force applied by the area on which it is acting. This assumes that the pressure is uniformly distributed over the sample. However, it has been shown in the past⁽⁴⁴⁻⁴⁹⁾ that this method of estimating the pressure does not take into consideration the pressure gradients in the sample due to friction forces. As a result, the maximum pressure in the sample may be as much as two times the average pressure as determined by the force/area (F/A) ratio. Specifically, it has been shown that the pressure varies from the center of the sample to a higher^(47,50,51) or lower^(44,45,49,52,53) value near the edge depending on whether deformable (metallic) or nondeformable (diamond) anvils are used respectively.

The introduction of metallic washer around the sample tends to reduce the pressure gradients. It has been shown by Myers et al.⁽⁴⁷⁾ that for sample diameter to thickness ratios of 15 or more, the pressure gradients are reduced to

less than 10%. Jackson⁽⁵³⁾ has treated this problem theoretically and predicts that the pressure gradient will be reduced and that the actual pressure experienced by the sample will depend on the ratio of its yield strength to that of the washer material. While the mathematical analysis of this situation is very difficult, it appears that if the ratio of yield strength is greater than one, the pressure on the sample will be greater than the average applied pressure (as determined by Force/Area). On the other hand, if the sample has a lower yield strength than the washer—a most likely situation when metal washers are used—the pressure on the sample will be less than the F/A ratio.

An experimental example of the strong sample-weak washer combination is given by McWhan.⁽⁵⁴⁾ He has calibrated his apparatus by X-ray analysis of a sample surrounded by boron epoxy discs. In this case the average pressure on the sample is 1.5, or 2 times that determined by the F/A ratio.

There are no known calibration data on the reverse situation although several groups are known to be using high yield strength metals as washers when studying low yield strength compounds and liquids under pressure.⁽⁵⁵⁻⁵⁷⁾ Since most of the work with the cell described in this paper will deal with the latter case, it was necessary to provide such data.

Theory

Let us initially consider the case of a plastic material B surrounded by an elastic material A. (Figure 12.21). Later we will allow A to become plastic and examine how this affects the pressure to the sample. The axial stress on the sample (B) within the region $0 < r < c$ is given as⁽⁵³⁾

$$\sigma_z(B) = [\sigma_z(r=c) (B)] \exp \left(2 \frac{f(c-r)}{h} \right) \quad \dots(1)$$

where $\sigma_z(r=c) (B)$ is the axial stress on an increment of sample at $r=c$,
 f is the coefficient of friction between the sample and the anvil,
 h is the thickness of the washer.

Since the radial stress, σ_r , must be continuous between region A and B and from the yield criterion at $r=c$, one finds

$$\begin{aligned} \sigma_{r=c}(A) &= \sigma_{r=c}(B) \\ &= [\sigma_z(r=c) (B) - \sigma_0(B)] \quad (2) \end{aligned}$$

where σ_0 = yield stress of sample.

By substituting the boundary condition of Equation (2) into the general equations for the radial and tangential stresses in the elastic region, $c < r < a$, one can solve for $\sigma_z(r=c) (B)$ in terms of the elastic

A - Washer Material

B - Sample

FIGURE 12.21. Geometrical Model for Theoretical Calculation of Pressure Imposed on a Sample-Washer Combination Between Bridgman Anvils

stresses in (A). Such a substitution yields⁽⁵³⁾

$$\begin{aligned} \sigma_r(A) = & \frac{2f}{3n} \sigma_z(A) \left[-r + \frac{c^3 - a^3}{c^2 - a^2} \right. \\ & + \frac{a^2 c^2}{c^2 - a^2} \frac{(a - c)}{r^2} \left. \right] \\ & + [\sigma_{z(r=c)}(B) - \sigma_0(B)] \\ & \times \left[\frac{c^2}{c^2 - a^2} - \frac{a^2 c^2}{(c^2 - a^2)r^2} \right] \quad (3) \end{aligned}$$

and

$$\begin{aligned} \sigma_\theta(A) = & \frac{2f}{3h} \sigma_z(A) \left[r + \frac{c^3 - a^3}{c^2 - a^2} - \frac{a^2 c^2}{c^2 - a^2} \right. \\ & \times \frac{(a - c)}{r^2} \left. \right] + [\sigma_{z(r=c)}(B) - \sigma_0(B)] \\ & \times \left[\frac{c^2}{c^2 - a^2} + \frac{a^2 c^2}{(c^2 - a^2)r^2} \right]. \quad (4) \end{aligned}$$

Using the octahedral stress theory which states:

$$\begin{aligned} \sigma_0^2 = & \sigma_1^2 + \sigma_2^2 + \sigma_3^2 - \sigma_1 \sigma_2 \\ & - \sigma_2 \sigma_3 - \sigma_3 \sigma_1 \quad (5) \end{aligned}$$

where $\sigma_1 = \sigma_z$

$\sigma_2 = \sigma_r$

$\sigma_3 = \sigma_\theta$

σ_0 = Yield in tension or compression, and inserting the following

values of σ_z , σ_r , and σ_θ at $r=a$ (where yielding first takes the place in A)

$$\sigma_r(A) = \sigma_1 = 0 \quad (6)$$

$$\begin{aligned} \sigma_\theta(A) = \sigma_2 = & \frac{2f}{3h} \sigma_0(A) \left[a + \frac{c^3 - a^3}{c^2 - a^2} \right. \\ & - \frac{c^2(a - c)}{c^2 - a^2} \left. \right] + [\sigma_{z(r=c)}(B) \\ & - \sigma_0(B)] \times \left[\frac{2c^2}{c^2 - a^2} \right] \quad (7) \end{aligned}$$

$$\sigma_z = \sigma_3 \quad (8)$$

one finds that

$$\frac{\sigma_{z(B) \text{ ave}}}{\sigma_T} = \left[\frac{1}{VU} (1 - t^2) + t^2 \right]^{-1} \quad (9)$$

where $\sigma_{z(B) \text{ ave}}$ is the average axial

stress in B

σ_T is the theoretical stress on the washer and sample = $\frac{\text{Load}}{\pi a^2}$

$$t = \frac{c}{a}$$

V is the factor which relates $\sigma_{z(B) \text{ ave}}$ to $\sigma_{z(r=c)}(B)$ and equals

$$1/2 \left(\frac{h}{rc} \right) \times \left[\exp\left(\frac{2fc}{h}\right) - 1 \right]$$

U is the factor relating the $\sigma_{z(r=c)}(B)$ to the yield stress of region A and equals

$$\begin{aligned} \frac{1}{K} + & \left[(1 - 2X[]_1) \pm \left\{ (1 + 2X[]_1)^2 \right. \right. \\ & \left. \left. + (4X[]_1)(1 - X[]_1) \right\}^{1/2} \right] / 2A \end{aligned}$$

$$\text{where } X = \frac{2t^2}{3h}$$

$$[]_1 = a \left[1 + \frac{2t^3 - t^2 - 1}{t^2 - 1} \right]$$

$$A = \frac{2t^2}{t^2 - 1}$$

For typical values of $f/h = 10$, $\sigma_Z(A) = \sigma_0$, $k = 8$ and $c/a = 0.5$, one finds that $\sigma_{Z \text{ ave}}(B)/\sigma_T \approx 0.50$.

This says that a sample with a yield strength $1/4$ that of its containing washer will experience $\sim 1/2$ the theoretical pressure applied.

The situation in Equation (9) can be extended to a more realistic case under high pressures, namely, by allowing the washer material A to become plastic. The ratio of actual to theoretical pressure then becomes

$$\frac{\sigma_{Z \text{ ave}}(B)}{\sigma_T} = \left[\frac{Z}{\sqrt{U}} (1 - t^2) + t^2 \right]^{-1} \quad (10)$$

where Z relates $\sigma_{A \text{ ave}}(A)$ to the yield strength of region A and is a function of a , c , h , and f .

$$Z = \frac{h}{2f} \frac{1}{a - c} \left\{ \exp\left[\frac{2f}{h}(a - c)\right] - 1 \right\} \quad (11)$$

Substituting the same values used for the previous example, one finds $\sigma_{Z \text{ ave}}(B)/\sigma_T \approx 0.45$; that is, the pressure to the sample will be slightly less than $1/2$ that determined theoretically.

While the above values are rather sensitive to f/h and k and are not to be taken as precise values, both indicate that a significant pressure reduction can occur when a weaker material is placed into a stronger washer and both are compressed to high pressures.

Experimental Procedure

In order to test the validity of Equations (9) and (10), two methods of calibration were used:

1. The change in the lattice spacing of NaCl with applied load, and
2. The shift in the optical absorption edge of TlBr with applied load.

Both methods yield information about the absolute pressure experienced by a sample contained by a washer and the second method reveals the magnitude of pressure gradients that may exist in the sample. Both of these methods yield continuous calibration data and neither suffers from hysteresis effects as do most phase transitions.

The shift in the (200) spacing of NaCl was obtained from X-rays taken of the sample under various loads in the Diamond Cell. The X-ray camera described in Reference⁽⁴³⁾ was used for these studies. These changes were then compared to the equation of state for NaCl as derived by Decker.⁽⁵⁸⁾

The specimen to film distance was calibrated by X-rays taken of NaCl at 1 bar and assuming $d_{(200)} = 2.821 \text{ \AA}$. Exposure time depended on the amount of NaCl used (size of hole in washer) and typical values varied from 4 to 16 hr with Kodak no-screen film.

Thallium bromide is known to have an optical absorption edge that progresses to the red wave lengths as pressure increases.⁽⁵⁹⁾ Thus by noting the wave length of light at which the normally transparent TlBr becomes opaque the pressure on the sample can be determined. In addition, by noting

the wave length at which absorption begins in any part of the solid and the wave length at which the whole solid is opaque, the amount of pressure gradient experienced by a sample can be estimated.

The optical absorption edge was measured by use of a Zeiss Ultraphot II microscope. A gradation filter was used to vary the wave length of light passing through the sample. The absorption edge was noted visually at 100X magnification. The sample was cycled to maximum pressure several times before calibration data was recorded.

The theoretical pressure (F/A ratio) was calculated from load deflection curves for the disc springs and the area of the smallest face diamond. The diamond face areas varied from 2.00 to $2.46 \pm 0.01 \times 10^{-3} \text{ cm}^2$. The load deflection curves for the compression of the disc springs were determined at various strain rates and it was found that increasing the strain rate from 0.025 to 0.25 cm/min had no effect on the curves.

Figure 12.22a shows the load-deflection curves for three different sets of Ti-Al-V springs and two sets of steel springs as arranged in Figure 12.23. The differences in the slopes of the titanium Spring Set #1 and that of Set #2 and 3 stems from the fact that these washer sets were machined at different times. Since the load at any deflection is proportional to the third power of the washer thickness (0.107 cm in. Figure 12.23) normal machining tolerance can have a noticeable effect on the springs load characteristics. The variation between the titanium washers and Steel Set #1 is a result

FIGURE 12.22. (a) Load Deflection Curves for Disc Spring Arrangements in Figure 12.23. (b) Theoretical Calibration Curves for Disc Spring Arrangements in Figure 12.23 Combined with Designated Set of Diamonds.

of the difference in elastic modulus, $2.07 \times 10^6 \text{ bar}$ for steel and $1.14 \times 10^6 \text{ bar}$ for Ti-6 Al-4 V. It will be noticed that the slope of the load deflection curve for steel set #2 is almost four times that of Steel Set #1. The factor of two increase in load is due to stacking two washers in series and the reduction in maximum total deflection is due to having only 4 stacks in parallel as opposed to 7 in Steel Set #1.

It should be emphasized that the curves in Figure 12.22a are for the spring arrangements and dimensions in Figure 12.23, and one could get larger

FIGURE 12.23. Arrangement of Disc Springs Inside Diamond Cell

deflection with the same maximum load or larger load at the same deflection simply by placing the springs in series or parallel, or by altering the dimensions.

Figure 12.22b combines the load-deflection curves in Figure 12.22a with the areas of the anvil diamonds to yield typical theoretical calibration curves for various combinations of springs and diamond sets. Again, these curves are characteristic only of the arrangement in Figure 12.23 and virtually any slope could be obtained by proper design of the disc springs.

The washer material used to surround the samples was 0.018 cm thick molybdenum foil prepared in the manner described earlier.⁽⁴³⁾ Hole sizes

varying from 0.0158-0.0428 cm were drilled into the washer with a high speed microdrill press.

Results

Table 12.14 shows the results obtained from X-ray analysis and Table 12.15 lists results from TlBr absorption edge studies. In the case of NaCl c/a ratios varied from ~ 0.35 to ~ 0.83 and for TlBr from 0.31 to 0.88. Theoretical pressures ranged from 10 to 106 kbars by simple rearranging the disc springs. The accuracy of both methods of calibration is assumed to be ± 1 kbar below 20 kbars and ± 2 kbars above 20 kbars.

There are two general trends revealed by both tables:

TABLE 12.14. Summary of NaCl X-Ray Data Used For Calibration of Diamond X-Ray Cell-21 °C

Load Pounds	c/a	Lattice Parameter, A ± 0.004	Pressure kbar σ_Z ave (B)	Theoretical Pressure kbar σ_T	$\frac{\sigma_Z}{\sigma_T}$ ave (B)
50	0.35	2.789	9	10	0.90
	0.535	2.804	5	10	0.50
	0.682	2.799	6	10	0.60
95	0.530	2.778	12	20	0.60
	0.682	2.789	9	20	0.45
150	0.369	2.742	23	30	0.77
	0.535	2.768	15	30	0.50
	0.775	2.778	12	30	0.40
190	0.403	2.738	25	37	0.68
	0.825	2.772	73	37	0.35
205	0.369	2.736	26	40	0.65
	0.535	2.760	17	40	0.42
	0.775	2.763	16	40	0.40
285	0.403	2.715	33	56	0.59
	0.470	2.755	19	56	0.34
360	0.403	2.711	35	73	0.48
380	0.403	2.714	34	75	0.45
	0.470	2.730	28	75	0.37
410	0.403	2.700	42	80	0.53
550	0.403	2.696	44	111	0.39

TABLE 12.15. Summary of TlBr Optical Absorption Data Used for Calibration of Diamond X-Ray Cell at 21 °C

Load Pounds	c/a	Absorption Edge - A	Pressure kbar σ_Z ave (B)	Theoretical Pressure kbar	$\frac{\sigma_Z}{\sigma_T}$ ave (B)
50	0.311	4410	9	10	0.90
	0.490	4360	8	10	0.80
95	0.350	4560	15	18	0.83
	0.490	4460	11	18	0.61
	0.875	4350	7	18	0.39
100	0.490	4490	12	20	0.60
	0.875	4380	8	20	0.40
150	0.490	4510	13	30	0.43
	0.875	4475	11	30	0.37
190	0.350	4870	27	37	0.73
	0.490	4695	20	37	0.54
	0.875	4470	11	37	0.27
205	0.490	4565	15	40	0.37
	0.875	4585	16	40	0.40
285	0.35	5090	35	56	0.62
	0.490	4820	25	56	0.45
360	0.527	4770	23	73	0.31
	0.75	4600	16	73	0.22
380	0.400	5190	39	75	0.52

- There is a significant departure from theoretically predicted pressures at the higher loads, approaching $0.2-0.5 \sigma_T$.
- There is a definite relationship between the size of the hole in the washer and the fraction of theoretical pressure on the sample. Specifically the larger value c/a has, the lower the pressure on the sample.

These two trends are amply illustrated by plotting all of the data on a load versus pressure curve in Figure

12.24. Figure 12.25 shows the same data normalized to the theoretical pressure and again the departure is quite evident.

Photographs of the optical absorption in TlBr with and without surrounding washers are shown in Figure 7.26. It is obvious that the addition of a metallic washer around the sample considerably reduces the pressure gradients. Without washers, pressure gradients of as much as 40 to 50 kbars are found; whereas, with washers, the gradients are reduced to less than 10 to 15 kbars.

FIGURE 12.24. Experimental Calibration Curve for Diamond High Pressure X-Ray Cell

FIGURE 12.25. Pressure Reduction Factor for Diamond High Pressure X-ray Cell

Discussion of the Results

The major result of these tests is the demonstration that the pressure on a powdered sample surrounded by a metal washer which is compressed between diamond anvils, is considerably lower than that predicted by the theoretical F/A ratio. This is in direct contrast to the situation encountered when surrounding the sample with a soft washer material⁽⁵⁴⁾ or with no washer at all^(44,45,49, 52,53) Figure 12.26). The factor of 2 or 3 reduction is far in excess of any experimental error and the fact

that the same results were obtained with two completely independent methods of measurement tends to confirm the phenomena.

Not only is there a reduction in pressure, but the amount of reduction depends on the size of the hole in the washer. It was shown, again with both X-ray and optical analysis, that as the hole diameter increases, so does the deviation from the theoretical pressure.

This situation is exactly what is predicted by Equation 10, as is seen in Figure 12.27. In this figure the

FIGURE 12.26. Effect of Washers on the Pressure Gradients Between Bridgman Anvils as Determined by TlBr

ordinate is the experimentally measured average pressure and the abscissa is the pressure $\sigma_z \text{ ave}^{(B)}$ calculated from Equation 10. The values of c/a , σ_T , are used from Tables 12.14 and 12.15, f/h is assumed to be 10, the ratio of the yield strength of Mo to NaCl (or TlBr) is taken as 8 and $a = 0.025 \text{ cm}$ $h = 0.0125 \text{ cm}$. These latter two values have been experimentally determined. Bridgman⁽⁶⁰⁾ finds that the shear

strength of molybdenum at 50 kbars is $\sim 12 \text{ kbars}$ and that of NaCl at 20 kbars is 1.5 kbars so that at high pressures $k = 8$ is a reasonable value. Considering the fixed values of 'a' and 'h' the assumption of $f/h = 10$ may be somewhat low because $f \approx 0.05$. However, this f refers to the coefficient of friction between the plastic NaCl or Mo and the diamond. This is a factor of two lower than the reported coefficient of friction

between a smooth diamond surface and metal at 1 atmosphere.⁽⁶¹⁾ Considering the assumptions made in the derivation of Equations (9) and (10) it is thought that the use of this order of magnitude of the friction coefficient is justified.

Figure 12.27 should be viewed only in a qualitative manner. There are too many variables, h , c , f , k , and some simplifying assumptions in the theory of plasticity, to be able to obtain more precise information. For example, it was noticed in some tests that at the higher pressures the c/a ratio increased from its initial value by some 10 to 20%. Certainly the k ratio must be changed, probably

to lower values at higher pressures and the coefficient of friction will probably increase. Time did not permit a detailed study of all of these parameters.

The apparent qualitative agreement between experimental data and theory has great significance even if exact quantitative predictions cannot be made. It not only confirms the pressure loss that most workers in the field have noticed in the past, but it gives a semiquantitative way of predicting the magnitude of this loss. For sample washer combinations with values of $k > 8$, i.e. solidified liquids and very weak solids, theory predicts that the pressure loss will be significantly more than with materials such as NaCl and TlBr. On the other hand, if combinations with values of $k < 8$ are used, i.e. metallic powders, the pressure loss should be less than ~50 to 80% found in this experiment. Furthermore, theory and experiment predict that the larger the hole in the washer, the more the actual pressure to the sample will deviate from the theoretical pressure. One can also state that the higher the theoretical pressure, the more the sample pressure will deviate from it.

With these general guidelines and given c/a , f/h , and σ_T , it is thought that one could predict the sample pressure to within 20% by using Equation (10). While this does not preclude the necessity for reporting theoretical pressures, it can cut the uncertainty by a factor of 10 or more at extremely high pressures (>50 kbars).

FIGURE 12.27. Comparison of the Pressure on the Sample as Measured Experimentally (ordinate) and Calculated from Equation 10 (abscissa)

The reduction of pressure gradients within the sample when it is surrounded by a washer was evident in both X-ray and optical calibration measurements. In the X-ray analysis the reduced gradients resulted in sharper Debye-Scherrer lines. Considering the case of TlBr, the maximum pressure gradients were 10 to 15% (Figure 12.26). This reduction should make quoted pressure values much more meaningful than those associated with large pressure gradients. The homogeneity of the pressure will also tend to reduce the shearing effects on phase transitions as experienced in a previous study with diamond cells.⁽⁶²⁾ Since our diameter to height ratios are ~ 20 , the results of this experiment agree with those of Myers et al.⁽⁴⁷⁾ who found a 5% gradient for d/h ratios of 15 or more with metallic anvils.

It should be mentioned at this point that the change in the NaCl lattice parameter was entirely continuous up to 44 kbars at 22 °C with no evidence of any phase transitions. Pistorius,⁽⁶³⁾ Lawson,⁽⁶⁴⁾ Vereshchagin⁽⁶⁵⁾ claim to have observed a phase transition at 17 kbars, the latter group by high pressure X-ray analysis. However, Jameson,⁽⁶⁶⁾ Corll,⁽⁶⁷⁾ Johnson,⁽⁶⁸⁾ and Jeffery et al.⁽⁶⁹⁾ have tried unsuccessfully to duplicate their results. Our data supports this latter group of investigators.

Conclusions

Calibration of the Diamond High Pressure Cell by X-ray analysis of NaCl and by using the pressure sensitive optical absorption edge of TlBr has demonstrated that the pressure deviates by a factor of 2 or more from

those predicted by a simple force/area ratio. The proposed ratio fcc $\rightarrow$ bcc phase transition of NaCl at 17 kbars was not observed, which is in accordance with the more recent findings of high pressure scientists.

Design of Out-of-Reactor Ultra-High Pressure Equipment

To more fully understand the effect of pressure on the damage induced in neutron irradiated solids, an out-of-reactor ultrahigh pressure laboratory was established at BNWL. This facility is designed to complement information obtained from in-reactor high pressure equipment and, in many cases, to investigate the effects of preirradiation pressure-temperature treatments on materials of interest to nuclear metallurgists. The main purpose of this section is to explain the basic design features, calibration and general capabilities of the equipment. Future reports will deal more specifically with calibration and the various programs currently underway.

General Capabilities

The basic equipment was purchased from Physical Metallurgy Corporation which is a division of ManLabs Inc. of Cambridge, Massachusetts. Figure 12.28 is a photograph of the working system and its control equipment. There are three distinct units which will be described in more detail below.

- Press (left)
- Pressure and temperature control equipment (center and right)
- Tooling (inside press)

FIGURE 12.28. Ultra-High Pressure Equipment: Press (left), Control Equipment (center and right), Tooling (inside press)

The maximum pressure and temperatures attainable depend on the type of tooling used and are listed below:

	<u>Piston Cylinder</u>	<u>Belt</u>
Typical Sample Volume	1.52 cm diam x 3.05 cm long	0.68 cm diam x 1.52 cm long
Maximum Pressure	25 kbars	60 kbars
Maximum Temperature	1500 °C+	1500 °C+

The press has been modified to allow infinitely variable rates of platen advance from zero to 0.63 cm/min. Appropriate shielding for both radioactivity and rupture hazards is placed between the operator and press.

The pressure may be controlled to 0.1% although uncertainties in calibration far exceed this value. The samples in the high pressure region are heated with small graphite resistance furnaces. A 10 kVA power supply is used to produce temperatures in excess of 1500 °C on the sample. The rate of temperature increase may be varied with an electronic programmer in conjunction with a null detecting controller. This capability allows differential thermal analysis of the samples under pressure.

It is convenient to describe each set of tooling, Belt and Piston-Cylinder separately as their pressure-temperature-sample size characteristics are somewhat different.

Piston-Cylinder

Figure 12.29 is a schematic of this set of tooling. The upper picture shows the arrangement of the tooling in the die set and press while the lower schematic shows a magnified view of the tooling. The function of the clamping plates is to maintain proper alignment of the components and good electrical contact between the bottom of the specimen cell and the backing block which contains the current lead. The die consists of an inner tungsten carbide cylinder with various steel supporting rings shrunk and press fit around it. The assembly is equipped with a cooling chamber which is necessary at high operating temperatures as the strength of the die materials is considerably reduced above a few hundred degrees centigrade.

The piston is made of tungsten carbide and is beveled at its leading edge to accommodate a triangular cross-section steel ring which prohibits extrusion of the sample. As the load is applied, the ring is forced outward to the wall and extrudes into the gap between the piston and wall. Such an arrangement is sufficient when compressing solid media but obviously not useful in compressing liquid samples in the chamber.

Figure 12.30 is a schematic of a typical specimen cell equipped with a collapsible can arrangement. Figure 12.31 is a photograph of such a

FIGURE 12.29 Schematic of Piston-Cylinder Tooling

FIGURE 12.30. Schematic of Sample Cell Assembly for Piston-Cylinder Unit.

The electrical heating circuit is as follows: from the transformer through the current lead to the bottom backing block; from the backing block the current travels through the bottom steel closure to the graphite pad and through the thin graphite furnace. The rest of the circuit is out the top pad through the piston to ground of the press. This design offers a maximum of temperature capability with a minimum application of power. Temperatures of 1500 °C can be obtained with 9 kW of energy input.

This system gets its peculiar

Figure 1 consists of 14 sequential black and white photographs arranged horizontally, illustrating the process of cutting a metal rod with a wire saw. The sequence begins on the left with a complete, thick metal rod. As the wire saw (a thin wire with a small hook at one end) moves through the rod from left to right, the rod is progressively divided into smaller segments. The intermediate stages show the rod being split into two, then four, and finally eight pieces. The final stage on the right shows the wire saw having completely cut through the rod, leaving a single wire and a small fragment of the rod. A vertical ruler is positioned on the far right of the image, providing a scale for the dimensions of the rod and the resulting pieces.

FIGURE 12.31. Exploded View of Piston-Cylinder Sample Cell Assembly.

FIGURE 12.32. Schematic of Belt Tooling

also contains an enlarged cross-sectional view of the tooling. In contrast to the Piston-Cylinder arrangement, there are two conical pistons acting on the sample. These pistons are thrust into each end of a conically shaped tungsten carbide die by the 500 ton press. The pressure is generated by the advancing pistons as they compress the specimen cell and its surrounding gaskets. Unlike the Piston-Cylinder tooling where an effort is made to prohibit extrusion of the pressure transmitting media, the lavite gasket material is allowed to extrude to its equilibrium thickness in the Belt tooling. This extruded gasket not only retains the sample but also provides some measure of support for the pistons.

The pistons themselves are usually made from tungsten carbide with various amounts of cobalt binder. It has been necessary to use 6% Co binder for the higher pressures (~ 60 kbars; whereas, 12% is sufficient below 50 kbars. The latter is somewhat more resilient at lower pressures, resulting in a much longer working life. The pistons themselves are supported by shrunk fit steel binding rings so that the stresses in the pistons are always compressive except at the very highest pressures. This is necessary because the tensile strength of WC is much lower than its compressive strength. The face diameter of the piston is 1.27 cm.

The die arrangement is similar to the Piston-Cylinder unit in that the WC die is supported by shrunk fit steel binding rings, and provisions are made for water cooling. The difference between the Belt and Piston-Cylinder is mainly the double conical shape of the die necessary to allow piston advance in the Belt system.

Figure 12.33 is a schematic of a typical specimen cell for the Belt tooling. Figure 12.34 shows an actual model and a blown-up view. The components are pointed out in Figure 12.29 and typical dimensions inside the can are: 0.56 cm diam by 1.45 cm long before compression, and 1.2 cm long (same diameter after compression. As in the case of the Piston-Cylinder unit, the specimen can volume (0.056 cc) can be used for other purposes when true hydrostaticity is not required.

The electrical heating circuit is (Figure 12.32) from the transformer through the current lead and bottom

FIGURE 12.33. Schematic of Sample Cell Assembly for Belt Tooling.

backing block to the bottom piston. From the bottom piston the current travels through the steel end pad, graphite pad, and through the furnace. The rest of the circuit is out through the top piston to the ground of the press. Typical temperatures of 1500 °C can be obtained by an input of 3 kW of power.

IN-FLUX OXIDATION AND HYDRIDING OF TITANIUM AND NIOBIUM

A. B. Johnson, Jr.

Titanium (Grade II), niobium, and niobium-1 wt% zirconium samples were exposed in the G-7 loop of the Engineering Test Reactor. Three 1-in. square specimens of each metal were

FIGURE 12.34. Exploded View of Belt sample Cell Assembly.

included in the test. Specimens of Zircaloy-2 were exposed in the same specimen holder.

The test conditions are summarized in Table 12.16. Table 12.17 summarizes the metallurgical conditions of the specimens. Out-of-reactor controls were exposed in an autoclave at Battelle-Northwest operated at 280 °C with pH-10 NH_4OH .

The activated titanium isotope (^{51}Ti) has a short half life (5.8 min) and decays by beta emission to stable ^{51}V . Impurities in the titanium (probably principally iron) account for some sustained activity. However, approximately six months after discharge, the specimen activity was ~60 mr/hr at 6 in. and the specimens could be weighed on the same out-of-cell microbalance used for pre-exposure weighings. Since the in-flux weight gains on titanium were small, the microbalance weighings were considered necessary to assess the effect of flux on oxidation. Gold interference colors were observed on in-flux and out-of-flux specimens. No attempt was made to weigh specimens of niobium and its alloy on the out-of-cell microbalance, since the in-flux weight gains were relatively large. The niobium specimens were weighed on an in-cell Mettler analytical balance.

Hydrogen samples were taken from the three in-flux titanium specimens and from one each of the niobium and Nb-1% Zr specimens. Hydrogen analyses were conducted by hot vacuum extraction.

In-flux and out-of-reactor weight gains and hydrogen pickups are summarized in Table 12.18, including

Zircaloy-2 specimens for comparison. The data show:

- Low oxidation rates for titanium in-flux and out-of-reactor. However, in-flux weight gains were 5 to 10 times higher than the out-of-reactor weight gains.
- Appreciable hydrogen absorption by titanium out-of-reactor. In-flux absorption represented 130 to 265% of the hydrogen generated by in-flux oxidation of the titanium.
- Relatively high oxidation rates for niobium and niobium-1% zirconium in-flux and out-of-reactor. Variations among identical coupons may indicate spalling of the oxide.
- No detectable in-flux hydrogen absorption by niobium or its alloy. However, pure niobium absorbed hydrogen in a 200-day out-of-reactor exposure.

Zircaloy-2 weight gains were equal in-flux and out-of-reactor. The Zircaloy in-flux weight gains were higher than those of titanium and lower than the mean in-reactor weight gains of niobium and its alloy. In-flux hydrogen absorption is comparable for Zircaloy-2 and titanium. Out-of-flux, Zircaloy absorbed hydrogen while titanium did not in 37 days.

The low weight gains for titanium in pH-10 NH_4OH at 280 °C out-of-reactor are consistent with out-of-reactor weight gains observed by Mettler and Shannon in pH-10 LiOH at 275 °C.⁽⁷⁰⁾

The high and variable weight gains on niobium are consistent with behavior observed at Battelle-Columbus on niobium in water at 316 °C.⁽⁷¹⁾

TABLE 12.16. *G-7 Loop Exposure Conditions - Quadrant 217*

Time at Temperature	37 Days
ETR Cycles	78 and 79
Time at Full Power	44 Days
Nominal Loop Pressure in Sample Section	2150 psi
Flux	5.9×10^{13} nv, >1 MeV
Integrated Flux	2.2×10^{20} nvt, >1 MeV
Water Chemistry:	
Coolant	pH-10 NH_4OH
Oxygen, ppm	<0.05
Hydrogen, cm^3/kg	45 ^(a)
NH_3 , ppm	9.1 ^(a)
NO_2 , ppm	0.01 ^(a)
NO_3 , ppm	<0.05 ^(a)
^{138}Cs , dis/min ml	4×10^4 ^(a)

(a) *Mean values*TABLE 12.17. *Metallurgical and Surface Conditions*

<u>Specimen</u>	<u>Fabrication</u>	<u>Surface Treatment</u>
Titanium (Grade II)	Initial Forging 900 °C Annealed 700 °C, 2 hr Stress Relief 480-540 °C 45 min.	Etched 90 parts H_2O 8 parts HNO_3 2 parts HF 10 min.
Niobium	Cold Rolled Stress Relief 900 °C 1 hr	Electropolished 90% H_2SO_4 10% HF 4 min., 1.5 A/cm ²
Niobium-1% Zr	Cold Rolled Stress Relief 900 °C, 1 hr	Same as Niobium

TABLE 12.18. In-Flow and Out-of-Reactor Weight Gains and Hydrogen Pickups at 280 °C pH-10 NH₄OH

Specimen	37-day Weight Gains		Δ Hydrogen Absorption			
	In-Flux	Out-of-Reactor	In-Flux ^(a)		Out-of-Reactor ^(b)	
	mg/dm ²	mg/dm ²	ppm	mg/dm ²	ppm	mg/dm ²
Titanium, Grade II	2.1	0.4	31	0.69	0	0
	2.1		30	0.67		
	2.9		28	0.62		
Niobium	11.7	78.	0	0	20	1.1
	51.2					
	60.0					
Niobium-1% Zr	41.5	52.	0	0	0	0
	13.4					
	79.9					
Zircaloy-2	11.1	11.0	27	0.5	42	0.78

(a) After 37 days in-flux.

(b) After 200 days in out-of-reactor autoclave.

The three in-flux titanium specimens all show appreciable hydrogen absorption. Movement of radiolytic hydrogen through the relatively thin oxide films appear to have been involved, since the absorbed hydrogen exceeded that generated by corrosion in all cases.

Conclusions regarding the in-flux behavior of titanium and niobium will be reserved until results of a 4 cycle G-7 loop exposure are available.

PRESSURE-VESSEL NEUTRON DOSIMETRY

A. DePino, Jr. and R. E. Dahl

We are investigating the possibility of using fission-track detectors as a fluence monitoring technique for pressure vessel surveillance. Glass, mica and plastic samples have been irradiated in test facilities in the Hanford K-Reactor to determine the

limiting exposure level at which the detectors become useless due to radiation damage. Plastics were found to be unsuitable for use at expected pressure vessel fluences. Fast-neutron irradiation damage limited the use of the micas investigated to fluences less than $\sim 5 \times 10^{15}$ n/cm², $E > 1$ MeV, (about 10 yr at the surface of a typical pressure vessel). Above this exposure, optical microscopy methods could not be used successfully.

Mica and glass samples were coated with a thin layer of highly depleted uranium and irradiated. This technique appeared to be an attractive way to monitor fast neutron and to minimize the tracks formed by thermal neutron-induced fission of ²³⁵U. This coating technique yielded satisfactory results only at fluences less than $\sim 10^{15}$ n/cm²

($E > 1$ MeV). Therefore, coating fissionable material on mica and glass is not suitable for pressure vessel applications.

Natural mica is a good candidate for a detector material within its exposure limits. Fission fragments are registered with 100% efficiency in mica, and processing techniques, including etching and data analysis, have been established. However, extension of the fluence range will require a more radiation resistant material. To determine whether the accuracy of the method will meet pressure vessel requirements, further investigation is required for detector calibration, determination of the reproducibility of the fission tracks, and effects on nonhomogeneous distribution of the uranium in the material.

The results of this work have provided a sound basis for further work. We have discussed the use of fission-track dosimetry with Washington-AEC personnel and will propose further work in a research proposal.

INTERCALIBRATION TEST OF COUNTING LABORATORIES

R. E. Dahl, J. A. Ulseth

The Irradiation Standards and Procedures Committee of the Irradiation Effects on Reactor Structural Materials Program has recommended that another intercalibration test be initiated of counting laboratories utilized by program members. The purpose of this intercalibration test is to insure that all participating counting laboratories are consistent in both their determination of monitor activities and conversion of these activities to fluence.

This test is being conducted in a manner similar to the previous intercalibration test.⁽⁷²⁾ Two monitors of each type (Fe, Ni, Ti and Al-0.1% Co) have been irradiated, counted and distributed to the participating laboratories by Pacific Northwest Laboratory (PNL).

Monitor materials were irradiated in the MTR Rabbit Facility to obtain two levels of activity differing by a factor of about 100. Half of the sets were irradiated for 2 hr and the other half for 196 hr. Each participating laboratory has been asked to report total activity (d/s), specific activity (d/s/mg) corrected to discharge date, and total fluence for the monitors received. All data will be returned to PNL for tabulation and comparison.

The eleven participating laboratories are: Argonne National Laboratory; Atomics International; Battelle Memorial Institute, Columbus, Ohio; Brookhaven National Laboratory, General Atomic, General Electric Company (NMPO) Cincinnati; General Electric Company (APED) San Jose; Idaho Nuclear Inc.; Oak Ridge National Laboratory, Pacific Northwest Laboratory, Richland, Washington; and Isochem, Richland, Washington (Isochem radiochem. lab. is utilized by PNL to analyze many dosimeters).

FAST REACTOR SUPPORTING STUDIES

FAST NEUTRON MECHANISMS

R. P. Allen

The objective of this program is to determine the effects of fast neutron irradiation and in-core environmental and service conditions on fundamental

aspects of hydrogen and helium behavior in fast reactor alloys. The probable neutronic hydrogen retention and distribution characteristics of representative iron- and nickel-base alloys are currently being evaluated as the initial part of a study of synergistic damage interactions involving hydrogen, helium, and the irradiation induced defect state.

Permeation Barrier Effects

The hydrogen generated in endothermically occluding metallic core components by (n, p) nuclear transmutation reactions will be quite mobile at normal fast reactor operating temperatures and after an initial transient period will effuse into the surrounding coolant at a rate equal to the neutronic hydrogen production rate. The resulting maximum neutronic-hydrogen concentration (C_m) in a particular component is a function solely of the hydrogen production rate (R) and hydrogen diffusivity (D_m) in the component material, if diffusion through the component is rate controlling for the effusion process. Thus, for an approximately planar component of thickness L with desorption occurring at both surfaces,

$$C_m = \frac{RL^2}{8D_m} \quad (1)$$

Calculated values of C_m for representative temperatures, component dimensions, and hydrogen production rate⁽⁷³⁾ are shown in Figure 12.35 for 18-8 type stainless steel.

If a desorption or effusion mechanism, rather than diffusion, is rate controlling, then the concentrations calculated using Equation (1) are no longer

FIGURE 12.35. Calculated Maximum Neutronic Hydrogen Concentrations for Ideal Effusion Conditions

applicable, but represent a lower bound for the retention of the neutronic hydrogen, with the condition of complete hydrogen retention as an absolute upper bound. This can be illustrated by considering the effect of surface permeation-barrier formation on expected concentration values. At equilibrium, the permeation rate through a surface region must equal the corresponding total internal hydrogen production rate; for a planar component with barrier regions of thickness T at both desorption surfaces:

$$C_s = \frac{RL^2}{8D_m} \left[1 + 4 \frac{D_m}{L} \cdot \frac{KT}{D_s} \right] \quad (2)$$

where C_s is the maximum internal neutronic hydrogen concentration for a surface permeation-barrier controlled effusion process; D_s is the hydrogen diffusivity in the barrier material;

and K is the ratio of the unit pressure hydrogen solubility in the component material to that in the altered region. The magnitude of C_s is thus a function of the hydrogen solubility as well as the hydrogen diffusivity characteristics of the barrier region.

In evaluating C_s , it is not necessary to know or separately determine K , T , or D_s . If P_m is the experimentally determined hydrogen permeation rate through a planar component with an ideal effusion surface; and P_s is the reduced permeation rate for identical conditions and component dimensions, but an altered effusion surface, then:

$$P_m/P_s = 1 + \frac{D_m}{L} \cdot \frac{KT}{D_s} \quad (3)$$

Combining this with Equation (2)

gives, for $P_m/P_s \gg 1$,

$$C_s/C_m = 4 P_m/P_s \quad (4)$$

Relative permeation rate data for alloys, surface conditions, and temperatures of interest are limited; those available are primarily for surface oxide films or layers on selected iron-base alloys, for which permeation rate reductions of up to three orders of magnitude have been observed.^(74,75) Specific permeation studies have, therefore, been initiated to determine the effect of complex surface carbide and nitride layers on relative permeation rates and hydrogen desorption behavior in 18-8 type stainless steels. The permeation facility, used, incorporates a mass spectrometer to permit the additional detection and identification of desorbing hydrogenous compounds.

The effect of constant but nonideal effusion conditions on the duration of the initial transient period and on the corresponding time-dependent maximum neutronic hydrogen concentrations is illustrated in Figure 12.36 for an assumed P_m/P_s ratio of 25. The approximate time required to reach the maximum steady-state concentration can be obtained by dividing this concentration by the hydrogen production rate; similarly, for times less than this, the maximum transient concentration is given by the integrated hydrogen production rate, which is equivalent to a condition of complete hydrogen retention.

FIGURE 12.36. Calculated Time-Dependent Maximum Neutronic Hydrogen Concentrations for Representative Effusion Conditions

EBR-II IRRADIATIONS

J. J. Holmes, R. E. Robbins, B. Mastel,
J. L. Brimhall, R. W. Barker

The objective of this program is to determine the combined effects of environment and fast reactor irradiation on the mechanical properties of candidate fast reactor cladding and structural alloys. The program currently in progress is specifically directed at providing a basis for selection of optimum alloys for FFTF applications and at providing a description of material behavior in fast reactor service.

A detailed irradiation program for the EBR-II has been formulated. This effort involves the use of two full subassemblies in the EBR-II—a single Row 2 "B-7" irradiation pin and various space-available A-19 irradiation pins in the reactor core and blanket.

Program emphasis is on creep and tensile properties of austenitic stainless steels in sodium after fast reactor irradiation. Specimens are to be irradiated in the EBR-II to fluence levels of 1×10^{22} , 3×10^{22} and 1×10^{23} and subsequently tested at temperatures from 700 to 1500 °F (370 to 815 °C) at stresses sufficient to cause rupture in time from 1 to 10^4 hr. In addition to postirradiation creep testing, postirradiation tensile testing is also planned along with studies on flowing sodium effects, aging response, and in-reactor creep.

The major portion of the fast reactor irradiations is to be conducted in the two subassemblies. One of the subassemblies will be irradiated to

1×10^{23} total fluence in a Row 2 position, the other in Row 7 will be used to accumulate dose levels of 1×10^{22} and 3×10^{22} . Each subassembly contains nearly identical sets of specimens encapsulated in gamma heated subcapsules designed to operate at three temperatures; namely, 900, 1200, and 1400 °F (482, 649, and 760 °C). Both units will contain mostly 304 and 316 tensile, biaxial creep, and uniaxial creep specimens; but backup FFTF alloys such as 348 and 321 are also included as well as Hastelloy X-280, Inconel 600, Incoloy 800, Inconel 718, Inconel 625, and Inconel X-750. Both subassemblies have been fabricated and placed in the EBR-II for irradiation. It is anticipated that the Row 7 subassembly will be returned to PNL in October.

Comparison of irradiation effects occurring in sodium with those occurring in helium will be evaluated by conducting a limited number of tests in helium and in reactor sodium at ambient temperatures. A MK-B-7 irradiation pin in Row 2 containing 198 miniature specimens in a helium environment, is currently being irradiated to a target fluence of 1×10^{23} . The specimens in this pin are primarily AISI 304 and 316; some 348 and 321 specimens are also included. Twelve MK-A type space-available pins have been charged in the EBR-II core. Subassembly X-019 contains eight of these pins and is scheduled for an exposure of 3.0×10^{22} fast fluence. Subassembly X-020 contains four pins and is scheduled for an exposure of 6.0×10^{22} fast fluence. These pins

contain 124 tensile and creep test specimens of AISI 304 and 316 SS, evenly divided, of which half are exposed to bulk sodium.

Initial tensile test results on annealed AISI 304 SS irradiated at 1000 °F (540 °C) to about 1.7×10^{22} total flux in the EBR-II have been obtained. Tensile properties of annealed 304 SS irradiated in the EBR-II are presented in Figures 12.37 through 12.40 together with "as-received" control data.

The following general characteristics were observed:

- (a) Fast reactor irradiation produces an increase yield strength at all test temperatures in the range of 572 to 1600 °F (300 to 870 °C). Largest increases in yield strength occur at temperatures below 900 °F (480 °C) where a relatively constant increase of a factor of two is observed. Above 900 °F (480 °C) the increase in yield strength decreases monotonically to about 120% of the as-received condition at 1500 °F (815 °C). (Figure 12.37)
- (b) The ultimate strength is not appreciably changed by fast reactor irradiation except at room temperature where a 15% increase is found. (Figure 12.38)
- (c) Uniform elongation was decreased by about a factor of two at temperatures below 900 °F (480 °C). Between 900 and 1500 °F (480 and 815 °C) uniform ductility decreases monotonically with increasing temperatures from about 20% to 0.6% at 1500 °F (815 °C) but increases to 3.2% at 1600 °F

(870 °C). As-received material exhibits a similar decrease above 900 °F (480 °C) but obtains a minimum of about 12%. Total elongation follows the same general trends as uniform elongation; however, minimum ductility occurs at 1300 °F (705 °C). Uniform elongation in as-received material is nearly athermal. These characteristics are shown in Figures 12.39 and 12.40.

Irradiation damage to metals has been described in the literature in terms of two distinct behavior modes⁽⁷⁶⁾ operating in well-defined temperature ranges. Displacement damage is thought to cause increases in strength properties and losses in ductility at temperatures below one-half the absolute melting temperature ($T_m/2$). Above $T_m/2$ displacement damage recovers so rapidly that appreciable damage concentrations are not expected to build up during irradiation so strength and ductility are not affected. At temperatures greater than $T_m/2$, however, a different form of irradiation damage occurs which involves transmutation effects and drastically reduces ductility by weakening grain boundaries. Barnes⁽⁷⁷⁾ has suggested that the transmutation-induced grain boundary weakening is due to helium at grain boundaries from (n,α) reactions.

The recently-tested EBR-II specimens were irradiated at 1000 °F (535 °C) which is $0.501 T_m$, yet a significant increase in yield strength was observed at test temperatures as high as 1500 °F (815 °C) which is $0.67 T_m$.

FIGURE 12.37. Comparison of 0.2% Off-set Yield Strength in Annealed and EBR-II Irradiated AISI 304 SS.

FIGURE 12.38. Ultimate Strength of AISI 304 in the Annealed Condition and After Irradiation in the EBR-II to 1.7×10^{22} Total Fluence

FIGURE 12.39. Uniform Elongation of Annealed AISI 304 Before and After Irradiation in the EBR-II.

FIGURE 12.40. Total Elongation of Annealed AISI 304 Before and After Irradiation in the EBR-II to 1.7×10^{22} Total Fluence.

This is in marked contrast to classical behavior and indicates that either high exposure or high dose rate in the EBR-II produces a damage state that is relatively stable at temperatures in excess of $T_m/2$. Recent data on AISI 304 SS irradiated to 6×10^{21} (>1 MeV) in the ETR also show hardening persisting to temperatures well in excess of $T_m/2$ indicating that exposure, rather than exposure rate, produces the temperature-resistant defect state.⁽⁷⁸⁾

Thin film electron microscopy has been employed to clarify the nature of damage state in EBR-II irradiated specimens. Examination of foils in the as-irradiated condition and foils given an anneal at 900 °F (480 °C) revealed the following. The damage consists of two components: (1) dislocation loops 500 to 1000 Å in size containing stacking faults and identified as Frank sessile loops and (2) polyhedral voids or gas-filled cavities about 200 Å in size. (Figures 12.41 and 12.42) The number of point defects comprising the stacking fault loops was calculated and compared with the number of point defects comprising the voids. These two numbers were equal within experimental error. This is consistent with a model in which the interstitial atoms form the loops and the vacancies form the voids. Subsequent annealing at 1400 and 1500 °F (760 and 815 °C) for 1 hr completely removed the Frank sessile loops and greatly reduced the size and number of voids. At 1500 °F (815 °C) the voids were almost entirely absent from the structure.

The absence of any observable defect state at 1400 and 1500 °F (760 and 815 °C) suggests the primary

cause of hardening is not either the large Frank loops or voids but some unresolvable defect. The nature of this defect is not obvious at this time but must be considerably different than that thought to cause hardening at low temperatures. The dose dependence of high temperature hardening and indicated lack of dose rate effects suggests that some transmuted species such as helium is stabilizing displacement damage.

Electron microscopy studies have been supplemented with optical examination of fractured specimens. The fracture mode below 900 °F (480 °C) is entirely transgranular while intergranular fracture predominates above 1100 °F (600 °C). The intergranular fractures below 1500 °F (815 °C), while giving the general appearance of wedge fractures, do not start at triple points but instead are initiated away from the grain corners and propagate to them giving the appearance of wedge fracture. Below 1500 °F (815 °C) the cracks appear to propagate easily and are typically sharp pointed. At 1500 °F (815 °C) intergranular fracture is by cavitation and shows typical voids along the boundary and rounded crack tips.

It should be pointed out that premature grain boundary failure does not appear to be the cause of low uniform ductility in the EBR-II irradiations. Radiation hardening to the extent that the yield approaches the ultimate strength is sufficient for low uniform ductility (Figure 12.43). This is true regardless of any considerations of grain boundary effects. The nonuniform ductility, however, may be decreased by premature grain boundary

FIGURE 12.41. Annealed AISI 304 SS Irradiated to 1.7×10^{22} Total Neutron Fluence at 1000 °F (535 °C) in the EBR-II. (Electron micrograph shows (111) frank loops and polyhedral voids)

150,000X

FIGURE 12.42. Annealed AISI 304 SS Irradiated to 1.7×10^{22} Total Neutron Fluence at 1000 °F (535 °C) in the EBR-II. (Polyhedral activities possibly containing some gas phase are believed to precipitate. These cavities are almost entirely removed after annealing for 1 hr at 1500 °C (815 °C))

150,000X

FIGURE 12.43. Tensile Properties of AISI 304 SS after Irradiation in the EBR-II to 1.7×10^{22} Total Fluence

failure. Additional studies are needed to clarify this point.

LIQUID METAL CAPSULE IRRADIATIONS

K. R. Wheeler

This program is designed to provide high-temperature irradiations of candidate LMFBR program structural materials in flowing liquid metal environments. From these capsules will come information concerning neutron damage to structures and moving sodium-metal interactions, important to corrosion and mass transfer studies. In addition, the thermal stability of the various alloys at elevated temperature will be studied.

Liquid metal capsule GEH-22-3, containing 32 AISI 316 SS tensile samples, was discharged during shutdown of ETR Cycle 87. The capsule accumulated a fast fluence ($E > 1 \text{ MeV}$) of 1.38×10^{21} after 99.9 full power days of operation. One thermocouple located in the central sodium column functioned throughout the entire operating period indicating a sodium temperature of 1000°F (537°C). Based upon millivolt readings of the other capsule thermocouples prior to their loss, the peak sodium temperature was approximately 100°F (38°C) hotter than the continuous 1000°F (537°C) reading described above. The capsule has been returned to BNWL for disassembly and specimen retrieval.

A fourth generation liquid metal capsule GEH-22-4, containing a total of 40 AISI 304, 316, and 348 SS tensile samples, began an irradiation in ETR Cycle 87. A design modification increased the radial heat transfer path displacing the temperature control

range upward to realize a control point at 1400°F (760°C). As the curve of Figure 12.44 shows, the desired control point was achieved at a 50 Ar-50 He gas mixture. Other design changes should extend capsule life by removing restraints on thermocouple sheathing and the vacuum tube, these restraints producing severe stressing of components during differential thermal expansion. Figure 12.45 shows the tubular connectors between the outer and inner bulkheads. These connectors support the sodium chamber and provide stress-free exit paths for the thermocouples. The curved tube with tube fitting is the sodium filling port. Figure 12.46 presents an exploded view of the complete capsule including the specimen assembly.

GEH-22-4 has completed 15 full-power days of stable operation, with all thermocouples operating normally. The capsule will continue to increase its fluence level in future ETR cycles with discharge dependent upon capsule performance.

EXPERIMENTAL TESTING FACILITIES

R. H. Todd, J. A. Yount

The comprehensive investigation of the effects of irradiation and environment upon reactor structural metals and fuel cladding candidate materials requires a testing program and a facility which will accommodate numerous materials and operating variables. The testing conditions under consideration, which range up to 1400°F (760°C) in sodium, present environments in which standard materials have not been fully evaluated. The facility which will fill the need for

FIGURE 12.44. Temperature Versus Gas Composition - Liquid Metal Temperature Controlled Irradiation Capsule.

*FIGURE 12.45. GEH 22-4 Sodium Chamber
Suspension and T.C. Feed Throughs.*

*FIGURE 12.46. GEH 22-4 Sodium Capsule
Exploded View*

testing irradiated samples in sodium and controlled atmospheres is now being built. The assigned building has been improved to the point where the office spaces and workshop can be occupied. This portion of the work was completed on February 7 of this year. Closer supervision and coordination of the work is now possible with the offices and workspaces in close proximity of one another. The workshop is being equipped with the special and standard tools necessary for work with the test stands to be used in this building. Renovation of the second portion of this building, which will include the ventilation and exhaust system, will be done under a separate project proposal which is now awaiting AEC approval.

Four test stands from the CANEL equipment pool have been placed in the workshop. These units have undergone complete overhaul and cleanup, and the associated instruments and controllers

have been standardized. Except for conversion of the controller-recorders to chromel-alumel thermocouple signal input, these four units are ready to begin working. By placing these units in service at the earliest possible date, the data needed for FFTF design criteria will not be delayed as it would be if it were necessary to wait for the completion of the building improvement project. The balance of the CANEL test stands has been moved in to the 300 Area to Building 306, where they will be cleaned, overhauled, and checked. As this work is completed, the test stands will be moved to Building 321-A.

The prototype cell has been moved into the building and the lead brick put in place. The lead glass window has been delivered and was on hand for timely installation. The cell is now nearly complete. Some work is being held up because of lack of support facilities which are to be

provided by the building improvement project. A radiation leak check has been conducted which indicated weak points around the door and the loading port. The changes indicated have been made. A final radiation leak check will be made after all work has been completed on the cell and before it is accepted for operation. The first use of the cell will be in remote testing of the test stand, which are to be used in this cell and in the sodium loop facility. Design on these test stands cannot be considered complete until the complete test stand assembly has performed satisfactorily under remote conditions. Assembly and disassembly must be satisfactorily accomplished with master-slave manipulators. No radioactive materials will be introduced to the remote cell until these equipment tests have been completed. The test units which will operate in the prototype cell will require a vacuum system to attain the desired control of environment. The system is now being designed and will provide a vacuum manifold for the five test stands planned initially with capability for serving additional test stands later. The requirement that the manifold shall serve additional test stands at a later date does not increase the cost of providing adequate vacuum to the original five units. As reported in the last quarterly, the creep test rigs have been redesigned with a consequent reduction in size which makes it possible to install 15 test units where only 5 units of the original model could have been accommodated.

Initial installation will be limited to five units, but these will conform to a design which will allow addition of other test units whenever the additional units should be authorized.

The sodium loop which will provide high-temperature flowing sodium testing environments has been rescheduled for shipment from the manufacturer's plant. Shipping date has been set for April 5; the delay has been caused by late delivery of essential valves to the manufacturer. Design of the special creep test apparatus to be used in the sodium loop is complete. A prototype unit is being made up for use in performance tests which will be conducted first in the prototype cell, then in the nonradioactive test segment of the flowing sodium loop. Final acceptance of the design will hinge upon the satisfactory performance of the prototype unit in these tests. The test unit design for service in sodium features smooth-surfaced parts with shoulder-on-flange connections wherever possible rather than threaded connections. The sample loading system is only one-half the height of the original working model, and the extensometry has been moved outside the environmental chamber. The total result is that the overall height of the test unit is approximately half of the first working model devised; and the total cost, parts and machining costs, is also half of that for the original model. The overall simplifications and reduction in size of these test units make possible a considerable reduction in the size and cost of the shielding which will be built around

the test units for irradiated specimens. Design of the support piping and shielding for this portion of the recirculating sodium loop has progressed along with the design of the test stand. The scoping design package for this portion of the facility is 85% complete. It will be turned over to Laboratory Engineering Unit for integration into the overall building improvement project and actual construction.

A special dispenser for high purity liquid sodium has been devised for transferring precisely measured quantities of sodium from storage barrels to enclosed capsules or other containers. The receiving containers must be closed and fitted with valves and Swagelok fittings for vacuum, inert gas, and liquid sodium to prevent contamination of the sodium by the room atmosphere. This dispenser will adapt directly to an inert atmosphere glove box and can provide measured volumes of sodium with an estimated accuracy of $\pm 1.5 \text{ cm}^3$. Final assembly of this dispenser is now in progress.

The need for inert atmosphere facilities in which alkali metals can be stored, transferred, sealed in containers and cleaned from tools and containers has been recognized; and funds have been appropriated for two pieces of equipment. One will be a high purity inert atmosphere glove box in which test capsules can be loaded and sealed with an arc weld without contaminating the high purity alkali metal. The other will be an explosion-proof glove box with a low grade inert atmosphere and a steam

nozzle for cleaning equipment of any residual alkali metal. Specifications for these items of equipment are being written.

An investigation into the potential fire hazards of sodium and other alkali metals turned up the uncomfortable fact that there is no adequate fire fighting clothing now available. The materials normally used for protective personnel cover in other fire situations have been shown to be dangerous when exposed to alkali metal fires. An example of this is aluminized asbestos, which flares and burns when hit with burning NaK. A search for suitable materials has turned up two which may be used together to make protective blankets and clothing. The outer layer would consist of "pluton" cloth which is made of fibrous graphite. This would be backed up with an insulating layer of one of the several forms of Fiberfrax, a product of the Carborundum Company. Both of these materials have an extremely high-service temperature (above 2300 °F) and are known to be nonreactive when exposed to burning NaK. The graphite cloth retains some strength after direct contact but has essentially no insulating properties. The Fiberfrax material has excellent insulating properties; but, in low-density form, it cannot withstand direct contact with burning NaK. It is hoped that a suitable composite can be made up from these two basic materials. Technical personnel of the Mechanical Metallurgy Unit are working closely with the Safety Engineers for PNL and Atomics International on the development of this material.

RADIATION EFFECTS ON METALS - 05

PROGRAM - T. K. Bierlein, E. R. Bradley, J. L. Brimhall, H. E. Kissinger, G. L. Kulcinski, B. Mastel, and F. A. Smidt, Jr.

This program is concerned with establishing the combined effect of impurities and neutron radiation on the properties and structure of specific metals. Conditions during radiation of molybdenum, nickel, and rhenium are being varied to provide information on higher neutron flux, higher integrated exposure, and higher irradiation temperature. Postirradiation studies include thermally activated recovery of radiation induced changes.

Molybdenum

Foils of molybdenum have been annealed at an equivalent time and temperature that simulates the irradiation history of the molybdenum irradiated at 600 °C. This includes both unirradiated material and samples irradiated at 60 °C. The purpose of this experiment was to (1) compare the microstructure of samples annealed after low temperature irradiation with samples irradiated at elevated temperatures; (2) determine if any carbide precipitation could occur during the long time irradiation.

The unirradiated high purity molybdenum samples showed no apparent carbide formation after the long anneal. In the carbon-bearing molybdenum, carbides were present before the anneal, and no obvious precipitation of new carbides was found. Growth of existing carbides could have occurred, but this was not investigated quantitatively. This supports the conclusion that the

defects seen after irradiation are formed from vacancies and interstitial molybdenum atoms and are not carbides.

In the samples irradiated at 60 °C and annealed at 600 °C, there are some significant differences compared to samples irradiated at 600 °C. No denuding of the defects near grain boundaries occurs during postirradiation annealing of specimens irradiated at 60 °C (Figure 12.47). The average size of loops is smaller, 200 to 300 Å, although a few loops are as large as 1000 Å. The density of loops is significantly greater. In agreement with the results on samples irradiated at an elevated temperature, all measurable loops are interstitial in character. These observations point out that elevated temperature irradiation damage cannot be reproduced by annealing a material irradiated at a low temperature. Similar samples of molybdenum with carbon remain to be investigated.

Molybdenum, irradiated at 600 °C and given the further long anneal at 600 °C, showed no unexpected behavior. The microstructure was similar to samples annealed at 800 °C for short times. It is significant to note that the denuded zone did not increase during annealing.

In the carbon-bearing molybdenum, irradiated at 600 °C and annealed at 600 °C, direct observation of the glide motion of a small cluster was noted. Figure 12.48 shows the movement of a cluster after several minutes in the microscope while under observation. The motion was rapid and jerky, indicative of glide, and the direction was consistent with glide in the $\langle 111 \rangle$ direction. The driving force probably arose from

Neg 4633C

65,000X

FIGURE 12.47. Absence of Denuding of Defect Clusters in Molybdenum Irradiated at 60 °C to a Fluence of 5×10^{19} n/cm² and Annealed at 600 °C.

FIGURE 12.48. Sequential Micrographs Showing Motion of Cluster while under Observation in the Electron Microscope. (A portion of MO₂C is at upper left)

the differential heating of the matrix and nearby carbide. Whether this could occur in a bulk sample annealed or irradiated at elevated temperatures is not known. However, this may explain the increased concentration of clusters inside the large interstitial loops in samples irradiated at elevated temperatures.

Isothermal annealing studies of the elevated temperature irradiations are in progress.

Nickel

Nickel, irradiated to 1.7×10^{19} n/cm², has been annealed at temperatures up to 500 °C. The density of clusters and loops has decreased from approximately 2×10^{16} /cm³ to 4×10^{15} /cm³

after annealing at 500 °C. The average size of the loops has increased upon annealing. It is significant that no interaction of the loops to form nodes has occurred during annealing. From many observations of postirradiation annealing behavior of nickel, it appears that nodes cannot be generated by postirradiation annealing but only during irradiation. Although loop growth occurs during annealing, the concurrent decrease in density prevents any loop interaction. Specimens irradiated to fluences as high as 4×10^{19} n/cm² are available and will be examined.

During the investigation of the irradiated nickel, a significantly

higher defect concentration was observed at coherent twin boundaries as compared to incoherent twin boundaries and normal grain boundaries (Figure 12.49). This phenomenon is considered to be direct evidence for focusing collisions and channeling. The boundary interrupts the focusing event; this causes release of energy and, hence, displaced atoms. They remain in the vicinity of the coherent twin boundary because this cannot act as a sink for the point defects. A more complete analysis of these results has been written and submitted to the Journal of Applied Physics for publication.

The annealing of irradiation-induced defects in molybdenum single crystals is being studied by isothermal and isochronal annealing experiments. By observing changes in X-ray lattice parameter and sample dimensions during annealing, it is possible to make some quantitative estimates of the numbers and types of defects involved in each recovery stage.

Measurements of stored energy release during the recovery stages would provide additional information about the particular reactions involved. To this end, an apparatus has been assembled for use with specimens identical in shape and size to the rod samples used for X-ray and length-change measurements. This stored-energy calorimeter utilizes the differential thermocouple principle. An active sample assembly and a reference sample assembly provide a differential emf as both samples are heated in the same furnace. The differential emf provides a measure of the energy released or absorbed by the reacting sample.

Each sample assembly consists of three chromel-alumel thermocouple

junctions connected in cascade, so that the total output is the sum of the three individual emf's. The output of the reference sample assembly is opposed to that of the active sample in such a way that the net output to the detector is the difference between the two. The six junctions are arranged in a 3/4 in. diam circle. Samples are 1/8 by 1 1/2 in. rods having a small hole bored in one end to fit over the thermocouple junction.

Evaluation experiments have been of two types. In the first experiment, 0.017 g of pure lead was placed in small cavities drilled in three annealed copper rods weighing 8 g. The latent heat of melting of this amount of lead is 0.1 cal, which results in a heat absorption of 0.012 cal/g Cu upon melting. The apparatus detected this heat absorption readily. The second experiment utilized copper samples which had been severely cold-worked. These samples showed a heat evolution at 250 to 300 °C, the magnitude of the effect being about 10 times the deflection produced by the melting lead in the first experiment. This indicated a heat evolution of roughly 0.12 cal/g during the recrystallization of the copper; this value is in agreement with those reported by Clarebrough et al.⁽¹⁾

The capability of the calorimeter to have sufficient sensitivity to measure energy release in very small specimens has been demonstrated satisfactorily. Samples of single-crystal and polycrystal molybdenum and nickel have been prepared and submitted for irradiation. Until these samples are available, additional

1. L. M. Clarebrough, M. E. Hargreaves, and G. W. West. "The Release of Energy during Annealing of Deformed Metals," *Proc. Roy. Soc. (London)* **A232**, p. 252. 1955.

FIGURE 12.49. Direct Experimental Verification of Localized Displacement Damage Due to Focussing and Channeling at a Twin Boundary in Nickel Irradiated to a Neutron Fluence of 2×10^{19} nvt ($E > 1$ MeV). A Designates the Coherent Twin Boundary, and B Designates the Incoherent Boundary.

calibration experiments will be performed, and some preliminary work will be done on irradiated molybdenum and iron samples now on hand.

Isobaric-Isochronal Annealing Experiment

The objects of this program are:

- To measure activation volumes of migration for Stage III-IV annealing in molybdenum
- To determine the effect of post-irradiation compression on clustering of point defects
- To determine the effect of simultaneous irradiation and compression on point defects in molybdenum.

Initial samples for this study have been irradiated to approximately 5×10^{18} n/cm² (fast) and are currently being postirradiation characterized as to electrical resistivity, hardness, lattice parameter change, and microscopic structure. An unknown cause of smearable contamination picked up during irradiation has made these measurements most difficult.

A working model of the in-reactor hydrostatic high pressure cell has been successfully operating at 17 kbars for over a month in an out-of-reactor test. No noticeable diminution of pressure has been detected. The test will continue for another month.

Design of Out-of-Reactor Ultrahigh Pressure Equipment

To more fully understand the effect of pressure on the damage induced in neutron irradiated solids, an out-of-reactor ultrahigh pressure facility was established at BNW. This facility is designed to complement information obtained from in-reactor

high pressure equipment and, in many cases, to investigate the effects of preirradiation pressure-temperature treatments on materials of interest to nuclear metallurgists. The main purpose of this section is to explain the basic design features, calibration, and general capabilities of the equipment. Future reports will deal more specifically with calibration and progress on the various programs currently underway.

General Capabilities

The basic equipment was purchased from PhysMet Corporation which is a division of ManLabs Inc. of Cambridge, Massachusetts. Figure 12.50 is a photograph of the working system and its control equipment.

The maximum pressure and temperatures attainable depend on the type of tooling used and are listed below:

	Piston Cylinder	Belt
Typical Sample Volume	1.52 cm diam x 3.05 cm long	0.68 cm diam x 1.52 cm long
Maximum Pressure	25 kbars	60 kbars
Maximum Temperature	1500 °C +	1500 °C +

The press has been modified to allow infinitely variable rates of platen advance from zero to 0.63 cm/min. Appropriate shielding for both radioactivity and rupture hazards is placed between the operator and press.

The pressure may be controlled to 0.1% although uncertainties in calibration far exceed this value. The samples in the high pressure region are heated resistively with small graphite furnaces. A 10 kVA power supply is

FIGURE 12.50. Ultra High Pressure-Temperature Equipment; Press (left), Control Equipment (center and right), Tooling (inside press).

used to produce temperatures in excess of 1500 °C on the sample. The rate of temperature increase may be varied with an electronic programmer in conjunction with a null detecting controller. This capability allows differential thermal analysis of the samples under pressure.

It is convenient to describe each set of tooling—the Belt and the Piston Cylinder—separately because their pressure-temperature-sample size characteristics are somewhat different.

Piston Cylinder

Figure 12.51 is a schematic of this set of tooling. The upper picture shows the arrangement of the tooling in the die set and press, while the lower schematic shows a magnified view of the tooling. The major components

are also pointed out. The function of the clamping plates is to maintain proper alignment of the components and good electrical contact between the bottom of the specimen cell and the backing block which contains the current lead. The die consists of an inner tungsten carbide cylinder with various steel supporting rings shrunk and press fit around it. The assembly is equipped with a cooling chamber that is necessary at high operating temperatures as the strength of the die materials is considerably reduced above a few hundred degrees centigrade.

The piston is made of tungsten carbide and is beveled at its leading edge to accommodate a triangular cross-section steel ring which prohibits extrusion of the sample. As the load

FIGURE 12.51. Schematic of Piston-Cylinder Tooling.

is applied, the ring is forced outward to the wall and extrudes into the gap between the piston and wall. Such an arrangement is sufficient when compressing solid media, but is obviously not useful in compressing liquid samples in the chamber.

Figure 12.52 is a schematic of a typical specimen cell equipped with a collapsible can arrangement. Figure 12.53 is a photograph of such a cell both in the assembled and disassembled condition. The major components of the cell are pointed out in Figure 12.52. The inside dimensions of the can are 1.42 cm diam by 2.85 cm long before compression, and 2.34 cm long (same diam) after compression. It should be pointed out that the volume occupied by the can (7.1 cm^3) could be used for other purposes such as production of

FIGURE 12.52. Schematic of Sample Cell Assembly for Piston-Cylinder Unit.

high pressure polymorphs or, in other cases, where the applied pressure need not be truly hydrostatic. The can may be filled with liquids to insure that the pressure to the sample is truly hydrostatic. At low temperatures n-pentane is used while at high temperatures, liquid metals such as sodium and NaK are necessary.

The electrical heating circuit is as follows:

- The current flows from the transformer through the current lead to the bottom backing block
- From the backing block, the current travels through the bottom steel closure to the graphite pad and through the thin graphite furnace
- The rest of the circuit comes out the top pad through the piston to ground of the press.

This design offers a maximum of temperature capability with a minimum application of power. Temperatures

FIGURE 12.53. Exploded View of Piston-Cylinder Sample Cell Assembly.

of 1500 °C can be obtained with 9 kW of energy input.

Belt System

This system gets its peculiar name from the design of the die which acts like a circular belt to enclose and contain the sample as the pressure is applied. Figure 12.54 is a schematic of the unit in the press and also contains an enlarged cross-sectional view of the tooling. In contrast to the Piston Cylinder arrangement there are two conical pistons acting on the sample. These pistons are thrust into each end of a conically shaped tungsten carbide die by the 500 ton press. The pressure is generated by the advancing pistons as they compress the specimen cell and its surrounding gaskets. Unlike the Piston-Cylinder tooling where an effort is made to prohibit extrusion of the pressure transmitting media, the lavite gasket material is allowed to extrude to its equilibrium thickness in the Belt tooling. This extruded gasket not only retains the sample, but also provides some measure of support for the pistons.

The pistons themselves are usually made from tungsten carbide with various amounts of cobalt binder. It has been

FIGURE 12.54. Schematic of Belt Tooling.

necessary to use 6% Co binder for the higher pressures (approximately 60 kbars); whereas, 12% is sufficient below 50 kbars. The latter is somewhat more resilient at lower pressures resulting in a much longer working life. The pistons themselves are

supported by shrunk fit steel binding rings so that the stresses in the pistons are always compressive except at the very highest pressures. This is necessary because the tensile strength of WC is much lower than its compressive strength. The face diameter of the piston is 1.27 cm.

The die arrangement is similar to the Piston-Cylinder unit in that the WC die is supported by shrunk fit steel binding rings and provisions are made for water cooling. The difference between the Belt and Piston-Cylinder is mainly the double conical shape of the die necessary to allow piston advance in the Belt system.

Figure 12.55 is a schematic of a typical specimen cell for the Belt tooling. Figure 12.56 shows an actual model and a blown-up view. The components are pointed out in Figure 12.55 and typical dimensions inside the can are 0.56 cm diam by 1.45 cm long before compression, and 1.2 cm long (same diameter) after compression. As in the case of the Piston Cylinder unit, the specimen can volume (0.056 cm^3) can be used for other purposes when true hydrostaticity is not required.

The electrical heating circuit is from the transformer through the current lead and bottom backing block to the bottom piston, see Figure 12.54. From the bottom piston the current travels through the steel end pad, graphite pad, and through the furnace. The rest of the circuit is out through

FIGURE 12.55. Schematic of Sample Cell Assembly for Belt Tooling.

FIGURE 12.56. Exploded View of Belt Sample Cell Assembly.

the top piston to the ground of the press. Typical temperatures of 1500 °C can be obtained by an input of 3 kW of power.

ATR GAS LOOP SUPPORTING STUDIES

MODEL GAS LOOP

P. M. Jackson

The model gas loop is a 2100 °F (1093 °C) recirculating helium loop which is operated to determine the effect of the helium atmosphere on materials and specimen holders, and to see if prolonged use will affect the operating characteristics of the new heater. This loop is a one-tenth size model of the gas loop that is being constructed for the Advanced Test Reactor at Idaho Falls.

The gas loop was operated a total of 3110 hr since installation of the new heater; 2130 hr were accumulated this fiscal year, and 920 hr this quarter. Corrosion test assemblies installed in both the vertical and horizontal test sections have undergone 770 and 1305 hr of exposure respectively. Results of periodic examinations of specimens exposed in both locations are reported elsewhere.

A variable frequency generator of foreign manufacture for the gas-bearing blower in the loop has a history of maintenance outages. Since the last modification, however, it has operated 920 hr without trouble.

Studies to Upgrade the Model Gas Loop

Specimens that are irradiated in the ATR Gas Loop will reach temperatures of 2500 °F or more. A study of methods of upgrading the model gas loop in order to provide out-of-reactor specimen evaluation at this higher

temperature was completed. A supplemental resistance heater similar to the primary heater will be placed in the vertical test section. This unit will consist of several molybdenum tube series connected electrically to an external power source. The corrosion specimens will be placed inside the molybdenum tubes and subjected to only a portion of the total helium gas flow. The incoming helium gas at about 1700 °F will split in about a 20 to 80% ratio; the greater percentage bypassing the corrosion specimens. The 20% portion of the gas will pass through the heater section around the specimens and exit at about 2500 °F. Attemperation of the hot fraction with the 80% bypassed gas, will yield a bulk outlet temperature of about 1900 °F. This gas is then further cooled by operation of the attemperator further downstream to a temperature compatible with the recuperator piping (about 1800 °F). A scope drawing of this heater-specimen holder is shown in the following Figure 12.57.

TEST ASSEMBLY EVALUATION

L. D. Blackburn

During the past several quarters, a refractory metal test assembly has been undergoing high temperature exposure in the model gas loop. The purpose of this experiment is to evaluate the basic design of the assembly itself, and also to study the effects of long term exposure in the high temperature helium atmosphere on the properties of refractory metals. Figure 12.58 illustrates the test assembly. Tensile specimens are mounted in slots in a series of concentric rings. The rings are attached to a central sleeve forming an integral

FIGURE 12.57. Supplemental Heater for Test Section

FIGURE 12.57. Supplemental Heater for Test Section

FIGURE 12.58. Prototype ATR Test Assembly

cylindrical unit, or tier. Tiers slide onto a central rod to form the completed test assembly. Each tier contains eighteen tensile specimens; the unit shown in Figure 12.48 consists of three tiers, while final ATR Gas Loop assembly will contain twelve tiers.

Table 12.19 summarizes the materials in the assembly and the exposure history in the model gas loop. Under the given exposure conditions, the test assembly performed satisfactorily. There was no significant warpage or distortion of the assembly from the

elevated temperature exposure and thermal cycling, nor was there any indication of diffusion bonding within the assembly; specimens were easily removed from the tiers, and the tiers were readily removed from the central rod. The only problem was binding of the retaining nut on the central rod, which resulted from thread galling during initial assembly operations.

Visual examination at selected intervals during the exposure indicated that specimens reacted with contaminants in the helium environment.

TABLE 12.19. Summary of Test Assembly Exposure in Model Gas Loop

Materials:	Nb, Ta, Mo, W Specimens SCb 291 Alloy (Nb-10Ta-10W) Holder
Temperature (Nominal)	940 °C
Axial ΔT	10 °C
Radial ΔT	35 °C
Time at temperature	775 hr
Number of thermal cycles (25 °C-940 °C-25 °C)	18
Average heating time	5 hr
Average cooling time	5 hr
Normal Helium Purity:	H ₂ O, N ₂ , H ₂ , O ₂ , CH ₄ each 1 ppm

Early in the exposure, the surface of molybdenum samples developed a gray, cloudy appearance. Later, tantalum samples developed a purple and then a black surface discoloration. Finally, niobium samples also developed a black discoloration, and tungsten samples developed a gray, cloudy appearance.

Electron microprobe analysis of the Nb-10 Ta-10 W holder material (which also exhibited black discoloration) revealed that significant quantities of molybdenum, chromium, manganese, iron, and oxygen were present on the surface. It seems most probable that the molybdenum and at least some of the oxygen contamination resulted from oxidation of the molybdenum heating elements in the model loop heater. With oxygen contamination in the loop, the heating elements may well oxidize to form MoO_3 , which then volatilizes and is transported in the gas stream to the test assembly. The chromium, manganese, and iron contamination probably results from evaporation of

the Haynes 25 liner in the high temperature (1150 °C outlet section of the heater, and these vapors are also carried in the gas stream to the assembly. It may be noted that there was no indication of contamination by nickel or cobalt, the other major alloying elements in Haynes 25.

Results of room temperature tensile tests on specimens from the test assembly, along with those for as-received specimens, are summarized in Table 12.20. The specimens of niobium, tantalum, and molybdenum were recrystallized in the as-received condition. Niobium samples from the model loop exposure show significant increases in strength, while tantalum exhibits a smaller increase. Although neither material shows large changes in room temperature ductility, it seems quite likely that the ductile-brittle transition temperature increased, but still lies well below room temperature. In contrast to the property changes in niobium and tantalum, molybdenum seems

TABLE 12.20. Room Temperature Tensile Properties of Refractory Metals After Model Gas Loop Exposure

Specimen	Yield Strength, psi	Tensile Strength, psi	Elongation % in 1 In.	Reduction of Area, %
<u>As received</u>				
Nb G24	16,400	30,500	53	98
Nb G26	14,100	29,900	46	98
Ta G15	41,600	50,600	39	97
Mo G32	75,100	75,800	6.3	1.9
<u>Exposed in Model Gas Loop at 940 °C for 775 Hr</u>				
Nb G18	37,000	47,000	35	95
Nb G22	34,100	46,000	35	95
Ta G4	45,500	58,000	44	90
Mo G34	73,300	80,000	4.9	4.8

to be unaffected by model loop exposure. At this time, it is believed that interstitial contamination by oxygen and/or nitrogen plays a major role in the strengthening of niobium and tantalum, although surface contamination by metal species may also contribute to the observed effects. Additional experiments and metallographic analysis are in progress to verify this hypothesis.

SUPERALLOY AND REFRACTORY METAL CORROSION STUDIES

L. A. Charlot

The weight change data from superalloy and refractory metal samples exposed in the ATR Model Loop and reported last quarter⁽³⁾ suggest that metal contamination of niobium and tantalum could have been partially responsible for their weight gain. Refractory metal samples taken from the 2100 °F test section and examined for possible contamination were first scanned through their cross section with an electron beam microprobe, and, in the case of the tantalum sample, the reaction surface itself was scanned directly. The analytical results for samples exposed 504 hr in the Model Loop are discussed in the following section.

Molybdenum

Nothing unusual in microstructure was observed. Some iron and chromium presumably from the evaporating superalloy samples, were present as thin film on sample surface. Cobalt and nickel were not detected. Hardness was not determined.

Tungsten

Nothing unusual in microstructure was observed. A trace of iron, and a possible trace of chromium were found on the sample surface. Cobalt and nickel were not detected. Hardness was not determined.

Tantalum

Some small inclusions were noted in the microstructure. A possible trace of chromium was found on surface. Black, sooty surface deposit was not identifiable by either microprobe or X-ray diffraction analysis. The black deposit may bear a similarity to the surface case found on the niobium sample.

Niobium

The microstructure of this sample is shown in Figure 12.59. In addition to the surface case, contamination is evidenced by a large number of inclusions in the base metal. Microprobe analysis failed to reveal any metal contamination of the sample surface. X-ray diffraction investigation of the surface case showed it to be a Nb-N-O ternary compound, possibly of stoichiometry near $\text{Nb-N}_{0.9}\text{-O}_{0.1}$.⁽⁷⁹⁾ This finding implicates atmospheric gases as the major contaminant source, at least in the case of niobium.

Hardness traverses were made through the cross sections of the contaminated niobium and tantalum samples, and as-received samples of the same materials, to gain an idea of the extent to which their mechanical properties had been altered during the 504-hr Model Loop

250X

FIGURE 12.59. Microstructure of Niobium Sample Exposed in Model Loop for 504 hr at 2100 °F (Etchant: 30 Lactic Acid, 10 HNO₃, 10 HF)

exposure. Results of the traverses are shown in Table 12.21.

Note that there is a great deal of contamination-induced hardening in the exposed samples, indicating appreciable contaminant absorption. Diffusion calculations show that both nitrogen and oxygen will diffuse rapidly enough through the sample coupons to effect almost complete homogenization within the time duration of any one run. Because the samples were cooled slowly from the operating temperature, it is not possible to determine contaminant

levels or meaningful contaminant distributions by comparison with the microhardness data of other workers, nor is it possible at present to conclude which contaminant, oxygen or nitrogen, is the principal hardening agent.

Duplicate samples were added to the test holder in place of those analyzed. These samples have been through two cycles for a total of 763 hr at temperature. Table 12.22 summarizes the weight change of all samples to date.

TABLE 12.21. Microhardness Traverses, VHN, 1 kg Load

<u>Nb Sample</u>		<u>Ta Sample</u>	
<u>As-Received</u>	<u>Contaminated</u>	<u>As-Received</u>	<u>Contaminated</u>
69	250	83	379
71	242	90	370
77	270	96	379
73	250	94	360
85	256	91	355
76	242		374
	245		365
			351

TABLE 12.22. Corrosion Samples Exposed to Model Loop Conditions

Material	Sample Number	Average Temperature	Weight Change, mg/cm ²	Time at Temp., hr	Number of Cycles ^(a)
Hastelloy X	2	2100 °F	-4.85	1267	6
Haynes 25	2	High Gas	-4.56	1267	6
Haynes 25	7	Velocity	-3.68	1055	3
Hastelloy X	6	Section	-3.25	763	2
Haynes 25	6		-3.17	763	2
Molybdenum	4		-0.04	763	2
Niobium	3		1.83	763	2
Tantalum	4		1.12	763	2
Tungsten	4		-0.02	763	2
Niobium, 1 Zr	1		2.39	535	1
Hastelloy X	3	2025 °F	-1.96	1267	6
Hastelloy X	4	Low Gas	-1.81	1267	6
Haynes 25	3	Velocity	-2.26	1267	6
Haynes 25	4	Section	-2.10	1267	6
Molybdenum	3		-0.04	1267	6
Niobium	2		2.12	1267	6
Tantalum	2		1.26	1267	6
Tungsten	3		-0.05	1267	6
Hastelloy X	5		-1.80	1055	3
Haynes 25	5		-2.10	1055	3
Niobium	5		1.95	1055	3
Tantalum	3		1.15	1055	3
Niobium, 1 Zr	2		1.82	535	1
TZM	1		0.01	535	1

(a) A cycle refers to loop "start up" which consists of loop opening, closing, evacuation, and refilling with high purity helium (<1 vpm impurity) to 300 psi.

Future work will entail continued exposure of the corrosion samples to loop conditions. Samples will be added to or taken from the test primarily in an effort to determine the extent of contamination in niobium and tantalum.

ATR GAS LOOP OPERATION AND MAINTENANCE

G. A. Last

MODEL GAS LOOP STUDIES

The Model Gas Loop has operated a total of 3108 hr since installation of the new heater. Although the gas impurity level is maintained at less than 1 ppm by the by-pass cleanup system, there is evidence, from corro-

sion specimens in the loop, of in-leakage of air. Tests are planned to determine the source and amount of this in-leakage.

The refractory metal test assembly has been dismantled following a total exposure of 775 hr at 1700 °F (925 °C) in the Model Gas Loop. The Nb-10 Ta-10 W holder and the Nb and Ta specimens showed considerable black discoloration, while the Mo and W specimens exhibited gray, cloudy discoloration. Electron microprobe analysis revealed significant amounts of Mo, Cr, Mn, Fe, and O contamination on the surfaces of the holder. It seems probable that MoO₃, Cr, Mn, and Fe vapors are formed in the heater by

oxidation of the Mo heating elements and evaporation from the Haynes 25 liner, and these vapor species are then carried through the gas stream to the test assembly.

Room temperature tensile tests demonstrated significant strengthening of Nb, only modest strengthening of Ta, and no strength change of Mo as a result of the model loop exposure. Since no large changes in ductility accompanied the strengthening of Nb and Ta, the ductile-brittle transition apparently still lies well below room temperature.

Specimens irradiated in the ATR Gas Loop will reach temperatures up to 2500 °F (1370 °C). A study is being made of methods to raise the specimen temperature within the vertical test section of the Model Gas Loop in order to more closely simulate these temperatures. Three methods have been considered: (1) heating the gas with a resistance type heater, (2) induction heating the specimens, and (3) resistively heating tubular sections which would contain the test specimens. The third method has been selected as the one having the greatest potential of success and requiring the least amount of development effort. This arrangement would utilize a number of molybdenum tubes that are electrically connected in series. The test specimens would be placed inside these tubular heating elements and completely insulated (electrically) from them. The cluster of tubes would be arranged to flow a portion of the helium over the specimens and by-pass the remainder of the gas through the annular passage between the present test section and

the auxiliary heater. The specimen temperatures will be controlled by varying the gas flow or the power to the auxiliary heater. A computer program will be used to determine the specimen temperatures that can be reached by this method under varying flows and power settings.

ANALYTICAL INSTRUMENTATION FOR THE ATR GAS-COOLED LOOP

The transfer facility for the gas loop is designed to operate with dry air at a dew point of -150 °F (-100 °C). The instrumentation for measuring the dew point was inspected by PNL and Ebasco personnel to determine acceptability of the units.

Performance of the hygrometers was monitored by supplying them with a gas of known dew point. The gases were generated by use of a tempering coil and cold bath. Liquid nitrogen was used to prepare a gas that contained less than 2.5×10^{-19} mm water vapor. Although this value is considerably lower than the hygrometers will measure, it did provide the zero point gas to determine whether the zero suppression on the hygrometers was adequate to give a base line within the accuracy of our specifications.

After the system had been dried out, the recorder indicated a reading of 0.06 ppm which corresponds to a frost point of -136 °F (-93 °C) at the local atmospheric pressure. This put the minimum reading within the tolerance of the specifications.

Other monitors in the system are used at the higher range of water vapor and the accuracy in these ranges was checked and found to be adequate.

Based on our requirements and specifications, the system was accepted for shipment.

Continual use of the Greenbrier chromatographs on the Model Gas Loop has shown minor weaknesses in the equipment associated with an instability of the base line. This instability appeared to be mainly from the loss of voltage regulation in the electrometer. The manufacturer has supplied two new electrometers, one for the chromatograph and one for the total impurity monitor. These units have been installed and are in the process of checkout.

Recent studies in the Model Loop have proven that extreme sensitivity in the chromatographs will be required to monitor impurity levels in the ATR Gas Loop. Work will continue on the units to ensure that total impurities can be measured at levels below 1 ppm.

INVESTIGATION OF TYPE 304H STAINLESS STEEL LONGITUDINAL PIPE WELDS

In compliance with a request by the Idaho Operations Office, gas tungsten-arc welds made by Swepco Tube Corporation were evaluated by W. F. Brown, Battelle-Northwest. These welds were from a section of the pressure containment pipe for the ATR Gas Loop. The Solar Aircraft Company, who is fabricating the piping assembly, purchased the 12 in., Sche 40, type 304H SS piping from Swepco Tube Corporation.

Swepco's manufacturing process consists of forming plate stock into a tubular configuration and welding the longitudinal square groove butt joint using their gas tungsten-arc welding process called "Syncroweld." The

Syncroweld process utilizes two torches positioned so that welding is performed from the underside of the joint as well as the upper side simultaneously without filler wire addition. This welding procedure yields a joint configuration comparable to a double-welded butt joint.

Evaluation of the 7 1/2 in. long weld was based on the results of visual examination, X-ray examination, fluorescent penetrant inspection, and metallographic examination. The criterion for evaluation was Section III, Nuclear Vessels, ASME Boiler and Pressure-Vessel Code.

Fluorescent penetrant inspection revealed several surface cracks. Metallographic examination revealed that over 50% of the joint length had incomplete fusion penetration. Although diffusion bonding had occurred in the unfused areas from high temperature and pressure, the bond was of relatively poor quality compared with the fused area. Although X-ray could not detect this condition, the metallographic examination indicated that the process was out of control.

Based on this investigation, it was concluded that the welds submitted for evaluation were not acceptable and did not meet the intent of Section III of the ASME Boiler and Pressure Vessel Code.

SUPERALLOY AND REFRACTORY METAL CORROSION STUDIES

Corrosion coupons exposed in the ATR Model Loop (504 hr total exposure at temperature) continue to follow the weight change trend previously reported. Metallography and microprobe analysis of the samples taken

from the high gas velocity section (2100 °F) (1150 °C) is presently being completed. Cursory examination of the microstructure of Hastelloy X-280 and Haynes 25 which had weight losses of 2.77 and 2.67 mg/cm² respectively, show metal loss from grain boundaries and what appears to be subsurface voids. Niobium, 2.24 mg/cm² total weight gain, shows a continuous surface case approximately 8 μ thick. The base metal also contains what appear to be intermetallic inclusions. After removal from the Model Loop, the tantalum sample, 1.77 mg/cm² weight gain, exhibited a loose, easily wipeable black deposit on the surface. However, metallography failed to show a well defined subsurface layer. The contaminant has yet to be determined for either niobium or tantalum.

Duplicate coupons were added to the test holder in place of those analyzed and the assembly will again be exposed to loop conditions.

Electron microprobe analysis of the tantalum, niobium, molybdenum and tungsten samples taken from the 2100 °F (1150 °C) heater discharge section of the Model Loop after a 504 hr exposure failed to show foreign metal contamination of their respective surfaces. Constituents of the black surface noted on tantalum and the surface case on niobium were not identifiable by microprobe. X-ray diffraction of these two samples did reveal the nature of the contamination on the niobium sample. Strong diffraction lines suggest Nb-N-O, a ternary compound of niobium, nitrogen, and oxygen of stoichiometry Nb-N_{0.9}-O_{0.1}. The contamination on tantalum, although not identified by X-ray diffraction, may be similar to the niobium contamination.

Corrosion coupons in the lower temperature (2025 °F) (1105 °C) discharge section of the loop have about 732 hr total exposure time at temperature. The coupons replacing those taken from the higher temperature section now have 228 hr exposure. Within experimental tolerances, these coupons follow the weight change behavior established by the first set. The samples are continuing their exposure in the Model Loop.

THERMAL-HYDRAULIC ANALYSIS

A steady-state heat transfer program was set up to calculate temperatures of a niobium hanger rod and test section liner under an assumed "worst case" condition in the ATR helium loop. Temperatures were calculated for a representative horizontal cross section through the test section. Thermal radiation effects were accounted for. Helium temperature was taken as 2000 °F (1095 °C), and a heating rate of 25 W/g was assumed.

At the above conditions, the hanger rod temperature will be 2530 °F (1390 °C), the liner temperature will be 3450 °F (1900 °C), and the tube wall will be at approximately 950 °F (510 °C). Since the liner will not bear any structural load other than its own weight, the temperature, while it is high, should not lead to any gross structural failure. The strength of the niobium hanger rod at 2530 °F (1390 °C) will be investigated, and some means of lowering the rod temperature may have to be devised.

It is planned to run more computer cases to calculate hanger rod and liner temperatures with varying helium temperatures, materials, and heating rates.

Experimental measurements of the pressure drop across the test assembly in the Model Gas Loop at several temperatures and gas flow rates have been obtained. Pressure drops at temperatures above 500 °F (260 °C) were higher than those obtained in similar experiments conducted two years ago. In both sets of experiments, however, the measured pressure drop multiplied by the helium gas density is a linear function of the square of the mass flow rate. This is the expected relationship for frictional losses in a fluid flow system. Extrapolation of the higher pressure drop data from the recent experiments to ATR operating conditions still leads to the conclusion that the maximum allowable pressure drop for the ATR specimen test assembly will not be exceeded with the current design.

VENDOR DATA REVIEW

About 60% of the vendor data on the ATR Gas Loop has been reviewed, and comments have been submitted to the architect-engineer.

Contract submittals on the most recent ATR Gas Loop orders have been reviewed. Some of the submittals from the various vendors, such as the heat exchangers, require a complete review of the analysis to determine code and material compliance. The superalloys used in the regenerative exchanger are being reviewed thoroughly based on our past experience with these materials in the Model Gas Loop.

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