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**QUARTERLY PROGRESS REPORT
IRRADIATION EFFECTS
ON REACTOR STRUCTURAL MATERIALS
MAY, JUNE, JULY, 1968**

SEPTEMBER 1968

**AEC RESEARCH &
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ON REACTOR STRUCTURAL MATERIALS
MAY, JUNE, JULY, 1968

Participating Organizations:

Argonne National Laboratory
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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
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September 1968

Edited by:

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TABLE OF CONTENTS

SUMMARY	2.1
ARGONNE NATIONAL LABORATORY	3.1
The Effects of Irradiation on the Tensile Properties of EBR-II Type 304 Stainless Steel - R. Carlander	3.1
Examination of Irradiated Type 304 SS and V-20 wt% Ti by Transmission Electron Microscopy - S. D. Harkness	3.4
A Model of Void Formation - S. D. Harkness	3.10
BATTELLE MEMORIAL INSTITUTE	4.1
The Effect of Radiation on the Mechanical Properties of Cladding Materials - M. Kangilaski, S. L. Peterson, J. S. Perrin, and R. A. Wullaert	4.1
Correlation of Radiation Effects on Mechanical Properties as Caused by a Predominantly Fast Flux and a Mixed Fast and Thermal Flux - M. Kangilaski, J. S. Perrin, and R. A. Wullaert	4.6
Radiation Effects on the Mechanical Properties of Type 347 SS Fermi Wrapper Tube - J. A. DeMastry, J. W. Sheckherd, and R. A. Wullaert	4.8
Determination of the Fracture Toughness of Specimens from the PM-2A Pressure Vessel - J. A. DeMastry, J. W. Sheckherd, and R. A. Wullaert	4.10
BROOKHAVEN NATIONAL LABORATORY	5.1
Effect of Microstructure on Tensile Properties of Irradiated Stainless Steel - J. G. Y. Chow and R. Jones	5.1
Effect of Irradiation on Tensile Properties of Irradiated Superplastic Fe-Ni-Cr Alloys - J. G. Y. Chow	5.2
Radiation Hardening of Iron - P. Soo and R. Jones.	5.4
IDAHO NUCLEAR CORPORATION	7.1
Irradiation Effects on Reactor Structural Materials - J. M. Beeston and C. R. Brinkman	7.1
NAVAL RESEARCH LABORATORY	9.1
Research Progress - L. E. Steele, J. R. Hawthorne, C. Z. Serpan, Jr., Uldis Potapovs, and R. A. Gray, Jr.	9.1
Irradiation Response of a 4 in. A533-C, Class 2, Submerged Arc Weldment	9.1
Relative 550 °F Irradiation Response of a Heavy Section A533-B Electroslag Weldment	9.3
Special A533-B Steel Heat for Variable Radiation Embrittlement Studies	9.7
Thermal Stability Evaluation of Ni-Cr-Mo Weld Deposits.	9.8
Thermal Stability Characteristics of 12 Ni-5 Cr-3 Mo Maraging Steel Plate Evaluated at 550, 650, and 740 °F Using Charpy-V Notch Specimens.	9.8
Long-Term Irradiation of Pressure Vessel Steels in the Big Rock Point Reactor	9.10
Plate Directionality Characteristics of Irradiated La Crosse Reactor Pressure Vessel Steel	9.14
Preliminary Study of the Irradiation Response of A537 Grade B Steel at Less Than 250 °F	9.18
NUCLEAR MATERIALS AND PROPULSION OPERATION	10.1
Radiation Effects on Fast Reactor Cladding and Structural Materials - J. Moteff	10.1

Status of Irradiations - F. D. Kingsbury, J. P. Smith, and W. S. Chenault.	10.1
ORR Facility	10.1
EBR-II Facility.	10.2
Heat-Resistant Alloy Program	10.2
Creep-Rupture Testing - J. P. Smith and and R. A. Joseph	10.2
Low Cycle Plastic Fatigue - J. P. Smith and J. T. Berling	10.3
Refractory Metal and Alloys Program	10.5
Creep-Rupture - F. Kingsbury and R. Treinen.	10.5
Resistivity Studies - L. K. Keys and J. Moteff	10.7
Transmission Electron Microscopy of Vanadium - R. C. Rau, F. Secco d'Aragona, and R. L. Ladd	10.8
OAK RIDGE NATIONAL LABORATORY	11.1
Solid State Division	11.1
Radiation Metallurgy - D. S. Billington and M. S. Wechsler.	11.1
Introduction - M. S. Wechsler	11.1
Inhomogeneous Distribution of Ta and W in Zone-Refined Niobium - R. E. Reed	11.2
The Effect of Interstitial Impurities on Post- Irradiation Annealing Phenomena in Niobium - J. M. Williams, J. T. Stanley, and W. E. Brundage	11.6
Pseudo Snoek Peak in Niobium - J. T. Stanley	11.14
Radiation Anneal Hardening in Niobium - S. M. Ohr and R. P. Tucker	11.21
Dislocation Channeling in Neutron Irradiated Niobium - R. P. Tucker and M. S. Wechsler.	11.25
The Temperature Dependence of Radiation Hardening - S. M. Ohr	11.54
Metals and Ceramics Division	11.57
Irradiation Damage to Type 316 and Titanium-Modified Type 316 SS - E. E. Bloom and J. R. Weir	11.57
Mechanical Properties of Titanium-Modified Type 304 SS - E. E. Bloom and J. R. Weir, Jr.,	11.60
Electron Microscope Evaluation of Titanium- Modified Type 304L SS - G. T. Newman	11.63
Drawing of Stainless Steel Tubing - G. A. Reimann	11.65
The Effect of Cyclotron-Injected Helium on the Creep-Rupture Properties of Type 304 and 0.2% Titanium-Type 304L SS - R. T. King and J. R. Weir, Jr.	11.66
Electron Microscopy of Irradiated EBR-II Fuel Cladding - J. O. Stiegler, E. E. Bloom, and J. R. Weir, Jr.	11.73
Some Effects of Grain Size on the Radiation Damage of Incoloy 800 - D. G. Harman	11.76
Voids in EBR-II Irradiated Vanadium - F. W. Wiffen and J. O. Stiegler	11.79
Comparison of Laboratory and Commercial Heats of Titanium - Modified Hastelloy N - C. E. Sessions and H. E. McCoy.	11.82

PACIFIC NORTHWEST LABORATORY.	12.1
Irradiation Damage to Reactor Metals - A. L. Bement	12.1
Reactor Metals Research	12.1
Irradiation Facilities Operation - R. W. Barker.	12.1
Irradiation Damage to Stainless Steel - A. J. Lovell	12.2
Transmission Electron Microscopy of Irradiated Austenitic Stainless Steel - H. R. Brager	12.4
Irradiation Damage to Nickel Base-Alloys - I. S. Levy	12.18
In-Reactor Measurements of Mechanical Properties - E. R. Gilbert and N. E. Harding.	12.19
Grain Boundary Stabilization - E. R. Bradley	12.20
Fracture Studies - R. G. Hoagland and J. A. Williams	12.23
High Pressure Studies - G. L. Kulcinski.	12.28
In-Reactor Corrosion of Zirconium Alloys - A. B. Johnson, Jr.	12.30
Fast Reactor Supporting Studies	12.33
Fast Neutron Mechanisms - R. P. Allen	12.33
Test Facilities - R. H. Todd	12.37
LMFBR Fuel Cladding Information Center - F. B. Shober	12.38
Radiation Effects on Metals-05 Program - T. K. Bierlein, E. R. Bradley, J. L. Brimhall, H. E. Kissinger, G. L. Kulcinski, T. J. Larson, B. Mastel, and F. A. Schmidt, Jr..	12.41
Electron Microscopy.	12.41
Some Observations on the Annealing of Neutron Irradiated Single-Crystal and Polycrystalline Molybdenum	12.45
ATR Gas Loop Operation and Maintenance - J. E. Minor	12.52
ATR Gas Loop Support	12.52
ATR Water Loop Support	12.52
WESTINGHOUSE ELECTRIC CORPORATION	13.1
Fracture Mechanics Evaluation of Reactor Vessel Steels - T. R. Mager, F. O. Thomas, and W. S. Hazelton	13.1

SUMMARYARGONNE NATIONAL LABORATORY

The yield and tensile strengths of AISI Type 304 SS at test temperatures between 450 and 750 °C increased and the ductility decreased as a result of irradiation in EBR-II to a nominal fluence of 5×10^{22} at 371 to 480 °C.

The irradiated strength was greater at a test temperature of 450 °C than at test temperatures of 550, 650, and 750 °C (whereas, the irradiated ductility was low

at all test temperatures) for material irradiated to approximately the same fluence and irradiation temperature.

Material irradiated to approximately the same fluence and tested at the same test temperature had higher strength and lower ductility when irradiated at approximately 371 °C than when irradiated at approximately 480 °C.

BATTELLE MEMORIAL INSTITUTE

Specimens of UMo-50, Haynes Stellite 25, Tenelon and Nickel 200 were tested in the 450 to 650 °C range after irradiation. These specimens contained varying nickel contents and were irradiated to establish if nickel was necessary for the elevated temperature embrittlement. These specimens had been irradiated to a fast fluence of 1.5 to 1.6×10^{21} n/cm² in the ETR at 50 °C. Results of the tensile tests showed that all of the specimens were embrittled by irradiation. Consequently, it was concluded that the presence of nickel was not necessary to cause elevated temperature embrittlement in irradiated materials. Stress rupture tests on irradiated (2.2×10^{22} n/cm²) Type 347 SS were continued at 650 °C.

Tensile tests were performed on

specimens from the Fermi wrapper tube which had been irradiated to a fast fluence of 3×10^{20} n/cm² (E > 1 MeV). These tensile tests indicated that the mechanical properties were not significantly changed at any of the testing temperatures.

Fracture toughness tests have been performed on both 1X WOL and IT WOL type specimens which were cut from the irradiated PM-2A pressure vessel. It was found that the fracture toughness at -20 °F was strongly dependent on fast fluence. Specimens receiving a fast fluence of 4.2×10^{10} n/cm² exhibited an average K_{IC} value of 105 ksi√in. while specimens receiving a fast fluence of 7.7×10^{18} n/cm² had a fracture toughness of 69 ksi√in. It appears that the specimen size does not affect the K_{IC} values.

BROOKHAVEN NATIONAL LABORATORY

Altering the structure of Type 304 SS by cold swaging 92% and annealing at 760 °C greatly improves the postirradiation ductility between 650 and 800 °C.

Various compositions of 2 phase, Fe-Ni-Cr alloys that exhibit superplastic properties at temperatures above 750 °C are being irradiated. Preliminary results indicate that irradiation to a fluence of 1.3×10^{20} nvt destroyed the superplastic properties of these alloys.

A fast neutron dose of 10^{18} nvt ($E > 1.45$ MeV) at 80 °C causes a change in the rate-controlling slip mechanism in iron at temperatures below about 250 °K. The new mechan-

ism involves dislocation interactions with small vacancy loops which are created during irradiation. The slip process between 250 to 350 °K is, however, basically the same as that operating in unirradiated material, although the data indicate that irradiation causes some strengthening of the controlling obstacles to slip. Above about 400 °C, dynamic strain aging is important and the free carbon interstitial is most likely responsible. The large increase in the athermal component of the flow stress is believed to be caused by the presence of depleted zones which are formed by incident fast neutrons.

IDAHO NUCLEAR CORPORATION

Delayed failure tests on A542 steel in the normal quenched and tempered condition at initial hydrogen concentrations of approximately 3.2 and 5 ppm indicate that hydrogen reduces the ductility, but has only a slight tendency towards inducing cracking. When heat treated to R_c 25-26, the A542 steel was susceptible to hydrogen induced cracking and failure such that with 6 ppm a lower critical stress of 50% of the ultimate strength was obtained,

while after heat treatment to R_c 34-37 the lower critical stress was 35% of the uncharged ultimate strength. The ductility of A302B steel in tensile tests as represented by percent reduction in area at fracture shows a significant decrease (from 65 to 30%) with increasing hydrogen content to 5 ppm.

The first two subassemblies containing fatigue specimens had received in EBR-II Run 28, 1126 MWd.

NAVAL RESEARCH LABORATORYIRRADIATION RESPONSE OF A 4 IN.
A533-C CLASS 2 SUBMERGED ARC
WELDMENT

The Charpy-V notch ductility properties of a 4 in. A533-C, Class 2 plate and submerged arc weld deposit were compared before and after irradiation at 550 °F to a neutron fluence of approximately 1.2×10^{19} n/cm² (E > 1 MeV). Both the plate and weld metal exhibited high radiation embrittlement sensitivity (180 °F and 205 °F transition temperature shifts, respectively), although the losses in full shear energy absorption were relatively minor. Recent studies of variable radiation embrittlement sensitivity of A302-B steel have demonstrated that copper contents in amounts found in the 4 in. A533-C plate and weld deposit (~0.2%) can contribute significantly to radiation embrittlement sensitivity, which suggests one explanation for the large transition temperature increases observed.

RELATIVE 550 °F IRRADIATION RESPONSE
OF A HEAVY SECTION A533-B ELECTROSLAG
WELDMENT

The irradiation response of a commercially produced 5 3/4 in. thick A533-B, Class 1 electroslag weldment has been investigated. For the preirradiation condition, the base plate exhibited a lower

Charpy-V 30 ft-lb transition temperature than the weld metal (-60 °F versus 15 °F). Postirradiation test data for a fluence of 1.6×10^{19} n/cm² (E > 1 MeV) at 550 °F revealed a threefold difference between Charpy-V transition temperature responses of weld metal and base metal. The radiation embrittlement, however, is consistent with the high level of sensitivity observed for submerged arc weld metals for this type steel.

SPECIAL A533-B STEEL HEAT FOR VARI-
ABLE RADIATION EMBRITTLEMENT STUDIES

At the request of the U.S. Atomic Energy Commission (AEC), a special tonnage heat of A533-B steel is being procured with compositional control for limiting 550 °F radiation embrittlement sensitivity. A 30 ton electric furnace heat was air melted on May 23, 1968, and split between two 62 in. wide by 22 in. thick ingot molds after vacuum stream degassing. During the casting of one ingot, an addition of pure copper shot was made to permit an evaluation of the tolerance of this steel to 0.13% (aim) copper versus only trace quantities of copper. The ingots are to be rolled to 6 in. plate using a 1:1 cross rolling ratio, whereupon each plate will be split with individual sections heat treated to Class 1 and Class 2 properties, respectively.

THERMAL STABILITY EVALUATION OF Ni-Cr-Mo WELD DEPOSITS

Thermal stability evaluations have been completed on six commercial and experimental Ni-Cr-Mo weld deposits which were used in a cooperative NRL-Babcock and Wilcox (B&W) radiation embrittlement sensitivity program. Irradiation results from this program were summarized earlier. All weld deposits showed good thermal stability following long term (~1200 hr) aging at 550 °F.

THERMAL STABILITY CHARACTERISTICS OF 12 Ni-5 Cr-3 Mo MARAGING STEEL PLATE EVALUATED AT 550, 650, AND 740 °F USING CHARPY-V NOTCH SPECIMENS

The response of unirradiated Charpy-V specimens machined from two heats of 12 Ni-5 Cr-3 Mo maraging steel (160 to 185 ksi yield strength) has been determined for thermal exposures of 1900 and 3300 hr at 550, 650 and 740 °F. The exposures were matched to time-at-temperature histories for neutron irradiations of specimens from these same two plates. The results indicate that the aging processes continue at these temperatures and produce increases in the transition temperature which counteract (or conceal) the annealing of radiation damage typical for this temperature range. Thus, higher than expected transition temperatures result from the radiation exposure at elevated temperatures. The need is indicated for development of time-at-temperature versus property change data for the 12 Ni-5 Cr-3 Mo maraging

steel composition if it is to be seriously considered for future elevated temperature reactor applications.

LONG-TERM IRRADIATION OF PRESSURE VESSEL STEELS IN THE BIG ROCK POINT REACTOR

Notch ductility characteristics of A302-B and A543 steels have been evaluated after a 9720 hr irradiation at 575 °F in an accelerated surveillance location of the Big Rock Point Reactor (BRPR). The high neutron fluence for the exposure (~ 6 to 7×10^{19} n/cm², $E > 1$ MeV) resulted in transition temperature increases of 175 and 105 °F, respectively. When compared to data for 550 °F irradiation, the approaching condition of saturation of radiation-induced embrittlement for both of these steels becomes evident.

PLATE DIRECTIONALITY CHARACTERISTICS OF IRRADIATED LA CROSSE REACTOR PRESSURE VESSEL STEEL

Transverse plate orientation notch ductility properties of two heats of La Crosse Boiling Water Reactor (LACBWR) steel were developed to complement the longitudinal orientation properties reported earlier. The transverse specimens showed initial full shear energy shelf levels less than half those of the longitudinal orientation. Upon irradiation to 1.0×10^{19} n/cm² ($E > 1$ MeV) at 550 °F, the transverse specimens indicated a decrease in shelf energy proportional to that exhibited by longitudinal specimens after a 2.1×10^{19} n/cm² ($E > 1$ MeV)

irradiation also at 550 °F. The 30 ft-lb level transition temperature increases were also in proportion. These data suggest that the limiting structural factor in the LACBWR vessel may be the transverse fracture toughness. Nevertheless, these data indicate that the LACBWR vessel should be capable of accepting lifetime neutron fluences of 2.1×10^{19} n/cm² (E > 1 MeV) or more without undue concern for vessel ductility.

PRELIMINARY STUDY OF THE IRRADIATION
RESPONSE OF A537 GRADE B STEEL AT
LESS THAN 250 °F

Longitudinally oriented Charpy-V specimens from a 3/4 in. thick plate

of A537-B steel have been irradiated at less than 250 °F to a fluence of 5.1×10^{18} n/cm² (E > 1 MeV). Test results indicate that this steel is quite similar in radiation response to A302-B steel under these conditions. Transversely oriented specimens revealed a full shear energy absorption shelf just over one third that of the longitudinal orientation. Special attention will be given to longitudinal versus transverse properties of this steel in future studies.

NUCLEAR MATERIALS AND PROPULSION OPERATION

Creep-rupture testing at 705 °C of Incoloy 800 irradiated in a thermal (GETR) or in a fast spectrum (EBR-II) reactor show that the effect of spectrum on high temperature mechanical properties is very minor at comparable fast fluences. Post-test metallographic examination of fractured Incoloy 800 indicates that when transgranular fracture of the unirradiated specimens predominates, the changes in rupture properties are minimal.

Low cycle plastic fatigue testing of Type 316 SS at 650 °C at a strain rate of 4×10^{-5} /sec has shown that fatigue life (N_5) is reduced by a factor of approximately 3.5 following an irradiation at ambient (~70 °C) temperatures to fluences of 3×10^{18}

n/cm² (E > 1 MeV) and 2×10^{19} n/cm² (thermal).

The effect of irradiation temperature on the creep-rupture properties of polycrystalline molybdenum (2 carbon levels, 38 and 220 ppm) at 1000 °C was determined. In general, irradiation at 70 °C resulted in the least effect on the time to rupture; irradiation at 700 and 1000 °C showed increases by factors of approximately 1.4 and 2.5, respectively. Delayed creep was observed in the 700 and 1000 °C irradiated specimens. The results suggest that, for the carbon contents investigated, the major influence of carbon is on the unirradiated elevated temperature plastic

flow characteristics and not on the stabilization of the irradiation-induced defects.

An isochronal resistivity recovery study of molybdenum after irradiation to a fluence of 1.5×10^{21} n/cm² ($E > 1$ MeV) shows only a single large recovery which occurs at about $0.31 T_m$ and believed to be due to vacancy migration (Stage IV). A residual resistivity remains after the high temperature annealing treatment which is attributed to the presence of technetium (and ruthenium) formed from thermal neutron (n, γ) transmutation reactions.

Hot-microhardness data were determined for irradiated Mo-0.5 Ti and unirradiated vanadium and tungsten. The influence of hold times on the hot-hardness properties has been initiated. A study of room-temperature hardness of irradiated (1.2×10^{20} n/cm², $E > 1$ MeV) Ta and Ta-10 W following elevated-temperature anneals has defined the recovery characteristics of these materials. Thermal hardening was observed at 400 °C with recovery beginning at 750 °C. Complete recovery occurred at approximately 1360 °C.

Transmission electron microscopy studies were begun on irradiated and unirradiated vanadium single crystals. As-received unirradiated specimens exhibited domain boundaries and, less extensively, dark precipitates and small planar fringed regions. Annealing at 300 °C eliminated the domain boundaries but enhanced the precipitates and fault-like regions. The precipitates appear to be exceedingly thin and have been tentatively identified as V₂C. Irradiated specimens showed the presence of radiation-induced defect clusters which increased in average size and decreased in number upon annealing in the 330 to 1175 °C temperature range. Above 500 °C, many of the clusters were believed to be interstitial in nature. The clusters appeared to be more persistent, on a homologous temperature scale, than other bcc metals. Circular halos surrounding matrix particles were observed in the irradiated material. They are believed to be damage shells surrounding boron-containing precipitates similar to those observed in Fe- and Ni-base alloys.

OAK RIDGE NATIONAL LABORATORY

SOLID STATE DIVISION

This report covers recent progress in the study of radiation damage and radiation hardening in niobium. An investigation of methods to purify

and prepare niobium materials shows that tungsten and tantalum impurities exist in layers after electron-beam zone refining. This was detected by autoradiographic techniques which are capable of distinguishing between

the tungsten and tantalum impurities and yielding, potentially, a quantitative measure of their distributions.

The influence of interstitial impurities, particularly oxygen and carbon, on the response to radiation has been studied by internal friction and resistivity measurements. Previous work showed that oxygen atoms become trapped at radiation-produced defects at about 150 °C in neutron-irradiated niobium. In this report, the de-trapping process upon annealing above about 300 °C is described. The internal friction results indicated a more rapid return of oxygen to solid solution than did the resistivity results. Extraneous contributions to the internal friction may be involved; the evidence for a "pseudo Snoek" internal friction peak is given. In the case of carbon impurities, the removal of carbon from solid solution occurred between 250 and 300 °C. This process took place in irradiated and unirradiated niobium containing carbon, although to a larger extent for the irradiated material.

The trapping of interstitial impurities at radiation-produced defects is believed to be responsible for radiation-anneal hardening, in which a further increase in strength is observed when irradiated metals are annealed at low temperatures. Additional evidence for this point of view is presented; a decrease in the carbon content caused a corresponding decrease in the magnitude of the radiation-anneal hardening. Also, the hardening caused by carbon trapping was found to be consistent with the kinetics of carbon migration. Dislocation channeling (the sweeping away

of radiation-produced defect clusters by slip dislocations) was absent in the radiation-annealed niobium. This is consistent with the idea that the defect clusters are stronger barriers to dislocation motion when interstitial impurities are trapped at the defect clusters.

The dislocation channeling phenomenon in irradiated niobium was investigated in detail using diffraction contrast and stereo transmission electron microscopy techniques. The strain in the channels was estimated from the offset in microstructural features where they were crossed by the channels; the strain corresponded to the passage of several dislocations per atomic plane. The crystal geometry was also determined; in most cases the channel plane was {110}. A discussion is given of possible mechanisms of the channeling.

Finally, some comments concerning the theory of dispersed barrier hardening are given, with particular application to the effect of the barriers on the temperature dependence of yielding.

METALS AND CERAMICS DIVISION

The Oak Ridge National Laboratory program on radiation effects in fuel cladding materials includes studies of the high-temperature irradiation embrittlement and other technologically important aspects of radiation damage at high neutron fluence. The work centers around Types 316 and 304 SS, Incoloy 800, Hastelloy N, and the effects of titanium concentration on radiation damage. Associated with the program are studies

of the effects of tubing fabrication variables on the properties of Types 304 and 316 SS and related compositions.

The results of postirradiation creep-rupture testing of Types 314 and 316L SS indicate that the properties are dependent upon the irradiation temperature. As the irradiation temperature is increased in the range 550 to 750 °C, the creep rate and ductility increase. We believe this is due mainly to thermal aging. A few tests on the titanium-modified Type 316 SS compositions indicate the alloy to be stronger and more ductile for similar irradiation and testing conditions.

Further postirradiation testing of the large heat of Type 304 SS containing 0.2% titanium shows that its resistance to high-temperature embrittlement is improved over standard Type 304 SS. A study of the precipitates and precipitation processes in titanium-modified Type 304L SS indicates that the size of the small (500 Å diam) precipitates is reasonably stable at 700 °C. Some evidence has been obtained that sulfur exists in high concentrations in the titanium-bearing precipitate particles.

Fabrication of the first experimental lots of tubing from both the high- and low-carbon titanium-modified Type 304 SS heats is nearing completion. These studies indicate that both compositions can be satisfactorily fabricated.

The resistance of the modified Type 304L SS to helium embrittlement under creep-rupture testing has been demonstrated. Thus far, we have examined the effects of cyclotron-injected helium to concentrations of 20 ppm.

Annealing of EBR-II driver fuel cladding after irradiation indicates a strong dependence of the annealing behavior of voids on their initial size. Voids of 187 Å maximum diameter are removed in 1 hr at 700 °C, whereas 800 °C is required to anneal voids 300 Å in diameter.

Incoloy 800 specimens that have various carbon and titanium contents and grain size have been irradiated at 700 °C and the postirradiation tensile and creep testing is underway. As we expected from our previous experience with stainless steels, there are important effects of titanium and carbon concentration in addition to the expected effects of grain size.

We have recently begun an electron microscopic examination of unalloyed vanadium irradiated as fuel cladding in EBR-II at 630 °C to a fast neutron fluence of 1.7×10^{22} n/cm². Voids were observed in some of the specimens having sizes in the range 70 to 220 Å in diameter.

The effects of titanium concentration on the postirradiation creep-rupture properties of laboratory and small commercial heats of Hastelloy N have been found to be self-consistent.

PACIFIC NORTHWEST LABORATORYIRRADIATION DAMAGE
TO REACTOR METALSREACTOR METALS RESEARCHIrradiation Facilities Operation

A total of 182 specimens were discharged from the ETR at the conclusion of Cycle 95. Included were tensile, creep, and corrosion specimens.

Tensile data reports have been recorded, processed, and stored in the Radiation Effects on Metals (REM) computer program for 5597 individual tests.

Irradiation Damage to Stainless Steel

The tensile data for AISI Type 348 SS irradiated in the G-7 hot water loop to 8.11×10^{21} ($E > 1$ MeV) have been evaluated. Tests were made at RT, 300, 500, 650, and 750 °C. The data show substantial increases in strength and losses in ductility.

Creep rupture testing in AISI Type 304 SS specimens was initiated. Tests at 540 °C show about a four-fold loss in rupture life after irradiation in the hot water loop to approximately 8×10^{20} ($E > 1$ MeV).

Transmission Electron Microscopy of
Irradiated Austenitic Stainless Steel

Electron microscopy was completed on samples cut from an EBR-II control rod thimble. The specimens of AISI Type 304 SS were irradiated over a

fluence range from 0.03 to 5×10^{22} n/cm² ($E > 0.1$ MeV). The irradiation-produced cavity size, number density, and the material microstructure, were a strong function of both irradiation temperature over the range 704 to 866 °F and fast fluence above 10^{22} .

Irradiation Damage to Nickel-Base
Alloys

Stress-aging treatments were initiated on a stainless steel alloy. Room temperature tests showed that increases of 450% in Proportional Elastic Limits (PEL) and 87% in 0.2% YS could be obtained while retaining 49% total elongation (versus 77% for the annealed condition). Better ductility, 71.5% total elongation, could be achieved with smaller increases in strength levels; namely, 300% in PEL and 53% in 0.2% YS. Tests at 1000 °F showed that the PEL could be increased 180%, and the 0.2% YS by 100% while maintaining 35.3% total elongation (versus 42.1% in the annealed condition).

In-Reactor Measurements of Mechanical
Properties

In-reactor and unirradiated control creep tests conducted at 1000 °C on Hastelloy-X were correlated with published postirradiation and control creep data at lower temperatures, by use of the Larsen-Miller rupture-life parameter. A faster creep rate was

observed for in-reactor measurements on Zr-2 at 300 °C and 11.6 kg/mm² stress; whereas, a slower creep rate was observed during in-reactor measurements on Zr-2.5 Nb at 300 °C and 58.3 kg/mm² stress.

Grain Boundary Stabilization

Nickel powders with and without helium have been compacted at pressures of 10, 30, and 50 kbars and sintered at 600 and 800 °C. The material containing helium showed improved sintering characteristics over the as-received material. Both materials recrystallized during the sintering treatments.

Nickel foils of different purity and grain size are being produced for elevated temperature irradiation.

Fracture Studies

Fracture toughness (K_{IC}) values were determined between -160 and -40 °C for ASTM A533, Grade B, Class 1 steel. Measurements were made by using DCB specimens up to 2 in. gross thickness. It was found that K_{IC} varies from about 40 ksi $\sqrt{\text{in}}$ at -160 °C to about 60 ksi $\sqrt{\text{in}}$ at -80 °C.

Initial work is being conducted to determine the contribution of deformation twinning as a mechanism for crack initiation in Zircaloy-2. Current mechanisms, based on defect impedance to slip or the formation of brittle hydrides, do not explain the saturation Δ loss in fracture toughness at low radiation exposures.

High Pressure Studies

The high pressure forms of Zr, PbO₂, and SiO₂ (coesite) show excellent radiation stability after irradiation to 2×10^{19} n/cm² (> 1 MeV). At the same fluence, cubic InTe shows a slight amount of reversion (~30%), while the high pressure form of phosphorous was rendered amorphous. Monoclinic B₂O₃ was converted to an amorphous structure after irradiation to 3.6×10^{20} n/cm².

In-Reactor Corrosion of Zirconium Alloys

Zirconium alloy specimens have been delivered to the Advanced Test Reactor for a "clean loop" corrosion test to differentiate between water chemistry and fission fragment effects on accelerated in-flux corrosion of zirconium alloys.

Specimens of the following materials have been prepared for in-reactor corrosion studies in the ETR G-7 loop: three lots of NPR Zircaloy-2 in as-etched and thick-film surface conditions; Inconel 600-Zircaloy-2 transition joints; Inconel 600; 304 and 348 SS.

FAST REACTOR SUPPORTING STUDIES

Fast Neutron Mechanisms

A significant accelerating effect of cold work on the hydrogen diffusion and gas evolution characteristics

of AISI 304L SS has been observed and studied as a function of plastic strain and deformation temperature. The results show a definite correlation between the enhanced hydrogen mobility in cold-worked material and the deformation-induced martensite content; and they reflect an apparent, high, intrinsic hydrogen diffusivity for the body-centered cubic martensitic phase.

Test Facilities

The facility to test irradiated and nonirradiated candidate structural and cladding materials for the FFTF is complete, except for installation of heating and ventilating equipment, and equipment is being moved in place. Creep tests of as-fabricated control specimens of 304 SS have been completed at 1200 °F (649 °C); tests at 1000 °F (538 °C) have been started. Negotiations with the vendor, regarding additional equipment needed to bring the liquid metal loop support section up to design requirements, have been completed. An inert atmosphere glove-box with vacuum oven for handling high purity alkali metals has been received and is being installed. A thermocouple calibration chamber, utilizing sodium as a heat transfer medium, has been placed in service.

LMFBR Fuel Cladding Information Center

The LMFBR Fuel Cladding Information Center, which utilizes a computerized storage and retrieval system incorporating the REM Program (Radiation

Effects to Materials), has been established at Battelle-Northwest. The purpose of this center is to provide complete information on LMFBR cladding material variables in the major areas of primary materials processes, composition, fabrication, properties, quality, fuel element fabrication, and irradiation history.

A computer program for storage and retrieval of information pertaining to manufacturing variables has been designed, and the terms related to manufacture have been identified and fields established. A manufacturing processes flow sheet for claddings and a questionnaire to aid in the collection of cladding manufacturers information has been prepared. An analysis of variables using existing information in the REM program file has been initiated.

RADIATION EFFECTS ON METALS - 05 PROGRAM

Isothermal annealing experiments of irradiated nickel show that the large voids grow at the expense of the smaller voids during annealing at 600 °C. Higher annealing temperatures produce very inhomogeneous disappearance of the voids which may be a result of grain boundary migration. The voids completely disappear during annealing at 800 °C. Nickel irradiated at 50 °C and annealed at 600 °C, does not show any voids. The annealing behavior of single crystal and polycrystalline rods of irradiated molybdenum have

been analyzed and compared. Polycrystalline material exhibits simultaneously a large lattice parameter recovery and stored energy release at 150 to 200 °C, while the length change is relatively slight. Single crystals, however, show almost no lattice parameter recovery and very little evolution of stored energy when annealed in the same temperature range. The interstitials and vacancies appear to anneal independently of one another without appreciable mutual annihilation. Accordingly, the so-called Stage III annealing in irradiated molybdenum may consist of both reactions proceeding simultaneously. From the behavior of the interstitial defects in the polycrystalline samples, a higher concentration of trapping sites is postulated. Whether these are a

result of difference in purity or a difference in the distribution of impurities is not yet clear.

ATR GAS AND WATER LOOP OPERATION AND MAINTENANCE

PNL is continuing to recommend fabrication and welding procedures for the critical component for the ATR Gas Loop. In addition, considerable research work is continuing in reference to problems associated with some of the super alloys that will be used in the final construction of the Gas Loop.

Programs for use of the ATR Water Loop are being completed, and data packages are being submitted to Idaho Nuclear Corporation for use of the ART Water Loop facility.

WESTINGHOUSE ELECTRIC CORPORATION

The preirradiation testing of the tensiles, Charpy-V notch, drop weight, 1X WOL, 1T and 2T Compact K_{IC} specimens fabricated from the 12 in. thick plate of A533 Grade B Class I steel was completed this report period.

The irradiation of the second three capsules continued in the Babcock and Wilcox Test Reactor (BAWTR). The total exposure accumulated by the capsules to date is about 100

EFPD or about 70% of the target exposure of 140 EFPD.

Testing of the irradiated Charpy-V notch and tensile specimens from the European forging grade steel, the 8 in. thick plate of A533 Grade B Class I steel, and the weldment (both weld and HAZ) was initiated. A considerable shift in the 30 ft-lb fixed energy transition temperature was exhibited for three of the four materials. The HAZ material appeared somewhat irradiation insensitive.

THE EFFECTS OF IRRADIATION ON THE TENSILE PROPERTIES
OF EBR-II TYPE 304 STAINLESS STEEL

Argonne National Laboratory

R. Carlander

Argonne National Laboratory has been engaged in a program to determine the effects of irradiation on the mechanical properties of Type 304 SS used in EBR-II as part of its responsibilities for operating the reactor. Results of early work in this program were given for tube burst tests conducted on specimens of irradiated driver fuel cladding.⁽¹⁻⁴⁾ As an extension of the tube burst investigations, the tensile properties of irradiated Type 304 SS from the guide thimble of EBR-II Control Rod No. 12 have been determined.

Control Rod No. 12 was located in Row 5, Sector C, Position 3 of EBR-II and accumulated a maximum fluence of approximately 5×10^{22} at temperatures between 371 and 480 °C during its residence in the reactor. The guide thimble, which served as a channel for the sodium coolant, was hexagonal in cross section with a dimension of 2.290 in. across the flats. It was constructed from 0.037 to 0.045 in. thick sheet stock. Tensile specimens were prepared from two of the six flats at 2 1/2 in. intervals with a Tensilkut machine using a solid carbide cutter rotating at 20,000 rpm. The specimens had a gage length of 1 in. and a gage width of 0.208 to 0.210 in.

The specimens were tested at a constant crosshead speed of 0.02 in./min and at temperatures of 450, 550, 650, and 750 °C. The total strain was determined by measuring the distance between gage marks which were 1 1/2 in. apart prior to fracture. The results are given in Table 3.1 and Figures 3.1 and 3.2.

EFFECT OF IRRADIATION ON STRENGTH

The strength of irradiated Type 304 SS increased with fluence but decreased with increased irradiation temperature; the magnitude of the increase in strength decreased as the test temperature was raised from 450 to 750 °C. These observations were indicated by the following:

- The yield and ultimate strengths increased with increasing fluence—the yield strength increasing at a faster rate than the ultimate strength. At a fluence of approximately 4.8×10^{22} , the yield and ultimate strengths of material tested at 450 °C were about the same—95 ksi versus 98 ksi (Table 3.1, Figure 3.1).
- The average irradiated yield strength of material irradiated to a comparable fluence and tested at 450 °C was approximately 80 ksi

TABLE 3.1. Tensile Properties of Irradiated EBR-II Type 304 SS

Test Temp., °C	Calc. Fluence, n/cm ² x 10 ²²	Irrad. Temp., °C	Yield Strength, ksi	Tensile Strength, ksi	Uniform Strain, in./in.	Total Strain, in./in.
450	--	--	24.2	62.1	0.256	0.266
450	0.4	460	35.9	63.4	0.184	0.193
450	2.0	480	65.4	73.6	0.058	0.075
450	2.0	480	65.4	74.6	0.075	0.083
450	2.0	371	74.7	76.7	0.025	0.037
450	2.0	371	85.9	89.1	0.027	0.040
450	4.8	420	95.3	98.3	0.020	0.027
550	--	--	18.9	55.7	0.222	0.234
550	<0.1	460	20.9	48.1	0.200	0.209
550	<0.1	460	25.6	55.1	0.197	0.208
550	0.6	480	41.7	57.2	0.133	0.149
550	0.6	480	41.6	55.6	0.130	0.141
550	2.0	371	69.4	70.9	0.020	0.032
550	3.6	460	70.3	74.0	0.021	0.025
650	--	--	22.9	39.4	0.138	0.194
650	0.4	460	25.3	37.1	0.090	0.110
650	1.4	480	32.9	37.5	0.049	0.067
650	1.4	480	34.8	40.8	0.046	0.071
650	2.6	480	37.6	41.4	0.029	0.035
650	2.6	480	35.8	39.4	0.025	0.039
750	--	--	17.9	23.9	0.116	0.214
750	<0.3	460	15.0	18.0	0.057	0.096
750	1.0	480	15.4	18.4	0.056	0.096
750	1.0	480	15.4	19.8	0.067	0.092
750	2.6	480	20.1	21.5	0.018	0.039
750	2.6	480	20.5	22.6	0.020	0.034

after irradiation at 371 °C compared to about 65 ksi after irradiation at approximately 480 °C (Table 3.1).

- The increase in yield strength of material irradiated between approximately 2.6×10^{22} at a com-

parable irradiation temperature was approximately 40 ksi at a test temperature of 450 °C compared to approximately 13 and 2 ksi, respectively, at test temperatures of 650 and 750 °C (Table 3.1).

FIGURE 3.1. The Effect of Fast Fluence on the Strength at 450 °C of Type 304 SS from EBR-II Control Rod Thimble

FIGURE 3.2. The Effect of Fast Fluence on the Ductility at 450 °C of Type 304 SS from EBR-II Control Rod Thimble

EFFECT OF IRRADIATION ON DUCTILITY

The ductility of Type 304 SS decreased with increased irradiation and with increased test temperature; however, in contrast to the observed recovery of strength as the test temperature was increased from 450 to 750 °C, the irradiated ductility was low at all test temperatures. For the same test temperature, irradiation at low temperatures resulted in lower ductilities than irradiation at high temperatures. These observations were indicated by the following:

- The total elongation of material irradiated at comparable temperatures and tested at 450 °C was about 19% after irradiation to approximately 4×10^{21} compared to about 8% after irradiation to approximately 2×10^{22} (Figure 3.2, Table 3.1).
- The total elongation of material irradiated to a comparable fluence and tested at 450 °C was about 4% after irradiation at approximately 371 °C compared to about 8% after irradiation at approximately 480 °C (Figure 3.2).
- The ductility of material irradiated to approximately 2×10^{22} at 371 °C was about the same as that of material irradiated at a slightly higher temperature (~ 50 °C) but to a fluence approximately 2 1/2 times greater (Figure 3.2).
- The total elongation was between 2 and 4% at test temperatures of 450, 550, 650, and 750 °C after irradiation to fluences greater

than approximately 2×10^{22} at comparable irradiation temperatures (Table 3.1).

CONCLUSIONS

The observed increase in strength and decrease in ductility were not unexpected since this has been observed for Type 304 SS irradiated to lower fluences. The changes in property values, however, do not appear to have peaked after irradiation to 5×10^{22} and likely indicates further significant loss of ductility at higher fluences. In addition, the lower ductilities and higher strengths occurred in areas where the void volume fractions reported by S. D. Harkness in this report were low. This latter observation indicates that under these experimental conditions, void formation may not affect the fracture mode of Type 304 SS.

EXAMINATION OF IRRADIATED TYPE 304 SS AND V-20 wt% Ti BY TRANSMISSION ELECTRON MICROSCOPY

S. D. Harkness

CONTROL-ROD THIMBLE

A Type 304 SS control-rod thimble irradiated to a peak fluence of 5×10^{22} n/cm² has been examined by transmission electron microscopy (TEM). Samples were taken from five positions along the thimble, as indicated in Figure 3.3. The observed microstructures in the high-flux region were similar to those reported^(5,6) earlier for Type 304L SS cladding.

This observation is important in that it implies that low levels of stress during irradiation may not be important in determining the resultant microstructure. In general, this microstructure is characterized by a homogenous distribution of polyhedral voids, a high density ($\sim 10^{15}/\text{cm}^3$) of Frank loops, and the lack of any cavities along grain boundaries. The microstructures observed in the control-rod thimble are presented in Figure 3.3 as a function of estimated fluence, estimated temperature, and the change in material density measured by immersion techniques. Note that the peak swelling, as measured by immersion density, did not occur at the position of highest fluence; thus, the importance of temperature in the void formation phenomenon is apparent.

Void-volume fractions were measured from the electron micrographs and compared with the immersion-density results in Figure 3.4. The actual values are listed in Table 3.2. The lack of agreement at smaller void sizes and lower irradiation temperatures is not fully understood. One possible explanation may be the inability of the electron microscope to resolve voids smaller than 30 Å. This factor would be of increased importance as the average void size decreased. A second uncertainty in the TEM measurements is the estimate of foil thickness.

The high density of dislocation loops formed at the lower irradiation temperatures should be emphasized. Figure 3.5 shows an area where the

FIGURE 3.3. A Summary of the Microstructural Results Obtained on the Control Rod Thimble

FIGURE 3.4. A Comparison of Density Changes in Irradiated Type 304 SS as Measured by Transmission Electron Microscopy and Immersion Techniques

loops are in good contrast. This type of microstructure is in good agreement with the very high yield strengths reported for this material by R. Carlander in this report.

V-20 wt% Ti

Samples of V-20 wt% Ti alloy irradiated to a fluence of 2×10^{22} n/cm² at a temperature of about 550 °C have been examined by TEM. The observed microstructures showed only a few dislocation loops and a polygonized dislocation structure. Part of the dislocation density may have been introduced during specimen preparation. Figures 3.6 and 3.7 are typical of microstructures seen in this alloy. These results are in sharp contrast with those obtained for Type 304 SS, in which large numbers of voids and dislocation loops would have formed under similar irradiation conditions.

The microstructural observations on the V-20 wt% Ti are in agreement with the tensile results for the same material reported by R. Carlander. He found little change with irradiation

TABLE 3.2. A Comparison of Immersion Density Results with Void Volume Measurements by Transmission Electron Microscopy

Sample	Est. Fluence, n/cm ² x 10 ²²	Est. Temp., °C	Void Volume Fraction (TEM), %	Measured Density	Percent Swelling from Density Measurement
1	2.4	375	0.13	7.853	0.53
2	4.5	410	0.21	7.795	1.26
3	4.1	450	0.34	7.763	1.67
4	3.1	470	1.06	7.804	1.15
5	2.0	480	a	7.873	0.28
6	1.0	480	a	7.890	0.06
Unirradiated Standard	0	0	0	7.895	0

a Not yet measured

FIGURE 3.5. Typical Microstructure of Irradiated V-20 wt% Ti Showing Dislocation Substructure ($nvt = 2 \times 10^{22}$, $T_{Irr.} = 550 \pm 50$ °C)

FIGURE 3.6. Typical Microstructure of Irradiated V-20 wt% Ti. Note presence of dislocation loops. ($nvt = 2 \times 10^{22}$ n/cm², $T_{Irr.} = 550 \pm 50$ °C)

FIGURE 3.7. A Micrograph Showing the Defect Structure from a Section of the Control Rod Thimble ($nvt = 2.4 \times 10^{22}$, $T_{Irr.} = 375^\circ C$)

in the mechanical properties after testing at both high- and low-strain rates.

A MODEL OF VOID FORMATION

S. D. Harkness

Void formation in structural materials subjected to fast-neutron bombardment has become a problem of major concern to fast reactor development. A model of void formation was made in an effort to understand the phenomenon and, as a result, to control void formation. Both nucleation and growth of voids are considered.

NUCLEATION

The vacancy cluster is the most stable configuration for point defects in all fcc materials when only a few defects are present. As more vacancies condense on the cluster, one of several secondary defects may become a more stable structure. For fcc materials, these secondary defects include the void, stacking fault tetrahedra, Frank loop, and perfect loop. The energy of formation of each of these defects is roughly independent of temperature, but will be a function of defect size, modulus, and the stacking fault and surface energy of the material.

For materials irradiated in a fast flux, however, two important differences exist. The first is that the damage event almost instantaneously creates a vacancy-rich core that can easily agglomerate into a cluster, which in effect allows the material to start with a cluster size of about $9 \text{ \AA}$ in diameter. The second major

discrepancy is that fast-neutron irradiation generates gas homogeneously in the material. This gas then may diffuse into the small clusters and thus lower the energy of void formation. This phenomenon has been observed in quenched materials by Clarebrough⁽⁷⁾ who observed void formation in copper and silver only, when each was quenched from a hydrogen-bearing atmosphere. One would thus expect that, for materials in which the void was not the secondary defect of lowest energy, gas generated by irradiation could eventually stabilize void formation.

Voids due to irradiation have been observed in three fcc materials: nickel, aluminum, and austenitic stainless steel. These voids occurred after fast-neutron irradiation in the same homologous temperature range ($1/4$ to $1/2 T_{mp}$). The energy-of-formation curves for secondary defects in these three materials have been calculated by using the equations given by Kuhlmann-Wilsdorf.⁽⁸⁾ For nickel, the void is stable up to very large sizes, which implies no stabilization by gas atoms should be needed. This prediction has been borne out by the work of Mastel and Brimhall⁽⁹⁾ who found voids in nickel after fluences of only 10^{19} n/cm^2 . For aluminum, the defect energies of formation for the void and the loop are quite similar, which again implies that little gas should be needed for void formation. Experimental verification⁽¹⁰⁾ was obtained from examinations of aluminum irradiated in the HFIR reactor to fluences less than 10^{22} n/cm^2 .

In austenitic stainless steels, on the other hand, the lowest energy form up to quite large sizes is the tetrahedra, while the void is the least stable. The defect energies for this material are presented in Figure 3.8. A higher gas concentration would be required to stabilize void formation. This is apparently verified experimentally since voids are not observed in austenitic stainless steel until fluences in EBR-II of about 10^{22} n/cm².

VOID GROWTH

To examine the importance of possible influences on void growth,

FIGURE 3.8. Defect Energies in Austenitic Stainless Steels

it is informative to consider the changes in free energy accompanying a cavity expansion from radius x to $x + dx$. The increment in free energy for this change is given by

$$dF = -4\pi x^2 dx \left(RT \frac{\rho}{M} \ln \frac{N_V}{N_{V_0}} + P_G + \sigma_A \right) + 8\pi x dx \gamma \quad (3.1)$$

so that at equilibrium

$$RT \frac{\rho}{M} \ln \frac{N_V}{N_{V_0}} + P_G + \sigma_A = \frac{2\gamma}{x}, \quad (3.2)$$

where

R = universal gas constant

$$= 8.3 \times 10^7 \frac{\text{ergs}}{\text{cm}^2 \text{ sec}}$$

T = absolute temperature,

ρ = density of material,

M = molecular weight of material,

N_V = number of vacancies/cm³ present
 $= C_V + N_{V_0}$,

C_V = concentration of vacancies in lattice due to neutron irradiation,

N_{V_0} = vacancies in thermal equilibrium,

P_G = internal gas pressure in cavity,

σ_A = 1/3 applied tensile stress for isotropic material,

and

γ = surface energy ≈ 1000 ergs/cm².

This expression shows a basic difference between the two types of cavities discussed in the literature.

Voids are the result of a dominant vacancy supersaturation term, while bubbles exist under conditions in which the internal gas-pressure term is the most important.

A calculation of the magnitude of the vacancy supersaturation term for austenitic stainless steel under EBR-II conditions will emphasize the importance of the term. The steady-state vacancy concentration due to the neutron irradiation must first be calculated, which involves solution of the equation

$$\frac{\partial C}{\partial t} = D_V \nabla^2 C_V + \phi \sigma_D - \beta D_V D_I C_V C_I.$$

If recombination is treated as a perturbation, the following solution for the vacancy concentration is found:

$$C_V = \left[\phi \frac{\sigma_D}{15D_V} (r^2) - \frac{\beta (\phi \sigma_D)^2}{72D_V} \right] N_O, \quad (3.3)$$

where

C_V = concentration of vacancies,

C_I = concentration of interstitials,

ϕ = instantaneous flux $\approx 3 \times 10^{15}$ n/cm² for EBR-II,

ϕ_D = displacement cross section $\approx 3 \times 10^{-22}$,

D_V = vacancy diffusion coefficient for γ iron $\approx e^{-32,250/RT}$,

D_I = interstitial diffusion coefficient,

r = sink spacing,

β = recombination parameter

$$= \frac{\pi^2}{R_O^2 D_V},$$

R_O = recombination distance,

and

N_O = number of atoms/cm³.

To determine the steady-state vacancy concentration at any particular flux and temperature, the mean free path between defects is assumed to be the sink distance.

Thus, for irradiation at 800 °K at a flux of 3×10^{15} n/cm² and with a sink distance of 2×10^{-5} cm, one finds

$$C_V = 1.2 \times 10^{-8} / \text{cm}^3 N_O.$$

The concentration of vacancies in thermal equilibrium at 800 °K is

$$N_{V_O} = N_O e^{-32,000/RT} = 2.1 \times 10^{-9} N_O;$$

therefore, the vacancy supersaturation term at 800 °K is equal to 230,000 psi, or much larger than any possible applied stress or internal gas pressure. This result then implies that only the vacancy supersaturation should be important in cavity growth at 500 °C.

As the temperature is increased, however, the vacancy supersaturation term goes rapidly to zero. For example, at 1000 °K

$$D_V = 10^{-7} \text{ cm}^2/\text{sec},$$

$$C_V = 2.4 \times 10^{-10} N_O,$$

$$N_{V_O} = 10^{-7} N_O,$$

and therefore

$$\ln N_V/N_{V_O} \approx \ln 1 = 0.$$

At lower temperatures, the vacancy supersaturation term becomes very high under EBR-II conditions. The vacancy mobility, however, is severely decreased with $D_V = 10^{-18}$ at 400 °K. The structure seen experimentally at low temperatures is the black dot observed frequently in irradiated samples. The excess vacancies could well be tied up in this defect.

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THE EFFECT OF RADIATION ON THE MECHANICAL PROPERTIES
OF CLADDING MATERIALS

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OBJECTIVE OF PROGRAM

The objective of this program is to determine the extent and nature of the radiation-induced changes in the mechanical properties of AISI Type 347 SS and other potential cladding materials for liquid-metal-cooled fast reactor applications.

APPROACH BEING USED

Type 347 SS specimens will be irradiated in the ETR to fast fluences ranging from 5×10^{21} to 30×10^{21} n/cm² ($E > 1$ MeV). Irradiated tensile specimens will be mechanically tested at room temperature and at 315 and 750 °C. Special emphasis will be directed toward investigating the ductility loss at elevated temperatures found in irradiated stainless steels and nickel alloys. This ductility loss is characterized by intergranular failure. Investigation of this intergranular fracture at elevated temperatures will be carried out by use of metallography, X-ray diffraction, chemical analysis, transmission electron microscopy, fractography, and electron-beam microprobe analysis. Stress-rupture tests will be performed at 650 °C at different stress levels. Also, cyclic-strain fatigue tests will be

performed on irradiated Type 347 SS specimens at room temperature and at temperatures (~ 650 °C) where the irradiation-induced embrittlement has been found. In addition, the effect of fast-neutron irradiation on the impact strength of Type 347 SS will be determined by testing Charpy-V notch and Izod specimens after fast fluences of 5.75×10^{21} and 30×10^{21} n/cm² ($E > 1$ MeV).

The ductility loss observed at elevated temperatures in irradiated materials has been found only for nickel-containing austenitic stainless steels and nickel-base alloys. Consequently, it appears that nickel bears some direct relationship to this irradiation-induced elevated-temperature embrittlement. It may be that the nickel-containing alloys are extremely susceptible to helium-induced intergranular failure or that the irradiation may produce other transmutations which, in combination with nickel, may cause elevated-temperature embrittlement. To further investigate this elevated-temperature embrittlement, it is planned to irradiate selected materials in the ETR and test them at elevated temperatures. The materials to be irradiated are pure nickel (99.999%), Nickel 200 (which contains about 1 wt% of other elements),

a nickel-free manganese austenitic stainless steel (Tenelon), and two cobalt alloys. One of the cobalt alloys will contain 10 wt% nickel (Haynes Stellite 25), while the other cobalt alloy will be nickel-free (UMCo-50). Testing of these materials at elevated temperatures, after irradiation, is expected to show the effect of nickel content on elevated-temperature embrittlement.

The elevated-temperature embrittlement in irradiated stainless steel may be caused by a diffusion-controlled process. To determine whether or not diffusion is an important factor, specimens irradiated at about 50 °C will be heated both slowly and rapidly to test temperature and then tested in tension. If the rapidly heated specimens exhibit substantially less embrittlement than the slowly heated specimens, this will indicate that a diffusion-controlled process is associated with the embrittlement behavior.

Since the elevated-temperature embrittlement in irradiated stainless steel is intergranular in nature, a better understanding of the phenomenon may be obtained by investigating the effect of grain boundaries at elevated temperature after irradiation. Single crystals of Type 347 SS (no grain boundaries) and polycrystalline specimens with elongated grains oriented parallel and perpendicular to the tensile axis will be irradiated and tested at elevated temperatures.

PROGRESS REPORTED PREVIOUSLY

The tensile properties of Type 347 SS irradiated in the ETR to fast fluences of 5.5×10^{21} n/cm² ($E > 1$ MeV) were reported in HW-74679.⁽¹⁾ In HW-83398⁽²⁾ and BNWL-CC-510⁽³⁾ the effects of radiation on the tensile properties of Type 347 SS after fast fluences of 16×10^{21} and 21×10^{21} n/cm², respectively, were reported. Tensile tests were performed on the irradiated material at room temperature and at 315 and 750 °C after the irradiated material had been annealed for 1 hr at 980 °C.

Results of the tensile tests at room temperature and 315 °C indicated that both yield strength and ultimate strength are increased by irradiation, with the yield-strength value approaching that of the ultimate strength. About a 50% reduction in total elongation at both room temperature and 315 °C occurred after a fast fluence of 5.5×10^{21} n/cm². No further decrease in elongation took place with increasing fast fluence to 21×10^{21} n/cm². Annealing the irradiated specimens for 1 hr at 980 °C restored the preirradiation strength properties at room temperature and 315 °C; approximately 90% of the unirradiated ductility was restored.

Tensile tests at 750 °C of specimens which had received a fast fluence of 21×10^{21} n/cm² showed no change in ultimate strength as compared to unirradiated values; however, the yield strength increased to the level of the ultimate strength and the total elongation was drastically

reduced from about 26% to only 0.5%. Annealing of two irradiated specimens for 1 hr at 980 and 1350 °C, respectively, was found to decrease the yield and ultimate strengths during testing at 750 °C. Elongation values remained about 0.5%. A specimen which had received a fast fluence of 0.5×10^{21} n/cm² had a total elongation of 15% when tested at 750 °C. This suggests that the ductility loss at 750 °C increases with increasing fast fluence. It was further found that the drastic irradiation-induced (21×10^{21} n/cm²) reduction in ductility occurred in the very narrow temperature range of 525 to 575 °C, where the total elongation was reduced from 21.2 to 1.9% (BNWL-532).⁽⁴⁾ Annealing of a highly irradiated (21×10^{21} n/cm²) stainless steel specimen at 980 °C for 1 hr resulted in almost complete restoration of preirradiation ductility when tested at 500 °C. However, testing of similarly annealed specimens at 600 °C indicated that only minimal improvement in ductility occurred (i.e., total elongation was increased from 0.7 to 3.6%, while the unirradiated value was 22.5%). It was also found that the embrittlement at 650 °C was not significantly diffusion dependent since the postirradiation ductility was not significantly affected by reducing the soak times before testing from 1 hr to 4 min. Replicas for electron fractography were taken from the fracture surfaces of both unirradiated and irradiated (21×10^{21} n/cm²) specimens which had been tested at 750 °C. The unir-

radiated specimen exhibited a ductile transgranular fracture, while the irradiated specimen exhibited an intergranular fracture.

The cyclic-strain fatigue tests at room temperature after fast fluences of 5.5×10^{21} and 11×10^{21} n/cm² indicated that for high strains the number of cycles to failure was decreased by irradiation, while for lower strains the number of cycles to failure was increased by irradiation (HW-84618).⁽⁵⁾ Testing of irradiated Charpy-V notch specimens indicated that irradiation to a fast fluence of 5.7×10^{21} n/cm² reduced the impact strength of Type 347 SS at -196 °C and room temperature, while it had no effect on the impact strength at 315 °C (HW-83398).⁽²⁾

An emission spectrographic analysis which was performed on the irradiated stainless steel (21×10^{21} n/cm²) indicated that some compositional changes had taken place as a result of irradiation. Of the major constituents, nickel, niobium, and silicon were decreased while the copper, zinc, and vanadium content was increased. Chemical analysis also indicated that 30 to 40% of the manganese was burned out by the long-term irradiation, while no significant changes in the sulfur content took place. Computer calculations based on thermal neutrons and known capture cross-section data predicted an increase in zinc content which was considerably smaller than the change indicated by the emission spectrographic analysis. This discrepancy could be due to the segregation of zinc or significant compositional changes produced by the

epithermal and fast neutrons, which were not considered in the calculations [BNWL-678⁽⁶⁾ and BNWL-710⁽⁷⁾].

WORK THIS QUARTER

Tensile tests were continued on irradiated specimens of Tenelon, UMoCo-50, Nickel 200 and Haynes Stellite 25. These tests were undertaken to determine the role of nickel on radiation-induced embrittlement at elevated temperatures. Tenelon is a nickel-free austenitic stainless steel. One of the cobalt alloys does not contain nickel (UMoCo-50), while the other cobalt alloy contains 10 wt% nickel (Haynes Stellite 25). Nickel 200 is a nickel alloy which contains a total of 1% other elements including iron, manganese, silicon and cobalt. These specimens were irradiated in

the ETR at a temperature of 50 °C. The Tenelon and Nickel 200 specimens received a fast fluence of 1.5×10^{21} n/cm² and UMoCo-50 and Haynes Stellite 25 specimens received a fast fluence of 1.4×10^{21} n/cm². The UMoCo-50 and Haynes Stellite 25 specimens were annealed for 1 hr at 800 °C before testing, and the Tenelon and Nickel 200 specimens were annealed for 1 hr at 700 °C before testing. These anneals were undertaken to remove the displacement type of radiation damage so that the radiation-induced embrittlement at elevated temperatures could be better studied. Preliminary results of tensile tests performed at elevated temperatures on irradiated and unirradiated specimens given an identical anneal are shown in Table 4.1. These tensile tests indicate that irradiation causes severe embrittlement in

TABLE 4.1. Tensile Properties of Irradiated and Unirradiated Tenelon, Nickel 200, UMoCo-50, and Haynes Stellite 25

Material	Test Temp., °C	0.2% Offset Yield Strength, 100 psi		Ultimate Strength, 100 psi		Uniform Elongation, %		Total Elongation, %	
		Unirr.	Irr.	Unirr.	Irr.	Unirr.	Irr.	Unirr.	Irr.
Tenelon	450	35.3	26.5	88.5	55.3	37.0	11.0	49.0	11.2
	550	30.3	26.0	77.5	48.5	32.5	6.4	33.0	6.5
	650	27.0	25.3	51.0	31.9	10.5	1.7	12.0	1.8
Haynes Stellite 25	450	36.1	29.9	96.3	70.3	40.0	22.2	42.0	22.8
	550	33.9	28.5	82.3	53.5	28.0	8.6	30.0	8.8
	650	32.3	29.8	69.3	40.5	14.0	4.5	17.0	4.5
UMoCo-50	450	28.9	26.0	82.5	75.1	23.0	29.7	24.5	30.7
	550	25.0	24.6	72.0	49.1	25.9	11.5	27.0	13.9
Nickel 200	400	10.8	12.4	50.6	31.6	32.0	12.4	44.6	16.4
	500	8.7	11.8	33.4	19.7	27.0	5.4	36.0	6.7

Tenelon, UMC0-50 and Haynes Stellite 25 at testing temperatures of 550 °C and above. The loss of uniform elongation is also accompanied by significant losses in ultimate tensile strength. At 450 °C, the tensile properties of irradiated and annealed UMC0-50 are comparable to the tensile properties of unirradiated material. However, irradiated and annealed specimens of Tenelon and Haynes Stellite 25 exhibit losses in both strength and ductility at 450 °C. Irradiation reduced the ultimate strength and uniform elongation of Nickel 200 at both 400 and 500 °C. However, at both of these testing temperatures the yield strength is

increased by irradiation. It is concluded that irradiation induced temperature embrittlement can occur independent of nickel content.

Stress-rupture tests at 650 °C were performed on Type 347 SS specimens which were irradiated to a fast fluence of 2.2×10^{22} n/cm² at 50 °C in the ETR. Results of the stress-rupture tests are shown in Figure 4.1. It can be seen that irradiation causes a significant reduction in rupture time.

Some specimens to be used in the study of the effect of nickel are still being irradiated in the ETR. The materials and their accumulated fast fluence are given in Table 4.2.

FIGURE 4.1. Effect of Irradiation on the Stress Rupture Properties of Type 347 SS at 650 °C

TABLE 4.2. Accumulation of Fast Fluence for Specimens Being Irradiated in the ETR at 50 °C

Capsule	Position	Material	Fast Fluence After ETR Cycle 95 (5/26/68), 10^{22} n/cm ² (E > 1 MeV)
BMI-24-2	Top	Type 347 SS	2.85
	Bottom	Type 347 SS	3.04
BMI-24-10	Top	Type 347 SS	2.66
	Bottom	Type 347 SS	2.51
BMI-24-16	Top	Type 347 SS	2.53
	Bottom	Type 347 SS	2.68
BMI-24-22	Top	Type 347 SS	2.52
	Bottom	Type 347 SS	2.80
BMI-24-25	Top	Haynes Stellite 25	0.395
	Bottom	UMCo-50	0.407
BMI-24-26	Top	Tenelon	0.282
	Bottom	Nickel	0.296

PLANS FOR FUTURE WORK

Tensile testing of the irradiated Tenelon, Nickel 200, UMC0-50 and Haynes Stellite 25 specimens will be continued. Metallographic examination of the tested specimens will be initiated. The Type 347 SS specimens that were tested in stress rupture at 650 °C will also be examined metallographically.

Cyclic stress fatigue testing of irradiated (2.2×10^{22} n/cm²) Type 347 SS specimens at 650 °C will be initiated.

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

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and R. A. Wullaert

OBJECTIVE OF PROGRAM

The objective of this program is to compare the effects of a predomi-

nantly fast-neutron flux and a mixed fast- and thermal-neutron flux on the mechanical properties of cladding materials.

APPROACH BEING USED

Tensile and fatigue specimens of the same lot of Type 347 SS irradiated in the ETR to fast fluence of up to 3×10^{22} n/cm² (E > 1 MeV) will be irradiated in the EBR-II to equivalent fast fluences. Since only a limited number of the original Type 347 SS specimens were available, additional specimens of Type 348 SS will be included in the EBR-II irradiation experiment. The Type 348 SS rod, from which specimens were machined, was obtained from the stockpile of structural materials kept at Battelle-Northwest for use in experiments to study radiation effects on structural materials. The Type 347 SS can be considered as a Type 348 SS since it meets the chemical

specifications for Type 348 SS (less than 0.10 wt% tantalum).

Tubular specimens of Type 347 SS and tensile specimens of Tenelon, UMo-50, and Haynes Stellite 25 will also be irradiated in the EBR-II. The tubular specimens of Type 347 SS are being incorporated into the program to better approximate the actual behavior of cladding materials in nuclear applications. A few tensile specimens of Tenelon, UMo-50, and Haynes Stellite 25 are included to determine the effects of a predominantly fast-neutron spectrum on their mechanical properties and to relate these effects or previous results obtained in a mixed flux.

The irradiated specimens of Type 347 SS will be tested at room temperature and at 315 and 750 °C to correlate the effects of predominantly fast neutrons, and mixed thermal and fast neutrons, on the mechanical properties. Tensile tests on specimens of Type 348 SS, Tenelon, UMo-50, and Haynes Stellite 25 will be carried out at elevated temperatures, to determine the susceptibility of these materials to radiation-induced embrittlement at elevated temperatures.

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, it is possible that the different irradiation temperatures will not allow a direct comparison of the postirradiation

mechanical properties of Type 347 SS. Therefore, a capsule will be irradiated in the ETR with specimen temperatures of 400 and 650 °C so that a direct correlation between the effects of radiation on the mechanical properties of Type 347 SS can be made. These specimens will be tested at room temperature, and at 315 and 750 °C after irradiation. In addition, some tensile specimens of Type 348 SS and tubular specimens of Type 347 SS will also be included in the radiation capsule.

PROGRESS REPORTED PREVIOUSLY

All of the test specimens for the irradiation capsules have been fabricated and the capsules have been assembled.

WORK THIS QUARTER

The capsules for the EBR-II and ETR have been inserted into their respective reactors. The type and number of specimens and irradiation conditions of these specimens in the EBR-II and ETR are given in Tables 4.3 and 4.4, respectively.

PLANS FOR FUTURE WORK

The elevated-temperature ETR capsule will be irradiated to obtain an estimated fast fluence of $5 \times 10^{21} \text{ n/cm}^2$. The EBR-II capsule will be irradiated to a maximum fast fluence of $3 \times 10^{22} \text{ n/cm}^2$.

TABLE 4.3. Specimens to be Irradiated in EBR-II

Type	Material	Number of Specimens	Irradiation Temperature, °C
Tensile	Type 347 SS	16	400
Tensile	Type 347 SS	4	650
Tensile	Type 348 SS	4	400
Tensile	Type 348 SS	4	650
Tensile	UMCo-50	6	400
Tensile	Haynes Stellite-25	2	400
Tensile	Tenelon	8	400
Tensile	Tenelon	4	650
Fatigue	Type 347 SS	4	400
Tubes	Type 347 SS	12	400

TABLE 4.4. Specimens to be Irradiated in ETR at Elevated Temperature

Type	Material	Number of Specimens	Irradiation Temperature, °C
Tensile	Type 347 SS	12	400
Tensile	Type 347 SS	6	650
Tensile	Type 348 SS	12	650
Tensile	Type 348 SS	6	400
Fatigue	Type 347 SS	6	400
Tubes	Type 347 SS	12	650

RADIATION EFFECTS ON THE MECHANICAL PROPERTIES OF TYPE 347 SS FERMI WRAPPER TUBE

J. A. DeMastry, J. W. Sheckherd,
and R. A. Wullaert

OBJECTIVE OF PROGRAM

The objective is to determine the radiation-induced changes in the mechanical properties of Type 347 SS irradiated as a wrapper tube in the predominantly fast flux of the Fermi Reactor, and to compare these changes with those for Type 347 SS irradiated in a mixed fast and thermal flux.

APPROACH BEING USED

Sheet tensile specimens will be prepared from both an unirradiated

and an irradiated Type 347 SS Fermi wrapper tube. The irradiated tube has received a predominantly fast fluence of 3×10^{20} n/cm² (E > 1 MeV). The wrapper tube is 2.5 in. square with a weld down one side, and has a thickness of 100 mils. Specimens will be cut from the longitudinal direction. The specimens will be tested in tension at room temperature and at 315 and 650 °C in the as-irradiated condition. Results from these tests will be compared with data obtained on Type 347 SS irradiated in a mixed thermal and fast flux.

PROGRESS REPORTED PREVIOUSLY

The program was initiated in FY 1968 and the first report can

be found in BNWL-678.⁽⁶⁾ Samples were machined from both an irradiated and unirradiated wrapper tube for tensile testing.

WORK THIS QUARTER

Sheet specimens machined from the Fermi wrapper tube were tested in tension at room temperature, 315 and 650 °C. The results of these tensile tests are shown in Table 4.5. For comparison, results of tensile tests at identical temperatures for Type 347 SS irradiated in a mixed flux in ETR to equivalent fast fluences are also included.⁽⁸⁾ These results indicate that irradiation in Fermi does not appear to cause significant changes in tensile properties of stainless steel at any testing temperature. The differences in strength properties of the irradiated and unirradiated specimens (i.e., ultimate strength at 650 °C) probably reflects variations of properties between different wrapper tubes. Irradiation in ETR causes

significant increases in yield and ultimate strength along with a reduction in elongation at both room temperature and 315 °C. At 650 °C, the irradiation induced property changes in the stainless steel irradiated in ETR are annealed out by the holding at the testing temperature before testing. This apparent contrast in effects of irradiation in Fermi and ETR can probably be explained by the differences in irradiation temperature. Bloom and Weir⁽⁹⁾ have shown that maximum irradiation induced strength increase results in irradiation at the 100 to 300 °C range with considerable annealing of irradiation effects taking place at higher irradiation temperature (i.e., 65,000 psi increase in yield strength at room temperature when irradiation at 300 °C and only a 12,000 psi increase when irradiated at 350 °C to equivalent fast fluences). The ETR irradiations were performed at 290 °C, while the Fermi irradiations occurred in the 320 to 380 °C range.

TABLE 4.5. Comparison of the Effects of Radiation on Tensile Properties

Temperature, °C	0.2% Yield Strength			Ultimate Tensile Strength			% Total Elongation		
	Unirr.	Thermal ^a	Fast ^b	Unirr.	Thermal ^a	Fast ^b	Unirr.	Thermal ^a	Fast ^b
		Irr.	Irr.		Irr.	Irr.		Irr.	Irr.
Room	57.0	~82.0 ^c	65.8	96.7	-	99.5	48	~34 ^c	45
Temperature	58.2		60.8	93.5		95.7			46
315	55.3	75.8 ^d	52.5	74.5	81.8 ^d	70.4	17	8.2 ^d	17
	55.7	78.0 ^d	51.0	74.5	83.0 ^d	67.9	18	7.7 ^d	19
650	18.8	~21.0 ^c	20.8	49.8	~37 ^c	35.8	25	30 ⁽³⁾	20
	18.9		52.6	52.6		35.5	27		24

a ETR

b Fermi Reactor, ~320 to 380 °C

c $\sim 3.0 \times 10^{20}$ nvt irradiated at ~290 °C, Reference 8

d $\sim 5.5 \times 10^{20}$ nvt irradiated at ~290 °C, Reference 8

PLANS FOR FUTURE WORK

This task is completed.

DETERMINATION OF THE FRACTURE TOUGHNESS OF SPECIMENS FROM THE PM-2A PRESSURE VESSEL

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and R. A. Wullaert

OBJECTIVE OF PROGRAM

The objective is to determine the fracture toughness of two types of wedge-opening-loading (WOL) specimens, designated 1X WOL and IT WOL, machined from the PM-2A reactor vessel, to evaluate brittle-fracture characteristics and fracture toughness of pressure-vessel materials.

APPROACH BEING USED

Criteria for designing pressure vessels are based on tests of large unirradiated pressure vessels and small samples of irradiated and unirradiated materials. The first opportunity to test a full-size irradiated reactor vessel arose when the Army Portable Medium Power Plant (PM-2A) at Camp Century, Greenland, was retired.

The PM-2A Reactor Vessel Test to obtain brittle fracture of an irradiated reactor vessel under a set of defined conditions including an intentionally introduced material defect is described in WAPD-TM-640. The results will be used to obtain a better understanding of conditions causing brittle fracture in an op-

erating reactor vessel and to improve criteria for reactor-vessel design and operation.

The purpose of the present program is to make further use of materials from the PM-2A vessel to evaluate brittle-fracture characteristics and fracture toughness. Cracks will be introduced into the irradiated 1X WOL and IT WOL specimens, and the cracked specimens will be tested in tension.

The fracture toughness of the irradiated specimens is unknown, and therefore approximate loads based on the results of testing unirradiated PM-2A 1X WOL control specimens will be used. The control specimens fatigue cracked at a load of 2200 lb. Initially, a fatigue load of 1800 lb will be applied to the irradiated specimens with a constant-load fatigue machine. It is expected that fatigue cracking will require about 10^6 cycles.

After initial cracking has been accomplished, the crack will be grown to a length of 0.125 in. by continued cycling. It is expected that about 20,000 additional cycles will be required for this crack growth. Visual inspection of the sides of the specimen will be used to determine crack growth. In addition, a slot 0.125 in. deep by 0.003 ± 0.001 in. wide will be machined into several of the 1X WOL specimens by electrodischarge machining (EDM).

After EDM slotting or fatigue cracking, the 1X specimens will be pulled to failure. Testing will be conducted at temperatures of

-20, and 72 °F. All specimens will be pulled at crosshead speeds of between 0.02 and 0.05 in./min. Table 4.6 shows the 1X WOL specimens of orientations designated "I" and "O" to be tested, the tensile-testing temperature, and the method of slot preparation. Table 4.7 outlines the tests to be run on the transverse specimens and the larger IT specimens.

PROGRESS REPORTED PREVIOUSLY

The program was initiated in FY 1968 and the first report can be found in BNWL-678.⁽⁶⁾

Two values of fracture toughness (K_{IC}) were determined at -20 °F. The fatigue cracking of two additional specimens was completed. From these results and previous alignment tests, it was concluded that alignment of

TABLE 4.6. Testing Schedule for 1X WOL Specimens

Temperature of Tensile Test, °F	Number of Tests to be Conducted for Indicated Orientation		Method of Preparing Defect
	"I" Orientation	"O" Orientation	
-20	3	4	Fatigue cracking
72	2	1	Fatigue cracking
-20	1	1 ^a	EDM slot
-20	2	1	EDM slot and treat ^b

a These specimens will be used to repeat any test which has produced questionable data. If they are not required as replacements, they will be cycled 500 times to 1.4 $\times$ the yield load as determined in Footnote^b and then tested as shown in Table 4.7.

b Specimens will be loaded at 72 °F and the yield point (point at which load-deflection curve becomes nonlinear) determined. The specimen will then be loaded to 1.4 $\times$ the yield. If there is no failure, the test will be terminated and the specimen retained, flushed with 1:1 HCl for 1 hr at 72 °F, water rinse, and tensile test within 10 hr.

TABLE 4.7. Testing Schedule for IT WOL Specimens and Transverse Orientation 1X WOL Specimens

Tensile Test, °F	Number of Tests to be Conducted for Indicated Orientation			Defect Preparation
	Transverse Orientation	"I" Orientation	"O" Orientation	
1X WOL Specimens				
-20	3	3	--	Fatigue cracking
IT WOL Specimens				
-20	--	1	2	Fatigue cracking

the fatigue load train was satisfactory and that ten additional specimens would be fatigue cracked.

WORK THIS QUARTER

The fatigue cracking of an additional ten specimens was completed. The fatigue crack length of 0.125 in. was produced in a minimum of 7.0×10^5 cycles and a maximum of 1.5×10^6 cycles. One specimen (PI-3) was fatigue cracked completely through because of a counter failure in the fatigue machine. The counter failure also prevented the determination of the cycles to failure.

After fatigue cracking, two specimens were tested in tension at -20°F . From the results, it was concluded that enough variation in fracture toughness existed at -20°F to warrant the use of the available specimens at that temperature.

Five more tension tests were run at -20°F . One specimen (PO-4) failed by gross deformation of the upper half with no crack extension. The other four failed in a normal manner.

Three specimens were tested in tension at 72°F . There was some difficulty with the specimen design. Two of the specimens failed at the threaded attachment to the grip, and the third by gross deformation of the upper half of the specimen with no crack extension. Therefore, the test at 150°F was eliminated because the fracture

toughness was too high for the specimen design.

The three larger IT specimens were tested at -20°F . The specimen from the outside of the vessel failed by plastic deformation. The two inside specimens failed by brittle fracture. The results shown in Table 4.8 indicate a strong dependence of the fracture toughness on the fluence. At -20°F , those specimens receiving 4.2×10^{18} n/cm^2 had an average K_{IC} value of $105 \text{ ksi}\sqrt{\text{in}}$. Those specimens receiving an average fluence of 7.7×10^{18} n/cm^2 had a fracture toughness $36 \text{ ksi}\sqrt{\text{in}}$ lower. The results from the larger IT specimens were in good agreement with the values obtained from the 1X specimens. On the basis of this limited comparison, the specimen size did not affect the values of K_{IC} .

Two specimens have been EDM slotted in preparation for the HCl acid treatment. Tests are now in progress to determine the yield (the point where the load-deflection curve becomes nonlinear). The three IT specimens have been fatigue cracked for subsequent testing.

PLANS FOR FUTURE WORK

The acid treatment will be completed and the specimens tested at -20°F . On the basis of all the previous tests, the final testing conditions for the three remaining 1X specimens will be determined.

TABLE 4.8. Results of Fracture Toughness Studies of PM-2A Materials

Specimen	Orientation	Approximate Fluence, n/cm ²	Temperature of Tensile Test, °F	Load at Fracture	Toughness, ksi√in.
1X Specimens					
PO-4	Outer 3/4 of vessel	4.2 x 10 ¹⁸	-20	10,200 ^b	K _{IC} > 112
PO-5	"	"	"	9,987	110
PO-6	"	"	"	10,480	112
PO-7	"	"	"	8,100	80
PI-7	Inner 1/4 of vessel	7.7 x 10 ¹⁸	"	5,400	48
PI-2	"	"	"	1,550 ^a	--
PI-9	"	"	"	6,220	67
PI-1T	"	"	"	6,130	65
PI-2T	"	"	"	9,060	91
PI-3T	"	"	"	5,880	48
PI-6	"	"	72	pull roll thread failure	
PI-8	"	"	"	specimen thread failure	
PO-8	Outer 3/4 of vessel	4.2 x 10 ¹⁸	"	10,500 ^b	K _{IC} > 112
IT Specimens					
PO-1	Outer 3/4 of vessel	4.2 x 10 ¹⁸	-20	15,000 ^b	--
PI-1	Inner 1/4 of vessel	7.7 x 10 ¹⁸	"	13,900 ^c	70
PI-2	"	"	"	14,300	73

a Yielded linearly to load given and then deformed irregularly until fracture at 4640 lb.

b Yielded linearly until load given. Further loading produced gross deformation of upper half of the specimen with no crack extension.

c Yielded linearly until load given. Load then relaxed linearly to 13,800 lb where brittle fracture occurred.

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IRRADIATION EFFECTS ON STRUCTURAL MATERIALS
Brookhaven National Laboratory

EFFECT OF MICROSTRUCTURE ON TENSILE
PROPERTIES OF IRRADIATED STAINLESS
STEEL

J. G. Y. Chow and R. Jones

In our previous progress report (BNWL-790), it was shown that modifying the microstructure of Type 304 SS significantly improved the elevated temperature ductility of the irradiated materials. However, metallographic examination indicated that part of the fracture followed the original austenite boundaries. Because of this, we decided to study the effect of more heavily deformed Type 304 SS.

We started out with a 1 in. diameter bar of Type 304, cold swaged it

at room temperature to 0.275 in. diameter (92% reduction in area) with the final and two intermediate anneals at 760 °C for 16 hr. The annealing temperature of 760 °C was selected to give a fully recrystallized structure that should be stable at 650 °C. The photomicrograph at 800X of the final structure is shown in Figure 5.1.

We irradiated this material to a fluence of 1.3×10^{20} ($E > 0.82$ MeV) at a temperature below 100 °C. The tensile properties of this material at 650, 750 and 800 °C in the irradiated and unirradiated conditions are given in the Table 5.1.

Although there is still an appreciable drop in ductility after

800X

*FIGURE 5.1. Type 304 SS Swaged 92%
and Annealed at 760 °C, 16 hr*

TABLE 5.1. Tensile Properties of Heavily Cold Worked* Type 304 SS

Test Temp., °C	Yield Strength, psi	Tensile Strength, psi	Total Elong., %
<u>Unirradiated</u>			
25	55,000	115,000	47
650	23,300	31,500	76
750	10,200	13,000	53
800	6,600	8,650	83
<u>Irradiated 1.3×10^{20} nvt (E > 0.82 MeV)</u>			
650	25,150	30,600	36
650	22,800	29,500	34
750	12,500	13,300	29.5
750	11,100	12,600	24
800	5,500	8,300	46.5

All test specimens tested at a strain rate of 0.2%/min.

* Cold swaged 92% with two intermediate anneals and the final anneal at 760 °C for 16 hrs.

irradiation, the irradiated specimen had average elongation values of 35, 27, and 46%, respectively, at 650, 750 and 800 °C. The materials deformed like sound engineering materials. The gage diameter was uniformly reduced throughout the gage length and with no visible sign of surface cracks.

We will continue to study this material and will extend our study to include Type 316 SS which is the designated cladding alloy for the FFTF.

EFFECT OF IRRADIATION ON TENSILE PROPERTIES OF IRRADIATED SUPERPLASTIC Fe-Ni-Cr ALLOYS

J. G. Y. Chow

Based on our study on the effect of microstructure on the high temper-

ature ductility of irradiated austenitic steels, we believe that a fine grained austenitic steel with precipitation of ferrite might be an interesting structure to test. We tried to develop this structure in Type 301 where the austenite is less stable than in Type 304. However, the martensite formed by low temperature deformation readily reverts to austenite when heated above 1000 °F.

Some of the superplastic alloys developed at International Nickel Company (INCO) by Hayden et al.⁽¹⁾ can be treated to develop the desired structure. Accordingly, we have obtained from INCO two superplastic alloys for tests. The two alloys are Fe-24.5 Cr-6.5 Ni and Ni-38 Cr-18 Fe. Specimens of these are being machined for irradiation.

We have also made a small heat of an alloy consisting of Fe-28 Cr-32 Ni-0.6 Ti.

The ingot size was 1 in. in diameter and about 4 in. long. The ingot was cold-swaged to about 0.5 in. in diameter, heated to 2100 °K for 15 min, and then swaged to 0.270 in. diameter rod. The rod was given a final heat treatment at 1875 °F for 2 hr before machining to test specimens.

Metallographic examination and chemical analysis at both ends of the rod indicates that the composition and structure were uniform. The microstructure of the alloy after the 1875 °F heat treatment is shown in Figure 5.2. It consists

of a fine grained austenitic matrix, some stringers of delta ferrite and a fine precipitation of ferrite.

The tensile properties of this alloy in the irradiated and unirradiated conditions are shown in Table 5.2. In the unirradiated condition, the alloy exhibited 175% elongation at 750 °C, whereas after an exposure of 1.3×10^{20} nvt, the elongation value at 750 °C was only 17%. Post-irradiation annealing for 2 hr at 1000 °C did not improve the ductility at 750 °C.

The fractured specimens are being examined metallographically for indication of the cause for drastic change in ductility after irradiation.

400X

FIGURE 5.2. Fe-28 Cr-32 Ni. Superplastic Alloy; The Matrix Is Very Fine Grained Austenite and the Precipitated Phase Is Ferrite

TABLE 5.2. Tensile Properties of 28 Cr-32 Ni-Fe
Base Superplastic Alloys

Test Temp., °C	Yield Strength, psi	Tensile Strength, psi	Total Elong., %
<u>Unirradiated</u>			
25	61,500	111,850	7
650	32,500	39,100	26.5
750	16,200	17,900	175
<u>Irradiated 1.3×10^{20} nvt (E > 0.82 MeV)</u>			
650	30,700	37,800	7
750	15,600	17,800	17
750*	11,100	16,200	9
800	5,700	11,500	12

* Postirradiation annealed at 1000 °C for 2 hr.
Strain rate for all tests 0.002 in./min.

RADIATION HARDENING OF IRON

P. Soo and R. Jones

In a previous progress report,⁽²⁾ it was shown that a fast neutron fluence of 10^{20} nvt changes the rate-controlled slip process in prestrained iron. The new mechanism is based on dislocation interactions with the stress fields around small vacancy loops. Some calculations by Arsenault⁽³⁾ indicate that doses of at least this order of magnitude are necessary to detect such a change. For smaller doses, the effects of the radiation damage on the slip process are masked by the intrinsic rate-controlling slip process.

The present program was undertaken to study the effects, if any, of a smaller neutron dose of 10^{18} nvt on the deformation of iron which has been given a prestraining treatment,⁽³⁻⁷⁾ no change was observed in the rate-controlling slip process. This could be caused by the tendency

of the freshly nucleated dislocations to reside in definite slip zones which rapidly become denuded of radiation-induced slip barriers. In such a case, the irradiated iron would be expected to show similar behavior to unirradiated material. By prestraining the iron prior to irradiation, uniform deformation is encouraged and the formation of defect-free zones is minimized. This would, therefore, increase the chances of observing a change in the rate-controlling slip mechanism if one occurs for a neutron dose of 10^{18} nvt.

The tensile specimens were identical to the Ferrovac E samples used in the previous study.⁽²⁾ The analysis of this is shown in Table 5.3. They each had a gage length of 1.125 in., a diameter of 0.120 in. and a recrystallized grain size of 1.2×10^{-3} in. All specimens were prestrained at room temperature to 9%

TABLE 5.3. An Analysis of the Ferrovac-E Starting Material

Element	ppm
Carbon	30
Oxygen	55
Nitrogen	5
Hydrogen	0.7
Nickel	150
Chromium	100
Manganese	10
Molybdenum	100
Aluminum	70
Silicon	60
Sulphur	40

elongation and irradiated at approximately 80 °C in the Brookhaven Graphite Reactor to an integrated flux of 10^{18} nvt ($E > 1.45$ MeV). Tensile tests were conducted on a floor-model "Instron" tensile machine at a strain rate of 3.0×10^{-4} /sec.

The nature of the stress-strain curves as a function of temperature is shown in Figure 5.3. Below 200 °K, the deformation is unstable and necking and fracture occur soon after yield. At approximately 250 °K, the work-hardening rate is relatively low but it shows a large increase for temperatures between 298 to 350 °K. At 400 °K, small regular serrations appear in the stress-strain curves which are indicative of a dynamic strain-aging effect. Periodically, the flow curve is interrupted by sudden large drops in the stress.

The 10% flow stresses (i.e., the stresses after deforming the specimens for an additional 1%) are

Neg 6-1339-68

FIGURE 5.3. The Nature of the Stress-Strain Curves for Iron Irradiated to 10^{18} nvt

given in Figure 5.4. For the low temperature tests which exhibit unstable deformation, the flow stresses were calculated from the maximum load since at this point the cross-sectional area is most accurately known.

Stress relaxation tests were performed on the specimens at 10% total elongation and the results are given in Figure 5.5. From these, activation volumes and activation energies were obtained using the

Neg 6-1340-68

FIGURE 5.4. The Effect of Fast Neutron-Irradiation on the 10% Flow Stress of Iron

Neg 6-1343-68

FIGURE 5.5. Stress Relaxation Data for Iron Irradiated to 10^{18} nvt

technique described previously.⁽²⁾ The results are given in Figures 5.6 and 5.7. All of the current 10^{18} nvt data are compared with those for the unirradiated iron and iron which has been irradiated to 10^{20} nvt ($E > 1$ MeV).

It is clear from the data in Figure 5.4 that a dose of 10^{18} nvt causes an enhanced temperature dependence of the flow stress and an increase in the athermal stress for slip.

The general behavior of the 10^{18} nvt samples is intermediate between the unirradiated and the 10^{20} nvt samples. As mentioned earlier, a fast neutron dose of 10^{20} nvt changes the rate controlling slip process in iron to one based on dislocation interactions with tetragonal distortions around small vacancy loops in the lattice. Fleischer's^(8,9) theory for tetragonal distortions predicts that the thermal

Neg 6-1337-68

FIGURE 5.6. The Effect of Fast Neutron-Irradiation on the Activation Volume for Slip in Iron

Neg 6-1341-68

FIGURE 5.7. The Effect of Fast Neutron-Irradiation on the Activation Energy for Slip in Iron as a Function of Temperature

component of the flow stress τ^* may be given by:

$$\tau^{*1/2} = \alpha + \beta T^{1/2} \quad (5.1)$$

in which T is the absolute temperature and α and β are constants. In turn, τ^* is given by $\tau - \tau_\mu$ in which τ is the applied stress and τ_μ is the athermal component of the flow system. Therefore, τ_μ must be determined before τ^* can be obtained. Normally, the value of τ_μ may be obtained from the flow stress data for which, at

high temperatures, the thermal stress becomes zero. In this range, $\tau = \tau_\mu = \text{constant}$. Unfortunately, the data for the 10^{18} nvt specimens (Figure 5.4) are complicated by the presence of the dynamic strain-aging phenomenon which causes the flow stress to rise at temperatures greater than 350 °K. However, if one estimates that $\tau_\mu = 14.0 \text{ kg/mm}^2$, then from Figure 5.8, the flow stress data closely follow the relationship:

Neg 6-1344-68

FIGURE 5.8. Plot of $\tau^{*1/2}$ Versus $T^{1/2}$ for Unirradiated and Irradiated Iron

$$\tau^{*1/2} = 11.0 - 0.54 T^{1/2} \quad (5.2)$$

which is represented by the line AB in Figure 5.4. The athermal temperature of 420 °K was obtained by extrapolating the data in Figure 5.8 to $\tau^{*1/2} = 0$.

A further prediction of the tetragonal distortion model is that the activation volume and activation energy for slip may be given respectively by:

$$V = \frac{\Delta G_0}{\tau_0^*} \left[\left(\frac{\tau_0^*}{\tau^*} \right)^{1/2} - 1 \right] \quad (5.3)$$

$$\Delta G = \Delta G_0 \left[1 - \left(\frac{\tau_0^*}{\tau^*} \right)^{1/2} \right]^2 \quad (5.4)$$

in which ΔG_0 is the activation energy at zero thermal stress and τ_0^* is the thermal stress at 0 °K. All other symbols have their previous meaning. The data in Figures 5.9 and 5.10 are plotted according to the above equations using a value

Neg 6-1338-68

FIGURE 5.9. The Activation Volume for Slip in Iron as a Function of $[(\tau_0^*/\tau^*)^{1/2} - 1]$

Neg 6-1336-68

FIGURE 5.10. The Activation Energy for Slip in Iron as a Function of $[1 - (\tau^*/\tau_0)^{1/2}]^2$

of $\tau_0^* = 121.0 \text{ kg/mm}^2$ which was obtained by extrapolating the data in Figure 5.8 to 0 °K. At the higher stress levels ($T \leq 250 \text{ °K}$), the results show a reasonable fit to the tetragonal distortion theory. It is, therefore, likely that in this temperature range the rate of slip is controlled by the presence of small vacancy loops in the lattice as was the case for the 10^{20} nvt specimens.

An interesting observation in the irradiated samples is that the loop sizes in the 10^{18} and 10^{20} nvt batches are different. This is best demonstrated by comparing the respective values of ΔG_0 . By extrapolating the low-energy line for the 10^{18} nvt data in Figure 5.10 to

$$\left[1 - (\tau^*/\tau_0^*)^{1/2}\right]^2 = 1, \text{ the value}$$

of ΔG_0 may be shown to be 1.58 eV as opposed to 0.91 eV for the 10^{20} nvt specimens. The size difference probably indicates that loop formation and growth was easier in the 10^{18} nvt specimens. This is quite likely since the two batches of samples were irradiated in different reactors under only approximately similar conditions. As far as the actual distribution of loops is concerned, the activation volume data in Figure 5.6 shows that the loops in the 10^{18} nvt data are more widely separated, which is to be expected because of the smaller amount of radiation damage.

Attention will now be directed at the mechanism controlling flow at temperatures between 250 and 350 °K. A change in the controlling

slip process at 250 °K is indicated by the abrupt change in the activation volume (Figures 5.6 and 5.9) and the activation energy (Figures 5.7 and 5.10). From Figure 5.7, the behavior of the unirradiated and 10^{18} nvt batches of specimens is similar insofar as the activation energies show a rapid increase at approximately 250 °K. It seems likely, therefore, that the same mechanism controls flow in the two cases. However, the data in Figure 5.11 show that for a given thermal stress the activation energies for the 10^{18} nvt specimens are always substantially larger. This indicates that, in some way, irradiation has strengthened the barrier controlling slip in iron between 250 and 350 °K. At the time of writing, this barrier has not been identified.

Consider now the test conducted at 400 °K. The serrated nature of the stress-strain curve is usually indicative of dynamic strain aging. Friedel⁽¹⁰⁾ has shown that the minimum temperature at which this behavior can occur may be given approximately by the expression:

$$\dot{\epsilon} = 10^9 D = 10^9 A \exp(-Q/kT), \quad (5.5)$$

in which $\dot{\epsilon}$ is the strain rate, D is the diffusion coefficient, A is a frequency factor, Q is the activation energy for the diffusion of the interstitial atom causing strain aging, k is Boltzmann's constant and T is the absolute temperature. Of the impurity interstitials present in the specimens, both hydrogen and oxygen have very limited solubility

Neg 6-1342-68

FIGURE 5.11. The Effect of Neutron-Irradiation on the Activation Energy for Slip in Iron as a Function of the Thermal Stress

in α -iron⁽¹¹⁾ and are, therefore, unlikely to be capable of causing strain aging. In the case of carbon and nitrogen, the data tabulated by Hume-Rothery⁽¹²⁾ give the values of A and Q for carbon as $6.2 \times 10^{-3} \text{ cm}^2/\text{sec}$ and 0.82 eV , respectively, and for nitrogen $3.0 \times 10^{-3} \text{ cm}^2/\text{sec}$ and 0.79 eV . By substituting the values into Equation (5.5), it may be shown that carbon theoretically causes dynamic strain aging at 407°K . In the case of nitrogen, the temperature is 398°K . This is in excellent agreement with the above experimental observation. Of the two interstitials, the greatest effect must come from the carbon because of its greater concentration.

A notable feature of body-centered cubic metals is that neutron-irradiation increases the athermal component of the flow stress. Little attention has been devoted to the identification of the athermal slip barriers in irradiated metals, but they appear to be connected with the presence of depleted zones which are introduced during irradiation. These zones are large compared with the usual type of slip obstacle and would be expected to be formidable barriers to slip. This is a fundamental property of athermal barriers. In addition, a simple calculation of the activation volume for the depleted zones indicate that they do not contribute to the thermal stress. This

may be shown as follows using the standard definition of the activation volume given by:⁽¹³⁾

$$v = b \ell X \quad (5.6)$$

in which b is the Burger's vector of the glide dislocation, ℓ is the spacing of depleted zones on the glide plane and X is the width of the zone which McRickard⁽¹⁴⁾ showed to be about $20.2 b$ for iron irradiated under conditions similar to the present specimens. The value of ℓ , in turn, may be shown to be given by $b/\sqrt{N_c}$ in which N is the number of adjacent slip planes over which a zone is an effective barrier to slip and c is the atomic concentration of zones. To a first approximation, N is equal to 20.2 . Eliminating ℓ from Equation (5.6), therefore, the activation volume becomes $4.5b^3/\sqrt{c}$. The concentration of zones may be calculated from the expression $N_0(\phi t)\sigma$ in which N_0 is the number of matrix atoms/cm³, ϕ is the neutron flux, t is the irradiation time and σ is the neutron capture cross-section. In the present case, $N_0 = 8.46 \times 10^{22}/\text{cm}^3$ and $\phi t = 10^{18}$ nvt. If one estimates that σ is of the order of 10^{-24} cm², the concentration of zones is $8.46 \times 10^{16}/\text{cm}^3$ or 10^{-6} zones/matrix atom. From this value, the activation volume for the depleted zone becomes $4500 b^3$. A comparison with the data in Figure 5.6 indicates, therefore, that these zones do not contribute to the thermal component of the flow stress in irradiated iron and are more likely

to be associated with the athermal component.

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GENERAL ATOMIC DIVISION OF GENERAL DYNAMICS
(No contribution this quarter)

IRRADIATION EFFECTS ON REACTOR STRUCTURAL MATERIALS

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HYDROGEN EMBRITTLEMENT OF STEELS

Hydrogen induced cracking of a reactor pressure vessel in service is considered most unlikely for the following reasons:

- Hydrogen induced fracture generally occurs only in high strength steels, i.e., hardnesses usually greater than R_C 25 and then at high stress levels.
- Pressure vessels when fabricated in accordance with the ASME Boiler and Pressure Vessel Code, Section III, Nuclear Vessels, have a relatively low yield strength and a high degree of toughness.
- Hydrogen induced cracking in high strength steels at design maximum stress levels requires a threshold hydrogen concentration of the order of 2 to 4 ppm (wt%) which is considered to be in excess of the concentration level possible as a result of metal-water corrosion reactions which are thought to be the major source of hydrogen in a reactor vessel in service.

However, it is possible to envision a series of events which would render a vessel susceptible, if a source for generation of sufficient quantities of monatomic hydrogen was also present. Irradiation damage as a result of exposing the material up to peak fluences, i.e., 3×10^{19}

n/cm² ($E > 1$ MeV) coupled by aging embrittlement involving nickel and/or aluminum⁽¹⁾ (e.g., ASTM A533, HY 80, A508, and A540 steels) might well render the material sufficiently brittle. An undetected flaw in an improperly heat treated region (HAZ) of a weld as a result of failure to follow Code practice coupled with an unusually high stress concentration or some complex combination of any of the above could well render the material susceptible to hydrogen induced cracking.

It was not the objective of this research to investigate complex processes by which a reactor pressure vessel might pick up several ppm of hydrogen, but rather to determine whether several of the medium strength pressure vessel steels in the irradiated condition were inherently susceptible to hydrogen induced cracking. Towards this end, plane and notched specimens were fabricated from three steels, i.e., ASTM A302B, HY 80, and A542 in their normal quenched and tempered condition. In addition, specimens of A542 were heat treated to different strength levels and microstructures to investigate the susceptibility to hydrogen induced cracking as a function of strength level and microstructure. There is some evidence that A542 steel has a very sensitive relationship between the

tempering temperature and notch toughness that leads to a disconcerting scatter in the measured values. (1, p. 15)

Stress-Rupture Testing of Unirradiated Hydrogen Charged Steels

Stress-rupture tests, i.e., constant load, were continued on notched unirradiated specimens of A542 steel

to establish base-line data for comparison with irradiated specimens. Figure 7.1 shows results obtained from stress-rupture tests conducted at two initial hydrogen concentrations (i.e., ~3.5 and 4.9 ppm) on A542 in the normal quenched and tempered condition. The data indicate that for the hydrogen concentrations considered, hydrogen reduces the ductility but has only a

Neg 68-2910

FIGURE 7.1. Time Dependent Fracture Studies Compared for Pre-irradiated A542 Steel

slight tendency towards inducing cracking. A similar effect was found in specimens that had been austenitized and furnace cooled as shown by the data in Figure 7.2. This steel can be made susceptible to hydrogen induced cracking and failure, however, as evidenced by Figures 7.3 and 7.4. For A542 steel notched specimens heat treated to R_C 25-26 containing approximately 3.5 ppm hydrogen (Figure 7.3), the lower critical stress for failure is approximately 75% of the uncharged notched tensile strength, while specimens containing initially 6 ppm can fail at stress levels approaching 50% of the ultimate strength. Figure 7.4 similarly shows in A542 specimens quenched and tempered to R_C 34-37 and charged to approximately 4 ppm hydrogen that the LCS is 35% of the uncharged ultimate strength.

Tensile Testing of Unirradiated Hydrogen Charged Steels

Plain tensile specimens of unirradiated A302B were hydrogen charged and tensile tested at room temperature and at a strain rate of 0.1/min. The effect of hydrogen concentration on the essential mechanical properties obtained from engineering stress-strain curves are shown in Figure 7.5. As is evident from the data, hydrogen has little or no effect on the engineering stress-strain curve of A302B steel. A similar lack of any effect was previously reported for HY 80 steel and for several mild steels by other investigators.^(2,3) However, the ductility as represented by percent reduction in area at fracture shows a significant decrease (from 65 to 30%) with increasing hydrogen content to 5 ppm

Neg 68-2911

FIGURE 7.2. Time Dependent Fracture Study for Preirradiated A542 Steel that Had Been Austenitized and Furnace Cooled; Hardness R_b 83-84

Neg 68-2712

FIGURE 7.3. Time Dependent Fracture Studies for Unirradiated A542 Steel that Had Been Austenitized, Water Quenched, and Tempered at 620 °C to R_c 25-26

Neg 68-2909

FIGURE 7.4. Time Dependent Fracture Study for Preirradiated A542 Steel that Had Been Austenitized, Water Quenched, and Tempered to R_c 34-37

Neg 68-2710

FIGURE 7.5. Effect of Hydrogen Content on the Tensile Properties (Ultimate and Yield Strength) of A302B Steel

as shown in Figure 7.6. Figure 7.7 shows photographs of the specimen fracture surfaces after testing at several hydrogen contents. A decrease in ductility and the ductile mode of fracture is evident.

Irradiated Specimens

Irradiation of the first assembly containing nine capsules with a total of 162 specimens of A302B, A542, and HY 80 steels, has been completed. Irradiation of a second assembly is continuing.

FATIGUE OF CLADDING AND STRUCTURAL MATERIALS

The program to determine the fatigue properties of fuel cladding materials for fast reactors (SS 304, 304 Ti modified, 316, 316 Ti modified, and Incoloy 800) and of structural materials for water power reactors (pressure vessel steels; ASTM A302B, HY 80, and A542) and for liquid metal fast breeder reactors has been discussed in previous reports.⁽⁴⁾ During the past

Neg 68-2711

FIGURE 7.6. Effect of Hydrogen Content on the Ductility (Reduction in Area) of A302B Steel

(a) Specimen #31-24 precharged, Neg 68-2708

(b) Specimen #31-46 1.1ppm H_2 . (c) Specimen #31-42 3.2ppm H_2 , Neg 68-2709(d) Specimen #31-3 5.8ppm H_2 .

5.5X

FIGURE 7.7. Fracture Surfaces of Hydrogen Charged Plain Tensile Specimens of A302B Steel

quarter, 210 specimens in two sub-assemblies have been placed in EBR-II for irradiation prior to testing.

The fatigue specimens are hour-glass shaped to reduce buckling problems from elevated temperatures and from the large strains which produce low cycle fatigue during the testing. The specimen is loaded in the axial push-pull mode and the diametral strain measured with an extensometer. From relationships between the axial strain and the diametral strain, which is measured, the axial strain is calculated and also reported. Stress and strain values which are directly comparable to the designers' usually calculated pressure stress, thermal stress, and stress concentrations, are useful for design purposes, as in ASME Code Section III. For this purpose, in strain controlled axial fatigue a stress amplitude, $S = E \frac{\Delta \epsilon}{2}$, is calculated from the total strain range, $\Delta \epsilon$, and Young's modulus E . The axial strain is computed from the diametral strain, since by definition the strain consists of an elastic and plastic component through the following relationships:

$$\frac{\Delta \epsilon}{2} = \frac{\Delta \epsilon_p}{2} + \frac{\Delta \epsilon_e}{2} = \epsilon_p + \epsilon_e = \epsilon_p + \frac{\sigma}{E} \quad (7.1)$$

$$\frac{\Delta \epsilon_d}{2} = \frac{\Delta \epsilon_{dp}}{2} + \frac{\Delta \epsilon_{de}}{2} = -\nu_p \epsilon_p - \nu_e \epsilon_e \quad (7.2)$$

where: $\Delta \epsilon$, $\Delta \epsilon_p$, and

$\Delta \epsilon_e$ = total, plastic, and elastic strain range, respectively;

ϵ_p , ϵ_e = plastic and elastic strain amplitude;

ν_p , ν_e = Poisson's ratios for plastic and elastic stress-strain;

and the suffix d indicates diametral directions. Solving for ϵ_p from Equation (7.2) and substitution into Equation (7.1) with the use of the absolute value of ϵ_d gives:

$$\epsilon = \frac{\sigma}{E} \left(1 - \frac{\nu_e}{\nu_p} \right) + \frac{\epsilon_d}{\nu_p} \quad (7.3)$$

which allows the axial strain amplitude, ϵ , to be determined from the stress amplitude, σ , from measurements of diametral strain amplitude, ϵ_d , Young's modulus, E , and Poisson's ratios for plastic, ν_p , and elastic, ν_e , stress-strain.

Specimens of the cladding and structural materials were encapsulated in inserts. Three inserts comprise a capsule containing 15 specimens. Specimens in the insert at the center location (core elevation) are thermally bonded in argon gas and are irradiated at high temperature (600 to 800 °C). Specimens in the two outer capsule locations (blanket elevations) are bonded in sodium and are irradiated at 400 °C. Seven capsules comprise a hexagonal shaped EBR-II subassembly. The first two subassemblies, X037 and X038, were loaded into the EBR-II in positions 7C3 and 7C5, respectively, on May 8, 1968. On June 15 at the end of Run 28, they had received 1126 MWd of irradiation. It is planned to remove X037 after 3000 MWd and X038 after 18,000 MWd (maximum fluences of 10^{21} and 10^{22} n/cm², $E > 1$ MeV).

Flux monitoring wires (Fe, Ni, Ti) with Al-Co in the outer inserts

were placed alongside the specimens. A few ampoules containing boron were placed in the hollow grip ends of the specimens in the outer inserts. Temperature monitors were placed in the majority of the hollow grip ends. The temperature monitors consisted of either fusible wires or small cylinders of porous self-bonded β -phase silicon carbide ("Crusilite") which has been shown^(5,6) to be a reliable temperature monitor for fast flux environments.

The sodium for bonding specimens in the outer capsule inserts was purified by cold trap and filtered at 180 °C, and zirconium gettered at 650 °C. The atmosphere in which the purified sodium was loaded into the inserts was less than 1 ppm oxygen and less than 15 ppm moisture. The oxygen content of the argon gas was less than 1 ppm.

A test enclosure has been nearly completed in which the irradiated samples can be fatigue tested at temperatures up to 1000 °C in an inert atmosphere. The pressure vessel steels are to be tested at 250 to 400 °C. The austenitic stainless steel specimens for fast reactor pressure vessels are to be tested at 320 to 650 °C with emphasis in the 480 to 650 °C range. For cladding service, the specimens are to be tested at 600 to 800 °C. The tests are to be conducted at a strain producing failure in the range of 10^3 to 10^5 cycles. The slope of the curve (stress or plastic strain versus number-of-cycles to failure) will be obtained from a limited number of tests at each set of conditions

(material, fluence, and temperature) by taking 3 points each at each end of the strain range. The significance of this result can be shown by statistical analysis since the variance of the slope of a line is minimized when the variances of the independent variables are maximized [i.e., when $(X_1 - X)$ is large]. Thus, one obtains the most precise measurement of the exponent, the plastic strain range, and the fatigue life with the minimum number of samples which are sensitive indicators of the fatigue damage⁽⁷⁾ as well as the design parameters as previously discussed.

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January 1967.

MATERIALS RESEARCH LABORATORY, INC.
(No contribution this quarter)

RESEARCH PROGRESS

Naval Research Laboratory

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IRRADIATION RESPONSE OF A 4 IN.
A533-C, CLASS 2, SUBMERGED ARC
WELDMENT

The A533-C steel is a manganese-molybdenum steel that contains 0.7 to 1.0% nickel and carries two strength classifications (50 and 70 ksi minimum yield strength for Class 1 and Class 2, respectively). Unlike the A533-B grade, which now has largely supplanted ASTM A302-B steel as the prime material for pressure vessel construction, the A533-C grade has not received ASME code approval at this time. The irradiation response of this material, however, is of significant interest and a 4 in. submerged arc

weldment of A533-C, Class 2, steel has been obtained from Lukens Steel Corporation for evaluation.

The chemical compositions and heat treatments of both parent plate and weld metal are summarized in Table 9.1. Pre- and postirradiation notch ductility characteristics are given in Table 9.2 and Figure 9.1. Charpy-V notch specimens from the plate and weld were taken with the notch oriented perpendicular to the surface in the longitudinal (RW) direction; weld metal specimens were oriented with their long axis perpendicular to the welding direction. Irradiation was performed in the Union Carbide Research Reactor (UCRR), Tuxedo, New York, using standard

TABLE 9.1. Chemical Composition and Heat Treatment
of the 4 in. A533-C Class 2 Weldment

Identification	Chemical Composition,* wt%										
	C	Mn	P	S	Si	Ni	Cr	Mo	Cu	V	Al
PLATE	0.19	1.25	0.010	0.013	0.22	0.83	0.07	0.48	0.22	-	0.021
	0.24	1.05	0.006	0.018	0.21	0.78	0.06	0.48	0.20	0.02	-
WELD	0.08	1.84	0.013	0.009	0.37	0.10	0.07	0.51	0.30	-	-
	0.13	1.68	0.016	0.015	0.33	0.14	0.05	0.48	0.27	0.02	-

* First number - courtesy of supplier
Second number - NRL determination

Heat Treatment

PLATE - Austenitized at 1650 °F, water quenched, tempered at 1225 °F, air cooled
WELD - Stress relief annealed at 1125 °F for 20 hr, furnace cooled

TABLE 9.2. Charpy-V Notch Ductility Characteristics of the A533-C Class 2 Plate and Weld Metal Before and After 550 °F Irradiation to 1.2×10^{19} n/cm² ($E > 1$ MeV)

Identification	C _V 30 ft-lb Transition Temp., °F			Apprx. Full Shear Energy Absorption, ft-lb	
	Initial	Irradiated	ΔT	Initial	Irradiated
PLATE:	Surface (Face)	-130	-	100	-
	Surface (Root)	- 60	-	96	-
	1/4 Thickness	- 50	130	108	96
	1/2 Thickness	- 50	-	108	-
WELD:	Face	~ - 20*	-	>45*	-
	Root	- 50*	-	>80*	-
	1/4 and 1/2 Thickness	- 40	165	86	59

* Courtesy of Lukens Steel Corporation

FIGURE 9.1. Notch Ductility Characteristics of a 4 in. A533-C, Class 2 Experimental Weldment Before and After 550 °F Irradiation Showing the Similarity in Performance of Base Plate Versus the Submerged Arc Weld Deposit

equipment and techniques developed by NRL for controlled temperature specimen irradiations.^(1,2) All specimens were irradiated at 550 °F to a fluence of 1.2×10^{19} n/cm² (E > 1 MeV).

It can be seen from Figure 9.1 that the preirradiation transition temperatures for the plate and weld metal are quite similar (-40 and -50 °F), although the full shear energy absorption shelf of the weld metal is significantly lower than that of the parent plate (86 versus 108 ft-lb). Since the normal Charpy-V, NDT correlation for this class material was not available, the conventional A302-B index of 30 ft-lb was used in reporting transition temperature changes. Other data on A533 material have suggested that the 30 ft-lb correlation may not always be applicable to this class of material;⁽³⁾ however, it is noted that the choice of energy index in the range of 30 to 60 ft-lb would not markedly alter the transition temperature shift values determined in this manner.

It is apparent from the Charpy-V data that both the parent plate and weld metal are quite sensitive to 550 °F irradiation with the weld metal being slightly more sensitive than the plate. The 30 ft-lb transition temperature increases were 180 and 205 °F, respectively. (A 140 to 150 °F shift would be projected for the A302-B ASTM reference plate material under the same irradiation conditions).

Recent studies of variable radiation embrittlement sensitivity in

A302-B steel⁽⁴⁾ have shown that residual copper in small quantities (approximately 0.2%) can contribute significantly to the radiation embrittlement sensitivity of this material. Since the respective copper contents of the 4 in. A533-C plate and weld metal were approximately 0.21 and 0.28%, the relatively high embrittlement sensitivity of this weldment is in good agreement with these observations. Similarly, when the irradiation response of this weldment is compared to the results obtained from the irradiation of a 7 1/2 in. A533-B submerged arc weldment,⁽³⁾ good correlation between relative embrittlement sensitivity and copper content is again apparent. In this instance, the A533-B base plate (0.12% copper) transition temperature increase following 550 °F irradiation to 1.7×10^{19} n/cm² (E > 1 MeV) was 70 °F, while its companion weld metal (0.22% copper) exhibited a transition temperature increase of 200 °F. The latter is very much in line with the transition temperature shifts recorded here for the relatively sensitive 4 in. A533-C plate and weld metal which was irradiated to a slightly lower fluence.

RELATIVE 550 °F IRRADIATION RESPONSE OF A HEAVY SECTION A533-B ELECTROSLAG WELDMENT

The irradiation response of weld metal and base plate of a commercially produced electroslag weldment has been explored. The results permit

an initial assessment of the relative performance of weld joints produced by electroslog versus submerged arc welding practices. The chemical composition, fabrication procedures, and heat treatment of the 5 3/4 in. thick A533-B, Class 1 electroslog weldment for this investigation are given in Table 9.3. A photograph of the weld joint cross section revealing the size of the fusion zone is shown in Figure 9.2. Charpy-V notch specimens taken from the quarter thickness position in base plate and weld region were irradiated at a controlled temperature of 550 °F in the UCRR. The neutron fluence received was approximately 1.6×10^{19} n/cm² (E > 1 MeV) based on data from iron wire neutron detectors included in the exposure assembly.

Postirradiation experimental results are compared against preirradiation data for plate and weld in Figure 9.3.

Referring first to the pre-exposure condition, it is noted that the base plate exhibited a lower Charpy-V 30 ft-lb transition temperature than the weld metal (-60 °F versus 15 °F). Data for both materials suggest good properties uniformity through the thickness. Nil-ductility transition (NDT) temperature determinations with a limited number of drop weight test specimens also suggest good uniformity in weld properties between surface (NDT, 20 °F) and mid-thickness positions (NDT, 40 °F). The results further denote that the Charpy-V 45 ft-lb temperature rather than the 30 ft-lb

TABLE 9.3. Chemical Composition, Fabrication Procedures and Postweld Heat Treatment of 5 3/4 in. A533-B Class I Electroslog Weldment

	Chemical Composition								
	C	Mn	P	S	Si	Ni	Cr	Mo	Cu
Plate	0.22	1.19	0.008	0.018	0.16	0.52	0.14	0.48	0.12
Weld	0.22	1.28	0.008	0.014	0.10	0.40	0.14	0.48	0.19
(Fabricator)	0.18	1.37	0.014	0.016	0.12	0.36	0.08	0.49	0.21

Fabrication Procedure

Preweld Setup: 1 3/8 in. gap between straight side walls

Filler Metal/

Flux: RACO Hi-Mn-Mo Filler, LINDE 124 Flux

Weld Parameters:

Double electrodes, 575 to 625 A, 48 to 52 V
48 in./min; oscillation with 5 sec dwell

Postweld Heat Treatment

Austenitized at 1675 to 1725 °F for 6 hr, brine quenched;
Reaustenitized at 1600 to 1650 °F for 6 hr, brine quenched;
Tempered at 1200 to 1225 °F for 6 hr, brine quenched;
Stress relief annealed at 1115 to 1135 °F for 30 hr,
furnace cooled at 40 °F/hr (max.).

FIGURE 9.2. Photograph of the Electroslog Weldment Showing the Etched Weld Metal Joint (hot 50% HCL). The Initial Gap Between the Straight-sided Base Plates Was $1 \frac{3}{8}$ in.

FIGURE 9.3. Experimental Results Developed for the 5 3/4 in. A533-B, Class 1, Electroslag Weldment. Notch Ductility Properties of the Parent Plate are Markedly Superior to Those of the Weld Metal Both Before and After 550 °F Irradiation.

temperature would be a more conservative index of the NDT temperature for all weld thickness positions. It is seen from Figure 9.3, however, that the comparison of irradiation responses is not highly dependent upon the particular index value selected.

Postirradiation test data for the listed fluence condition depict a threefold difference between the transition temperature responses of the weld metal and the base plate. Therefore, the initial difference in transition temperatures is fur-

ther compounded by neutron exposure, and it would appear that the in-service limitations of a comparable weldment would be governed by the notch ductility properties of its weld metal. Consistent with earlier findings on variable embrittlement sensitivity,⁽⁴⁾ the irradiation performance of the weld metal and plate can be related in part to their respective copper contents. However, the 0.2% copper content of the electroslag weld metal approximates that of the submerged arc weld metals examined to date and also found to

exhibit high radiation embrittlement sensitivity.⁽⁵⁾ Thus, it would appear that irradiation response is not influenced directly by the particular welding process used.

Studies of the notch ductility performance of irradiated electroslag weldments are continuing and will assess one weldment specially fabricated with low copper filler wire.

SPECIAL A533-B STEEL HEAT FOR VARIABLE RADIATION EMBRITTLEMENT STUDIES

At the request of the AEC, NRL has specified and initiated procurement of a special tonnage heat of A533-B steel with compositional control for limiting radiation embrittlement sensitivity. The major objective of this procurement is to determine if present mill capabilities and technology in concert with existing knowledge of variable radiation embrittlement sensitivity⁽⁴⁻⁶⁾ can reproduce or closely approach with tonnage heats that low level of radiation sensitivity developed

with several small laboratory heats of A302-B steel. The common feature of these laboratory heats was a close restriction on total residual element content. Two residual elements known to be particularly harmful in this case are copper and phosphorus (or phosphorus in combination with sulphur).

A 30 ton electric furnace heat of A533-B steel was produced on May 27, 1968 by Latrobe Steel Company in accordance with NRL composition specifications summarized in Table 9.4. The melt was vacuum stream degassed and split between two ingots with 0.13% copper (aim) added to one ingot in the form of pure shot as it was cast. This composition modification was specified in the interest of examining the tolerance of A533-B steel to some low level copper content (ingot 1) as opposed to only trace quantities of residual copper (ingot 2). Chemical analyses of hot top samples included in Table 9.4 indicate that composition specifications and tolerances were met.

TABLE 9.4. Chemical Composition of Special A533-B Steel Heat

Sample	Chemical Analysis, wt %															
	C	Mn	P	S	P+S	Si	Ni	Mo	Cr	Cu	V	As	Sb	Sn	Bi	As+Sb+Sn+Bi
Melt Aim*	0.20	1.30	0.007	0.007	0.012	0.24	0.55	0.55	LAP**	LAP**	LAP**	LAP**	LAP**	LAP**	LAP**	0.05
			max.	max.	max.				0.05	0.08		0.01	0.01	0.02	0.02	max.
									max.	max.		max.	max.	max.	max.	
Latrobe Steel																
Hot Top Analysis																
Ingot 1	0.18	1.38	0.006	0.006	0.012	0.20	0.56	0.53	0.06	0.12	-	<0.01	<0.01	<0.02	<0.02	<0.05
										(0.13% Aim)						
Ingot 2	0.18	1.37	0.006	0.006	0.012	0.20	0.56	0.53	0.06	0.03	-	<0.01	<0.01	<0.02	<0.02	<0.05

* As specified by NRL and prime contractor (Lukens Steel Corporation)

** LAP - low as possible

Lukens Steel Corporation (prime contractor) has scheduled to roll the 62 in. wide by 22 in. thick ingots during the month of July. The ingots will be reduced to 6 in. plate (3 1/2:1 reduction) using a 1:1 cross rolling ratio. Each plate will be split with individual sections heat treated to Class 1 and Class 2 properties, respectively. Plate qualification, inspection, and shipment to NRL is planned before September 1.

THERMAL STABILITY EVALUATION OF Ni-Cr-Mo WELD DEPOSITS

The effect of variable chemical composition on the radiation embrittlement sensitivity of commercial and experimental Ni-Cr-Mo weld deposits, investigated as part of a cooperative program between NRL and B&W was discussed in the preceding quarterly report.⁽³⁾ The results suggest that copper contents of the weld deposits had a very pronounced (detrimental) effect on their irradiation behavior - a conclusion which reinforced previously reported data concerning the embrittlement sensitivity of Type A302-B pressure vessel steel.⁽⁴⁾

The investigation of thermal stability of the two commercial and four experimental weldments used in the cooperative program has now been completed. The results indicate that 550 °F aging for a time equivalent to the in-reactor exposure (~1200 hr) has no measurable effect on the transition temperature characteristics of these weldments.

Specifically, data points obtained from Charpy-V specimens representing the thermally-aged weld metal condition fell well within the scatter band of the points from the as-welded and stress relieved deposits in all cases.

THERMAL STABILITY CHARACTERISTICS OF 12 Ni-5 Cr-3 Mo MARAGING STEEL PLATE EVALUATED AT 550, 650, and 740 °F USING CHARPY-V NOTCH SPECIMENS

Previous research on two heats of 12 Ni-5 Cr-3 Mo maraging steel (160 to 185 ksi yield strength), representing the lower and higher strength levels for this composition, revealed that transition temperature increases for irradiations at 550 to 740 °F became greater in magnitude with increasing temperatures.⁽⁶⁾ This is in contrast to prior studies on the more conventional low alloy structural steels wherein transition temperature increases become smaller with increasing irradiation temperatures.

To investigate one possible cause of this anomaly, transversely oriented Charpy-V specimens were conditioned both for 1900 and 3300 hr at 550, 650, and 740 °F matching the time-at-temperature histories for the irradiated specimens from the same plates. The results of evaluating the specimens are summarized in Table 9.5.

For the 185 ksi yield strength condition, the increase in embrittlement is seen in Table 9.5 to continue from 550 to 740 °F at a rather uniform monotonic rate as is witnessed by both the Charpy

TABLE 9.5. Charpy-V Notch and Hardness Properties of
12 Ni-5 Cr-3 Mo Maraging Steel Plate After Thermal Conditioning

NRL Code	Conditioning Temperature, °F	ΔTT at 30 ft-lb* Charpy-V Level, °F		Hardness Change,* Rc	
		1900 hr	3300 hr	1900 hr	3300 hr
J6	550	80	80	1	1
Aged 20 hr					
@ 900 °F	650	110	~130	1 1/2	2
184.4 ksi YS	740	140	~150	4	4
J8	550	65	65	2	3
Aged 2 hr					
@ 900 °F	650	185	~190	6 1/2	8
162.6 ksi YS	740	185	~190	7	8

* Initial values: J6, -175 °F and 42 Rc
J8, -195 °F and 38 Rc

and the hardness values. In the case of the lower strength condition, however, a plateau is reached at 650 °F. The initial aging treatment readily accounts for this difference in response. The lower strength material was held at the aging temperature (900 °F) for a period of 2 hr which was not sufficient to allow the steel to approach its thermally stable state as closely as did the higher strength steel which was aged at 900 °F for 20 hr. Thus, the low strength condition is the more sensitive of the two to temperatures below 900 °F. Further, in both cases, the preponderance of this "thermal embrittlement" occurs during the first 1900 hr, as might be expected.

The relationship of these control data to the irradiation experiments is graphically demonstrated in Figures 9.4 and 9.5 for the two yield

strength conditions. The thermal instability of the maraging steel is more than sufficient to account for the transition temperature increase resulting from high temperature irradiation which indicates some interaction between the two effects. As a result, the irradiation embrittlement for these maraging steels appears to be directly related to exposure temperature in the range 450 to 740 °F, while for carbon and low alloy steels it is inversely related. Additionally, it is apparent that the thermal stability of these steels is of major consequence for fluences up to 5×10^{19} n/cm² (E > 1 MeV).

On the basis of the available data, a simple relationship between neutron exposure conditions and loss of the shear energy absorption level is not readily apparent. However, the shelf energy after irradiation is great enough to avoid

FIGURE 9.4. Charpy-V Notch Ductility Behavior of 12 Ni-5 Cr-3 Mo Maraging Steel Plate (160 ksi Yield Strength) Before and After Conditionin at 550, 650, and 740 °F and After Irradiation at 550, 650, and 740 °F. Transverse (WR) Orientation Specimens Were Used.

low energy ductile tear problem in all but the most extreme environment of temperature and fluence.

Continuation of this study will involve conditioning of tensile specimens in the two strength conditions at 550 and 740 °F to measure more exactly the degree of aging involved.

LONG-TERM IRRADIATION OF PRESSURE VESSEL STEELS IN THE BIG ROCK POINT REACTOR

Neutron irradiation of metallurgical test specimens for long periods at elevated temperatures within the range of operating temperatures of power reactors is quite

FIGURE 9.5: Charpy-V Notch Ductility Behavior of 12 Ni-5 Cr-3 Mo Maraging Steel Plate (185 ksi Yield Strength) Before and After Conditioning at 550, 650 and 740 °F and After Irradiation at 550, 650, and 740 °F. Transverse (WR) Orientation Specimens Were Used.

difficult in test reactors because of the degradation of experimental assembly components and excessive neutron exposures. The data from such experiments are necessary, however, to allow the most meaningful interpretation of radiation effects data. Thus, it is quite fortuitous that an accelerated surveillance location of the Big Rock Point Reactor (BRPR) has been available for irradiation of steel specimens.

The location within the reactor is large enough to accommodate a number of irradiation capsules, and the ambient water temperature in the immediate region is about 575 °F. Therefore, it is only necessary to construct simple capsule (non-lead) assemblies which will permit neutron exposures over long periods of time at a constant temperature, in this case, 575 °F.

The first group of NRL capsules irradiated at 575 °F in the accelerated location of the BRPR has been received after an irradiation of 9720 hr. Duplicate specimens of the same steels contained in the capsules are presently undergoing thermal treatment at 575 °F at NRL for the same length of time. Thermal aging results will be reported in the future.

The notch ductility data for two of the eight steels irradiated in the BRPR are now available (Table 9.6) for A302-B plate in Figure 9.6 and in Figure 9.7 for A543 (3 1/2 Ni-Cr-Mo) plate. The reported temperature of 575 °F for these irradiations has been determined by observation of low melting point fusible alloy monitors contained within each capsule, coupled with the estimation of the temperature gradient occurring within the reactor at the accelerated irradiation location. The reported neutron fluences are based upon the assumption of a fission spectrum distribution of neutrons at the irradiation location and a 68 mb fission-spectrum-averaged

iron flux detector cross section. The calculated neutron spectrum for this location indicates that the neutron fluences for n/cm^2 of energies greater than 0.5 MeV would be smaller by the fraction 98.26/118.

In assessing experimental results for the two steels, it appears that the 575 °F irradiation temperature has had a more pronounced effect toward limiting the amount of radiation-induced embrittlement than an irradiation temperature of 550 °F. Specifically, a 550 °F test reactor neutron exposure of both A302-B and A543 steels revealed higher amounts of embrittlement, 190 and 150 °F, respectively.⁽⁷⁾ The full shear energy absorption values for the steels, however, indicated decreases very comparable to those exhibited by the two steels in the test reactor irradiation. These decreases are, nevertheless, relatively insignificant. The lower transition temperature increases for the 575 °F BRPR irradiation appear to be in proper proportion to the results of a 650 °F test reactor irradiation.⁽⁷⁾

TABLE 9.6. Notch Ductility Properties for Two Steels After Irradiation in the Big Rock Point Reactor Accelerated Surveillance Location at 575 °F

Steel	Fluence*	30 ft-lb Transition Temp., °F			Full Shear Energy Absorption	
		Unirradiated	Irradiated	ΔT	Unirradiated	$\Delta ft-lb$ (decrease)
A302-B	7.0×10^{19}	30	195	165	78	20
A543	5.9×10^{19}	-130	-25	105	74	20

* n/cm^2 ($E > 1$ MeV); σ 68 mb, ^{54}Mn , fission spectrum

FIGURE 9.6. Notch Ductility Characteristics of 6 in. Thick A302-B Plate Before and After Irradiation in an Accelerated Surveillance Location of the Big Rock Point Reactor

FIGURE 9.7. Notch Ductility Characteristics of 8 in. Thick A543 (3 1/2 Ni-Cr-Mo) Steel Plate Before and After Irradiation in an Accelerated Surveillance Location of the Big Rock Point Reactor

The most significant conclusions which can be drawn from these results are that both A302-B and A543 steels appear to be nearing a condition of radiation embrittlement saturation in terms of increasing transition temperature for irradiation conditions at the nominal operating temperatures for pressurized and boiling water reactors (550 to 575 °F) and that the upper shelf energy values for a neutron fluence approximately $6 \text{ to } 7 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) are not adversely affected.

Several of the irradiated specimens have been reserved for annealing heat treatments to determine recovery characteristics after the BRPR irradiation. Tensile specimens of these two steels are also being evaluated. The final assessment of these data will be made with reference to the results of matching long-term thermal treatment.

PLATE DIRECTIONALITY CHARACTERISTICS OF IRRADIATED LA CROSSE REACTOR PRESSURE VESSEL STEEL

The notch ductility characteristics of the longitudinal or strong orientation in plate sections of three heats of 4 in. thick A302-B steel from the La Crosse Boiling Water Reactor (LACBWR) have been reported.⁽⁶⁾ Because of the importance of the transverse direction properties in reactor pressure vessels, the pre- and postirradiation characteristics for this "weak" plate orientation in two LACBWR vessel heats have also been determined and are reported herein.

Because of physical limitations, it was not possible to irradiate longitudinal and transverse specimens to all the LACBWR heats in one assembly. Accordingly, transverse specimens of two heats were combined with several longitudinal specimens of these same heats to act as controls for reference with the first, all longitudinal specimen irradiation. This second assembly was irradiated at 550 °F in a core facility of the Union Carbide Research Reactor (UCRR), Tuxedo, New York, to a neutron fluence of $1.0 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$).

Notch ductility characteristics for the unirradiated condition of transverse specimens from heats NP-1057 and NP-1058 are shown in the lower halves of Figures 9.8 and 9.9; the unirradiated condition characteristics for the longitudinal specimens of these heats are presented in both upper and lower halves of the same figures. Both plates of heat NP-1057 displayed double transition behavior in longitudinal and transverse orientations, while both plates of NP-1058 displayed a smoothly rising fracture energy with increasing temperature. The high full shear energy absorption shelf levels for the longitudinal specimens are in marked contrast to the far lower values for the transverse specimens. The wide difference is not indicative of that expected as a result of the 1:1 cross rolling practice commonly used for heavy section pressure vessel steel. Drop-weight NDT temperatures determined for the two heats were both +20 °F

FIGURE 9.8. Notch Ductility Characteristics of 4 in. La-Crosse Reactor Vessel Steel Plate (heat NP-1057) from Both Longitudinal and Transverse Orientations Before and After 550 °F Irradiations to 1.0 and $2.1 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$)

which corresponded to Charpy-V energy level between 75 and 95 ft-lb for the longitudinal orientation and between 30 and 40 ft-lb for the transverse orientation. Because of these wide variations, 30 ft-lb was arbitrarily selected for transition temperature increase determinations.

The postirradiation notch ductility results for the transverse specimens of heat NP-1057 clearly showed the double transition behavior, but also showed a 30 °F transition temperature increase in contrast to the absence of meaningful transition temperature increases

FIGURE 9.9. Notch Ductility Characteristics of 4 in. La-Crosse Reactor Vessel Steel Plate (heat NP-1058) from Both Longitudinal and Transverse Orientations Before and After 550 °F Irradiations to 1.0 and $2.1 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$)

for both irradiations of longitudinal specimens from this heat. The transverse orientation full shear energy absorption shelf was less than half that of the longitudinal orientation and showed a small lowering as a result of the $1.0 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) irradiation. This drop was in contrast to the retention of full shear energy level ex-

hibited by the longitudinal specimens in the same irradiation, but in proportion to the energy level drop exhibited by the longitudinal specimens from the first, higher fluence irradiation.⁽⁶⁾

The postirradiation notch ductility results for the transverse specimens of heat NP-1058 indicate radiation response characteristics

similar to those exhibited by the longitudinal specimens of the same irradiation and also to that from the higher fluence irradiation.⁽⁶⁾ The transverse specimen full shear energy level decreased somewhat as a result of the 1.0×10^{19} n/cm² ($E > 1$ MeV) irradiation, again in contrast to the retention of pre-irradiation energy levels for the longitudinal specimens from the same irradiation. As for the case of heat NP-1057, the higher fluence longitudinal specimens exhibited a drop in full shear energy level in comparable proportion to that for the transverse specimens. The notch ductility data for both irradiations of LACBWR steel are summarized in Table 9.7.

The different transition temperature increases revealed by the two LACBWR steel heats are highlighted, however, by the far larger, 140 °F, transition temperature increase of a reference A302-B steel

plate irradiated under the same conditions. Furthermore, the difference in radiation sensitivity of the reference and LACBWR A302-B steel plates appear to be related to their respective copper contents. Previous observations have been made on the influence of copper and other elements regarding their effect upon radiation embrittlement sensitivity.⁽⁴⁾ From this research, one could expect less embrittlement from the 0.16% copper NP-1085 heat than from the 0.20% copper reference heat and even less embrittlement from the 0.11% copper NP-1057 heat. It is clear, however, that copper content alone is not responsible for the essentially nil sensitivity of the NP-1057 heat, but that other factors, as yet not fully understood, must be contributing.

The investigation has shown that production plates of reactor pressure vessel steel can be produced

TABLE 9.7. Summary of Notch Ductility Evaluations on LaCrosse Pressure Vessel Plates and Weldments

Plate Code	Thickness Location	Orientation in Plate	Neutron Fluence, n/cm ² * (E > 1 MeV)	Charpy-V 30 ft-lb Transition Temp., °F			Energy Absorption at Full Shear, ft-lb		
				Unirrad-iated	Irrad-iated	ΔT	Unirrad-iated	Irrad-iated	Δft-lb
Heat NP-1057									
LC2	1/4 - 3/4	Longitudinal	2.1 x 10 ¹⁹	-35	~25	<20	143	129	14
LC2	1/4 - 3/4	Transverse	1.0 x 10 ¹⁹	- 5	25	30	72	66	6
LC3	3/4	Longitudinal	1.0 x 10 ¹⁹	-45	-45	0	149	141	8
Heat NP-1058									
LC4	1/4 - 3/4	Longitudinal	2.1 x 10 ¹⁹	-25	40	65	150	120	30
LC4	1/4 - 3/4	Transverse	1.0 x 10 ¹⁹	25	85	60	76	65	11
LC5	3/4	Longitudinal	1.0 x 10 ¹⁹	-35	20	55	144	144	0

* σ^- mb, ⁵⁴Mn, fission spectrum

with transverse properties considerably poorer than longitudinal properties. Although the transverse full shear energy absorption levels from the two LACBWR A302-B steel plates were about half of the longitudinal orientation, the radiation-induced decreases were proportional. The relatively small transition temperature increases are essentially the same whether the 30 ft-lb or the actual NDT point is used for referencing the irradiation induced increase in both plate orientations for the two LACBWR heats. The longitudinal and transverse orientation properties determined for these two LACBWR steel heats suggest that the reactor can be subjected to lifetime neutron fluences in excess of 2.1×10^{19} n/cm² ($E > 1$ MeV) without undue concern for transition tempera-

ture increase problems or for potential fracture by low energy absorption tearing through neutron induced lowering of the energy shelf.

PRELIMINARY STUDY OF THE IRRADIATION RESPONSE OF A537 GRADE B STEEL AT LESS THAN 250 °F

The initial irradiation experiment performed on Charpy-V specimens from a 3/4 in. thick plate of A537-B has been completed. The results show that the material transition temperature at the 30 ft-lb level is similar in irradiation response to A302-B for exposures of 5.1×10^{18} n/cm² ($E > 1$ MeV) at less than 250 °F. Data for the longitudinally oriented specimens are presented in Figure 9.10. Additional test data not plotted on the graph show that the

FIGURE 9.10. Charpy-V Notch Ductility Behavior of A537 Grade B Steel Plate Before and After Irradiation at Less Than 250 °F

transverse Charpy full shear energy absorption level in the preirradiation state is 45 ft-lb. Metallographic examination revealed a high cross-rolling ratio which accounts for the unusually low Charpy energy shelf relative to that for the longitudinally oriented specimens (128 ft-lb).

The transverse tensile properties will be carefully monitored in view of the foregoing directionality variations. Also, specimens of this plate and of production heats of the Fort St. Vrain Reactor liner will be included in future irradiation experiments. Special attention again will be given to the transverse specimen energy shelf.

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RADIATION EFFECTS ON FAST REACTOR CLADDING AND STRUCTURAL MATERIALS

Nuclear Materials and Propulsion Operation
General Electric Company

J. Moteff

The objective of this program is to determine the effect of radiation on the time-, temperature-, and stress-dependent properties of selected heat-resistant alloys and refractory metals, 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 this class of metals and alloys is continuing. Direct observations of defect configurations by means of the transmission electron microscope, as well as analysis of the neutron-induced defects and their migration behavior, are being performed and correlated with the measured changes in the mechanical properties.

STATUS OF IRRADIATIONS

F. D. Kingsbury, J. P. Smith, and
W. S. Chenault

ORR Facility

Three capsules were irradiated in the Hydraulic Tube Facility No. 14 of the Oak Ridge Research Reactor (ORR) since the beginning of the calendar year. The capsules were designated as ORM-60, -61, and -62 and were back-filled with high-purity helium. Capsule ORM-60 contained

vanadium single crystal foils of two different crystallographic orientations for transmission electron microscopy (TEM) studies and Hastelloy R-235 resistivity and foil specimens with two different boron isotope concentrations (50 ppm ^{10}B or 50 ppm ^{11}B). ORM-61 contained Mo single crystal and polycrystalline hot-hardness specimens. Ultra-high purity triple-zone refined polycrystalline vanadium and vacuum arc cast Mo hot hardness specimens were contained in ORM-62. Each capsule was irradiated separately for time periods of 408 to 428 hr to an estimated fluence of $5 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$).

Three ORR irradiation capsules containing AISI 304 SS low-cycle plastic fatigue specimens are in various stages of design or fabrication. Each capsule will contain six specimens of the hour-glass design. Capsule ORM-63 is fabricated and will be irradiated at reactor ambient temperature to approximately $3 \times 10^{18} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) beginning in mid-May. Following this, capsule ORM-64 will be irradiated to approximately $6 \times 10^{17} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$). Both of these capsule designs are identical to that used for ORM-55 reported previously.⁽¹⁾ Capsule ORHF-1 is being designed for irradiating six fatigue specimens at 650 °C in NaK.

EBR-II Facility

Subassemblies X034 (Row 2) and X035 (Row 7) which contained ORNL, BMI, and GE-NSP capsules were inserted in the EBR-II in early April at the beginning of cycle 28. The specimens in the Row 2 position have accumulated an irradiation (May 13, 1968) fluence in excess of 1.7×10^{21} n/cm² ($E > 1$ MeV).

HEAT-RESISTANT ALLOY PROGRAM

Incoloy 800 has been creep-rupture tested at 705 °C following irradiations in a fast (EBR-II) or a thermal (GETR) spectrum reactor. These specimens were tested under similar conditions to evaluate the effects of different neutron spectra on radiation-induced changes of mechanical properties. Metallography by the use of the optical and transmission electron microscope has been initiated. Preliminary investigations of the influence of low fluence ($\sim 3 \times 10^{18}$ n/cm², $E > 1$ MeV) irradiations on the plastic fatigue properties of 316 SS have been implemented.

Creep-Rupture Testing -

J. P. Smith, and R. A. Joseph

Incoloy 800* Creep-rupture testing of Incoloy 800 continues with the primary effort during this reporting period to determine the effects of neutron spectra on the 705 °C creep-rupture properties. Considerably more time was utilized in obtaining the data for GETR unirradiated control specimens because

this material is 4 to 5 times stronger than the EBR-II controls, although both types of specimens were cut from the same heat of material and received the same final heat treatment after fabrication (1030 °C for 15 min). The only known differences are 1) specimen thickness (0.012 cm for EBR-II and 0.024 cm for GETR specimens) and 2) reactor temperature simulation treatment. The EBR-II control specimens were given a reactor temperature simulation treatment of 214 hr at 540 °C, while the GETR specimens were simulation treated for either 620 or 5200 hr at 540 °C. Also, one EBR-II type was aged 500 hr at 705 °C. The reactor simulation treatments are not felt to be the primary cause of the difference between the two types of specimens because:

- EBR-II controls tested at 705 °C (10.5 kg/mm^2) had rupture lives and minimum creep rates within 10% of each other regardless of whether they were as-fabricated, aged at 540 °C for 214 hr or aged at 705 °C for 500 hr.
- GETR controls tested at 705 °C (10.5 kg/mm^2) had rupture lives and minimum creep rates within 10% of each other regardless of whether or not they were aged at 540 °C for 620 or for 5200 hr.

Both types of specimens exhibit essentially similar ductility at 705 °C with the GETR type specimens possibly being more ductile.

Based on the above considerations, it appears that the GETR type specimens are stronger probably because

of the differences in specimen fabrication necessitated by going from the 0.024 to 0.012 cm (0.062 to 0.030 in.) thickness, and the resulting grain size difference.

The effects of the GETR irradiation are quite similar to that produced by EBR-II irradiation in that rupture life is slightly reduced as is minimum creep rate. Ductility is reduced by about a factor of 4 for all irradiations. These preliminary results indicate that spectrum plays a minor role in determining radiation damage and are in good agreement with the results discussed previously for Hastelloy X which showed that either EBR-II or ETR irradiation produced essentially the same decrease in 650 °C rupture properties.

Examination of 705 °C creep-rupture tested Incoloy 800 by optical and transmission electron microscopy has been initiated. Transmission electron microscopy revealed no apparent difference between the unirradiated or EBR-II irradiated specimens. Specifically, no bubbles, voids or unusual dislocation patterns were observed.

Preliminary optical examination of these same specimens, particularly in the fracture area, has been somewhat more enlightening. Unirradiated specimens tested at 10.5 kg/mm² exhibit primarily transgranular fracture, whereas specimens tested at 5.4 kg/mm² exhibit intergranular fracture. This is a particular significant observation since irradiation has only minor effects on rupture life and creep rate at the 10.5 kg/mm² stress level, but changes rupture life and creep rate by a factor of 2 to 3 at the 5.4 kg/mm² stress level. Pre-

liminary examination of the irradiated specimens indicates that the same trend holds true except that at the higher stress level the fracture appears to be more evenly mixed trans- and inter-granular rather than predominantly transgranular. Absolute evaluation is difficult because the grain size is fine and it has been necessary to utilize replica and electron microscope techniques.

Hastelloy X

The testing at 540 °C of Hastelloy X control and EBR-II irradiated material is also in progress. The present material is much stronger than a previously tested⁽²⁾ different heat of Hastelloy X and, therefore, the control specimens are resulting in a longer time to rupture than expected and are still on test; however, two irradiated specimens have been ruptured. The ratio of rupture lives of control to irradiated will be greater than 5 and minimum creep rate is increased slightly at 39.4 kg/mm² from a value of 1.2×10^{-8} sec⁻¹ for the unirradiated specimen to $\sim 1.8 \times 10^{-8}$ sec⁻¹ after irradiation. The major effect is that the amount of initial strain on applying the load decreases with increasing neutron exposure. This is presumably due to substructure hardening by point defects produced during irradiation.

Low Cycle Plastic Fatigue

J. P. Smith and J. T. Berling

Preliminary studies have begun on the effects of neutron irradiation

on the low cycle plastic fatigue properties of AISI 316 SS. Specimens are of the hour-glass configuration identical to those being used on Job 81516. The test technique has been described in detail previously.⁽³⁾ Briefly, it is a push-pull type test based on diametral strain control utilizing a closed-loop servo control system. By electronically correcting for Poisson's ratio, a true constant axial strain range can be maintained at constant strain rate. Initial tests were performed on specimens irradiated at reactor ambient temperature in the ORR to 3×10^{18} n/cm² (E > 1 MeV) and 2×10^{19} n/cm² (thermal). This relatively low dose was picked for initial tests to determine potential problem areas that might be encountered in handling irradiated specimens of this configuration, since several sharp corners and potential stress concentration points are present in the grip section and it was anticipated that these could cause problems in testing the less ductile irradiated specimens. Also, the above fluence should be representative of the expected fluence range in the pressure vessel, flanges and other hardware in the FFTF and FBR designs. The initial test temperature of 650 °C was selected because it is known that irradiation induced embrittlement occurs in most austenitic steels at this temperature and that this temperature is applicable for first generation FBR systems.

The results of the first series of tests are summarized in Table 10.1. All specimens, although cut from the same rod of 316 SS, showed results

on the unirradiated specimens which agree quite well with other rods from this same heat.

The data are primarily of interest to designers and illustrate that the effect of irradiation is to reduce fatigue life at a given strain rate by a factor of about 3.5 and is independent of the strain ranges considered.

Analysis of the data in Table 10.1 leads to the following conclusions:

- Stress range is unaffected by irradiation.
- Based on a comparison of N_0 in the control to that of the irradiated, a macrocrack is formed much earlier in the irradiated specimen.
- Based on a comparison of N_0/N_f irradiated, the crack propagation rate is faster for the irradiated specimen.
- Using N_5 as the criterion for fatigue life, the effect of irradiation is to reduce the fatigue resistance by a factor of about 3.6.

Analysis of the hysteresis loop (and the tensile data discussed below) indicates that irradiation does not affect either Poisson's Ratio or the elastic modulus.

Only macroexamination of the fractures have been performed to date. These results show no detectable difference in the fracture mode of the irradiated and unirradiated specimens. All appear to be predominantly intergranular with no fatigue striations visible at relatively low magnifications (50X) under stereomicroscope observations.

TABLE 10.1. Low Cycle Plastic Fatigue Properties of Irradiated and Unirradiated AISI 316 SS

Specimen Number	Test Temp., °C	Strain Range, %			Strain Rate, sec ⁻¹		Loading Frequency, cycles/sec	Stress Range, Δσ at N _f /2, kg/mm ²	Fatigue Life					
		Diametral, Δε _d	Total Axial, Δε _t	Plastic Axial, Δε _p	Diametral, ε̇ _d	Axial, ε̇ _t			N ₀		N ₅		N _f	
									cycles	hr	cycles	hr	cycles	hr
26-1 (a)	650	0.99	2.11	1.73	1.97 × 10 ⁻⁵	4.21 × 10 ⁻⁵	0.001	58.07	71	19.7	139	38.61	153	42.5
26-4 (a)	650	0.494	1.104	0.771	1.98 × 10 ⁻⁵	4.36 × 10 ⁻⁵	0.002	51.398	432	60.0	490	68.05	695	96.53
26-8 (b)	650	1.00	2.13	1.75	2.00 × 10 ⁻⁵	4.23 × 10 ⁻⁵	0.001	57.69	27	7.5	37	10.27	44	12.22
26-9 (b)	650	0.49	1.10	0.76	1.97 × 10 ⁻⁵	4.40 × 10 ⁻⁵	0.002	53.49	138	19.17	168	23.33	182	25.28

(a) Unirradiated

(b) Irradiated in ORR at $\sim 70^\circ\text{C}$

3.1×10^{18} n/cm² ($E \geq 1$ MeV)

2.4×10^{19} n/cm² (thermal)

It has been proposed that an important parameter in determining fatigue life is ductility. However, the ductility measured should be generated by a test performed at a true constant strain rate all the way to failure. This type of data is quite readily obtained by the diametral strain control and electronic closed loop system described above. To date, constant strain rate tensile tests were performed at the identical test conditions ($4 \times 10^{-5} \text{sec}^{-1}$ strain rate at 650°C) used for the fatigue tests described above. An important feature of the stress-strain curve is that the irradiated and unirradiated stresses follow identical paths, initially. The major difference is that the irradiated specimen breaks with less than 1/2 the ductility of the unirradiated specimen. This single pair of tensile tests tends to support the hypothesis that fatigue life is ductility limited; however, considerably more testing is needed to verify these preliminary conclusions. These results will be analyzed in light of the various pro-

posed parameters utilizing short time tensile strength and ductility to predict fatigue limits as more data becomes available.

REFRACTORY METAL AND ALLOYS PROGRAM

Data are presented on the effects of irradiation temperature on the 1000°C creep-rupture properties of Mo rod specimens (two carbon levels). Hot-hardness studies have been continued on irradiated or unirradiated Mo, V, and W as an aid to defining the recovery mechanisms. Room-temperature hardness results following elevated temperature anneals on irradiated Ta and Ta-10 W are given. Initial transmission electron microscopy studies on irradiated and unirradiated vanadium are reported.

Creep-Rupture -

F. Kingsbury and R. Treinen

Mo Rod

The initial study of the effect of irradiation temperature on the creep-rupture properties of Mo at 1000°C has been completed. The

effect of irradiation temperature was studied on recrystallized polycrystalline specimens irradiated at three different temperatures [70 (reactor ambient), 700, and 1000 °C] to a fast fluence of approximately 1.3×10^{20} n/cm² ($E > 1$ MeV). Tests were conducted on Mo rod material with two carbon levels; i.e., 220 ppm and 38 ppm. The majority of the tests were conducted at a fixed stress level of 7.00 kg/mm². In these tests, none of the specimens were tested to fracture due to limitation in lever arm travel. All tests were terminated in the latter portion of third stage creep; the reported rupture lives are believed to be within 1% of the true value. All specimens exhibited a ductility of 30% or greater.

The present study, at a test temperature of 1000 °C ($0.44 T_m$), was conducted to complement the previously reported⁽⁴⁾ series of tests at 750 °C. Mastel and Brimhall⁽⁵⁾ showed in their transmission electron microscopy studies on recrystallized Mo, irradiated at reactor ambient temperature (1×10^{19} n/cm², $E > 1$ MeV), that the spot and loop defect densities decreased rapidly (factors of 50 to 100) on annealing from 750 to 900 °C. Hence, mechanical testing of specimens above 750 °C ($\sim 0.35 T_m$) should show significant differences compared to the previous 750 °C tests.

A comparison of the creep-rupture properties of Mo at 1000 °C for specimens irradiated at the three different irradiation temperatures and a control specimen show that the

results for the case of the two carbon levels are in general, quite similar to those observed in the 750 °C tests. The specimens irradiated at ambient reactor temperature (~ 70 °C) tend to exhibit creep characteristics of unirradiated material. All specimens irradiated at 700 and 1000 °C exhibited "delayed creep," whereas the ambient irradiated specimens show no evidence of delayed creep for the low-carbon Mo and only a very brief (~ 0.4 hr) period of delayed creep for the high-carbon Mo. Specimens irradiated at 700 °C increased in rupture life by factors of 1.3 and 1.5 compared to the unirradiated specimens. Irradiation at 1000 °C increased the rupture lives by factors of 2.4 and 2.6. The high-carbon (220 ppm) specimens exhibited approximately twice the rupture life of the low-carbon (38 ppm) specimens for a given similar condition. The results suggest that, for the carbon contents considered in these experiments, the major influences of carbon are 1) to increase the unirradiated elevated temperature plastic flow resistance (strengthening) and 2) to have very little effect on the stabilization of irradiation-induced defects. However, an effect of carbon content on the behavior due to irradiation-induced defects could be prominent for specimens that contain a factor of 10 less carbon (i.e., ~ 2 to 4 ppm C).

The creep characteristics of the various specimens have been examined by plotting instantaneous creep rates versus strain. The high carbon specimen irradiated at 70 °C exhibited

behavior very similar to that observed in the 750 °C tests;^(4, p. 679) i.e., a rapidly decreasing creep rate initially, followed by a rapidly increasing rate reaching a maximum value. After reaching the maximum value, the creep rate decreases to the usual minimum second stage creep rate. The initial decreasing creep rate and inflection point ("delayed creep") was not observed in the low carbon ambient irradiated specimen.

The creep rate versus strain behavior of specimens irradiated at elevated temperatures show very similar characteristics for both carbon levels and irradiation temperatures. For a given irradiation temperature and strain, the low-carbon specimens exhibited a higher creep-rate compared to the higher carbon specimen. The unirradiated material also shows this behavior.

In the present tests at 1000 °C, the creep-rate versus strain behavior of the 1000 °C irradiated high-carbon Mo specimen differs from the 750 °C test results.^(4, p. 681) In the 750 °C tests, it was suggested that a more stable defect was possibly formed at an irradiation temperature between 700 and 1000 °C. This was evidenced by the lack of a marked inflection at the minimum creep rate and the lower creep rate. The appearance of the inflection point at the 1000 °C test temperature indicates that this defect interacts differently with the dislocations at the two temperatures. The minimum in the creep rate occurs at about the same strain value (0.6 to 0.7) irrespective of carbon level or irradiation

temperature. It is apparent that a change in controlling mechanism is occurring at the inflection point. Plastic strain rate and dislocation movement by the Taylor equation:

$$\dot{\epsilon} \approx \frac{1}{2} \rho b \bar{v} \quad (\text{Simple tension})$$

where ρ is the dislocation density

b is the Burger's vector

$\bar{v}$ is the average dislocation velocity

shows that the creep rate is affected; by the rate of dislocation movement (velocity) and the multiplication of dislocations (density). Determination of the nature of these interactions must await the results of planned transmission electron microscopy studies and aging studies. Both grades of unirradiated Mo are to be given a thermal aging history similar to the 1000 °C irradiated specimens. In addition, creep-rupture tests will be conducted on these control specimens.

Resistivity Studies -

L. K. Keys and J. Moteff

Molybdenum

The previously reported incomplete high temperature annealing resistivity study^(1, pp. 595-98) of molybdenum after fast neutron ($E > 1$ MeV) irradiation (1.5×10^{21} nvt) was completed. The result of this study shows that the recovery consists mainly of a single large recovery peak centered in the vacancy recovery region.^(1, p. 715) A result which stands out, and which has not

been observed at the lower neutron fluences, is the presence of a residual resistivity after the high temperature annealing treatment. Since all the defects should be annealed out by the highest annealing temperature, 1500 °C, it appears, similar to tungsten,⁽⁶⁾ that the influence of the transmutation products are also observed in molybdenum. This transmutation is apparently significant at the higher thermal neutron fluences and leads to a detectable concentration of transmutants after a thermal fluence of 2.7×10^{21} nvt. The probable transmutation products⁽⁷⁾ are ^{99}Tc , ^{101}Tc , ^{100}Ru and ^{101}Ru . The transmutation species are in much smaller concentration in molybdenum than in tungsten due to the lower thermal neutron absorption cross section. The induced residual resistivity for the irradiated molybdenum specimen after the high temperature annealing treatment is only about 0.1 times the preirradiation value, whereas that for tungsten for a comparable irradiation is about 4 times the preirradiation value.^(6, p. 87) The products of this transmutation reaction may very well lead, in part, to the high temperature hardening observed in molybdenum after neutron irradiation.

Transmission Electron Microscopy of Vanadium -

R. C. Rau, F. Secco d'Aragona, and R. L. Ladd

Microstructural studies, using transmission electron microscopy, have been begun on vanadium metal.

These studies include 1) the characterization of the microstructure of unirradiated pure vanadium source materials, and 2) the determination of the effects of neutron irradiation and subsequent annealing on the microstructure.

Unirradiated Vanadium

Several vanadium source materials are being evaluated for possible usage in radiation damage experiments. However, to date only one material has been extensively surveyed, a high purity {100} oriented single crystal rod obtained from Materials Research Corporation.

Transmission electron microscopy has been carried out on this specimen both in the as-received condition and after various vacuum annealing treatments. In the as-received condition, extensive randomly oriented boundaries were seen which showed fringe contrast effects. These extended fringe defects, which are not grain boundaries, twin boundaries, or stacking faults, have been interpreted as domain boundaries. These domain boundaries, as well as dislocations in their vicinity, often moved under the influence of electron beam heating.

These domains are believed to be produced by a tetragonal distortion of the body centered cubic vanadium lattice due to the preferential occupation of tetrahedral or octahedral interstices by impurity atoms. The boundaries separate regions where different sets of interstices are filled.⁽⁸⁾ Such domain boundaries

have previously been observed in niobium⁽⁹⁾ and tantalum,^(10,11) and have been attributed to the ordering of interstitial oxygen or carbon. In the present case, the domains are believed to represent the ordering of nitrogen, carbon, and/or oxygen, since chemical analysis indicated the presence of all three of these elements.

In addition to the domain boundaries, two other types of defects were also seen in the as-received single crystal. These defects were less extensive and consisted of dark precipitates surrounded by many dislocations, and small planar fringed regions which had the appearance of stacking faults.

Vacuum annealing eliminated the domain boundaries, but enhanced the appearance of precipitates and the fault-like defects. After 1 hr annealing at 330 °C, both the precipitates and fringed areas were quite pronounced. Annealing at 650 °C for 1 hr removed the small background precipitates and initiated the break-up of the larger particles and fringed defects. Finally, annealing at 1000 °C led to a general deterioration and redistribution of the defects throughout the matrix. Microscopy studies also show examples of punched dislocations emanating from precipitate particles. These dislocations arise due to stresses set up by differences in thermal expansion between the particles and the matrix.

The interpretation of these rather complex effects in nominally high purity vanadium is still somewhat tentative, but like the domain boundaries is believed related to the

presence of nitrogen, carbon and/or oxygen impurities. Although the microstructures appeared to contain relatively large amounts of precipitates and defects, only rarely was evidence of a second phase found in the electron diffraction patterns, even when the diffraction selector aperture was positioned on seemingly massive particles. This indicates that the precipitates must be exceedingly thin, and would not require a great deal of impurity atoms to form them. Such thin precipitates are imaged chiefly by interference effects [i.e., displacement fringes⁽¹²⁾] or by diffraction contrast effects from surrounding dislocations.

The various precipitates seen are believed to be vanadium carbides and/or nitrides, although oxides cannot be ruled out. In the few cases where distinct second phase electron diffraction patterns were obtained, they were identified as most likely being V_2C . Indexing and measurement of this pattern gave hexagonal unit cell dimensions of $a = 2.91 \text{ \AA}$ and $c = 4.60 \text{ \AA}$, in good agreement with the values of $a = 2.886 \text{ \AA}$ and $c = 4.573 \text{ \AA}$ reported for V_2C , but in poorer agreement with the values of $a = 2.837 \text{ \AA}$ and $c = 4.541 \text{ \AA}$ reported for V_3N .

Observations of similar precipitates and fringed fault-like defects have been reported in chromium,⁽¹³⁾ where the precipitate is believed to be Cr_2N , and in various vanadium-^(14,15) and niobium-^(16,17) containing stainless steels. In these steels, precipitation of V_4C_3 or NbC initially occurs in very thin layers

at dislocations and stacking faults, and upon aging the faults gradually lose fringe contrast and are replaced by small particles outlining the former faulted areas.

Irradiated Vanadium

The effects of neutron irradiation on the microstructure of vanadium were studied using the {100} single crystal rod described above. Slices 0.5 mm thick were irradiated at reactor ambient temperature ($\sim 70^\circ\text{C}$) to a fast ($E > \text{MeV}$) neutron fluence of $5 \times 10^{19} \text{ n/cm}^2$. Transmission electron microscope observations were made of the as-irradiated material, and after 1 hr vacuum anneals at various temperatures ranging from 330 to 1175 $^\circ\text{C}$ (0.28 to $0.67 T_m$). These examinations showed the presence of radiation induced defect clusters and the effects of postirradiation annealing on those clusters. Quantitative determinations of the defect cluster densities were made from high magnification micrographs, assuming foil thicknesses of 2000 $\text{\AA}$.

In the as-irradiated condition, a very high density of small clusters was present. These clusters appeared as black dots averaging approximately 25 to 50 $\text{\AA}$ in diameter, and were estimated to be present in quantities approaching $10^{17}/\text{cm}^3$. Postirradiation annealing caused the clusters to increase in average size and decrease in number, until after the highest temperature anneal (1175 $^\circ\text{C}$) the cluster density was near $10^{14}/\text{cm}^3$ and the average cluster size was

approximately 500 $\text{\AA}$. After annealing at temperatures of 510 $^\circ\text{C}$ or above, many of the clusters were seen to be resolvable as dislocation loops. Tilting experiments in the electron microscope⁽¹⁸⁾ indicated that these loops were probably interstitial in nature.

The results indicate that the radiation-induced clusters in vanadium were much more persistent at elevated temperatures, based on a homologous scale, than expected from comparisons with the annealing behavior of other body centered cubic metals.^(19,20) This unexpected stability is ascribed to the presence of impurities in the vanadium, as discussed in the preceding section, and the interaction of those impurities with the radiation induced defects. Close inspection of dislocation loops in the higher temperature annealed specimens showed that they were decorated with small precipitates, believed to be vanadium carbides or nitrides. These precipitates probably segregate to the loops and stabilize them during high temperature annealing.

A further radiation phenomenon thought to be related to impurities in the vanadium was the occurrence of circular halos of defect clusters surrounding certain discrete particles in the matrix. Although these halos were rather rare in occurrence, they appeared only in the irradiated specimens, and were consistent in size and shape. The halos were duplex in nature, consisting of a very dense inner halo of approximately 1.3 μ radius, surrounded by a less dense

outer halo of about 2.6μ radius. The halos were centered around small square-shaped particles of about $3000 \text{ \AA}$ edge length. After annealing at 1175°C , both the halos and the central particle are less distinct. In this case, the outer halo appears to have been replaced by a circle of particles, while the inner halo seems to have been replaced by tangles of dislocations. In comparison with the surrounding matrix, the halo area in general seems to be depleted of small defect clusters and/or precipitates.

The observed halos are believed to be planar segments of spherical damage shells surrounding boron-containing precipitates, and arise from the $^{10}\text{B}(n,\alpha)^7\text{Li}$ reaction. The inner and the outer halos, thus, represent the recoil ranges of ejected lithium ions and alpha particles, respectively. Similar damage halos have recently been found in neutron irradiated iron- and nickel-base alloys containing boron.⁽²¹⁻²³⁾ Although the ranges of the recoiling particles have not yet been calculated for a vanadium matrix, it is expected that they should not be too different from the ranges in iron or nickel. The electron micrographs seem to indicate that this is the case.

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RADIATION METALLURGY

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SOLID STATE DIVISIONINTRODUCTION

M. S. Wechsler

The Radiation Metallurgy Section has been engaged in a study of radiation damage and radiation hardening and embrittlement in the body-centered-cubic metals. The emphasis on this class of metals is motivated largely by the practical problem of the radiation embrittlement of pressure vessel steels. This type of embrittlement is experienced by the pressure vessels of many operating reactors, which increases their susceptibility to catastrophic brittle fracture. Such brittle fracture incidents could have serious consequences that bear on the public safety.

Our research into the cause of the radiation embrittlement of pressure vessel steels indicates that the embrittlement is closely connected with radiation hardening. The increase in the yield stress of the steels upon irradiation permits stresses to occur which exceed the inherent cleavage fracture stress, thus permitting the initiation and propagation of a running cleavage crack at higher temperatures than would be possible in the absence of irradiation. The precise origin of

the radiation hardening is not known as yet, but it is almost certainly due largely to the radiation-produced defect clusters, which serve as barriers to dislocation motion on which plastic deformation depends.

To obtain a better understanding of how and why radiation produces an increase in the yield and flow stresses, it is helpful to experiment with metals less complicated than steels. Pressure-vessel steels contain a variety of impurities and eutectoid decomposition products whose individual effects tend to be masked. For this reason, it is helpful to work with other materials capable of better characterization. Therefore, the effort on steel has been supplemented in the past by experiments on iron and on niobium. Iron and niobium have the same basic crystal structure and both exhibit a tendency toward the same type of low temperature brittleness as steel. Niobium is a particularly useful material for study because it can be prepared in rather high purities and in single crystal form.

The study of niobium has developed around two major themes: 1) the influence of impurities, especially interstitial ones, and 2) radiation hardening and its relation to the defect clusters seen in the electron

microscope. This report starts with a description of the application of autoradiography to the determination of impurity distributions in zone-refined niobium. Then, some resistivity and internal friction measurements are described in which the trapping and de-trapping of oxygen and carbon impurities at radiation-produced traps is detected. The trapping of these impurities at radiation-produced defects has an effect on mechanical properties, as is seen in the radiation-anneal hardening that takes place when irradiated niobium is annealed between 150 and 400 °C. Some observations of this type are also specified below.

The radiation-produced defect clusters with which the radiation hardening is associated can be observed as black spots or loops by transmission electron microscopy in niobium and many other metals. On the other hand, the defect clusters are not detected in iron and steel by this technique, except after very high exposures or upon postirradiation annealing. Since the defect clusters are visible in niobium, the density and size distribution of the clusters were correlated with the magnitude of the radiation hardening. Furthermore, dislocation channeling was studied, a phenomenon in which the dislocations set in motion by plastic deformation cause the defect clusters to disappear. This report gives the results of a fairly detailed study of the crystal geometry of these dislocation channels and the

extent to which strain is concentrated within the channels. The fact that strain is so concentrated is consistent with the coarse slip bands observed on the surface of irradiated-and-deformed metals. This is what causes the rate of work hardening and the ductility of nominally ductile metals to decrease upon irradiation.

Unfortunately, budgetary limitations in fiscal year 1969 require that the work on niobium be discontinued. This report and the immediately previous ones indicate the present status of the research. A separate report will be issued later in the attempt to review the entire program on niobium and to indicate the directions that this line of investigation should take when conditions permit.

INHOMOGENEOUS DISTRIBUTION OF Ta AND W IN ZONE-REFINED NIOBIUM

R. E. Reed

Introduction

The electron beam floating zone melting (EBFZM) technique is widely used for purification and single crystal growth of the refractory metals. In the case of niobium, previous work at ORNL^(1,2) has shown that the major purification mechanism is generally volatilization. The metallic impurities Ta and W in niobium were not appreciably affected by the EBFZM method because of their low vapor pressure and the high zoning speeds which were above those needed for effective zone refining.⁽³⁾

Barthel⁽⁴⁾ has shown that for W impurities in molybdenum, the EBFZM technique results in a "banded" structure. In this, the W was distributed in alternate "impurity rich" and "impurity poor" layers which were parallel to the liquid-solid moving interface. This was attributed to turbulent convection currents in the liquid due to thermal gradients which resulted in periodic remelting of the moving liquid-solid interface. In a later paper, Barthel⁽⁵⁾ has shown that this fluctuation in the growth rate will change the effective distribution coefficient towards one. In addition, the dependence of the distribution coefficient on the zoning speed as given by Burton et al.⁽⁶⁾ was eliminated by these fluctuations. Therefore, at any zoning speed the effectiveness of zone refining of impurities with distribution coefficients greater than one will be reduced by fluctuations in the zone speed.

This report describes some preliminary work using microradiography to detect this inhomogeneous distribution of W and Ta impurities in niobium single crystals.

Experimental Procedure

The starting material was 1/2 in. diameter niobium rod swaged from a 3 in. diameter electron beam melted ingot supplied by Wah Chang Corporation, Albany, Oregon. Table 11.1 lists the chemical analysis for this material. A 1/2 in. diameter niobium rod was given one EBFZM pass

TABLE 11.1. Chemical Analysis for Wah Chang Heat No. 50541

Element	Weight, ppm
C	20
O	130
N	46
H	3
Ta	586
W	338

at 10 cm/hr zoning speed and 1×10^{-6} torr vacuum. The electron beam power was shut off quickly at the end of the pass. A 3 cm length which included the finish end was cut from the as-zoned rod. This specimen was then sectioned longitudinally along the rod axis by spark erosion machining. One half was ground flat on 600 grit abrasive paper, while the other half was planed flat by spark machining. Both were chemically polished in a nitric-hydrofluoric acid mixture.

These two specimens plus four sets of "standards" consisting of niobium metal with various Ta and W impurity levels were irradiated in the center hole of the D₂O tank of the Bulk Shielding Reactor for 24 hr. The thermal neutron flux in this position is 1.2×10^{12} n/cm²/sec. The samples were allowed to radioactively decay for 24 hr.

Autoradiographs were taken by placing the samples and one set of the standards on 2 in. x 2 in. Ilford nuclear emulsion plates with 100 μ thick L-4 type emulsion. Figure 11.1 is a print taken from a autoradiograph plate showing a typical arrangement of specimens and standards.

FIGURE 11.1. Print from Autoradiographic Plate of Electron-Beam Zone-Refined Niobium Containing Ta and W Impurities (2X)

The light and dark areas on this print are reversed from those on the autoradiograph.

The percent light transmission through the exposed plate for the standard and zoned specimens was measured on a Jarrel-Ash Recording Microphotometer Model 23-100. A calibration curve was made from the measurements on the standard specimens and used to find the variation of Ta and W content in the zoned specimen.

Results and Discussions

The autoradiographs were taken using β^- emission from neutron activated Ta and W impurities in the niobium. Table 11.2 lists the nu-

TABLE 11.2. Nuclear Data for Ta and W

Property	Ta	W
Nuclear Reaction	$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	$^{186}\text{W}(n,\gamma)^{187}\text{W}$
Abundance	^{181}Ta 99.98%	^{186}W 28.4%
Thermal Neutron Cross Section (barns)	19	34
Radionuclide	^{182}Ta	^{187}W
Type Decay	β^-	β^-
β^- energy (MeV)	99% 0.52	15% 1.3; 85% 0.63
Radionuclide Half Life	115 days	24.1 hr

clear properties necessary to calculate the relative activities of the β^- emissions. The activity due to a neutron-activated impurity element can be calculated as follows:

$$A = Nf\sigma SD$$

where

A = Activity from the impurity in dis/sec

N = Number of atoms of the impurity element that are present

f = Flux of thermal neutrons in $\text{n/cm}^2/\text{sec}$

σ = Thermal neutron cross section

S = Saturation factor = $(1 - e^{-\lambda t_i})$

where $\lambda = \frac{0.693}{T_{1/2}}$, t_i = irradiation time,

and $T_{1/2}$ = half life of radionuclide

D = Decay factor = $e^{-\lambda t_d}$ where

$\lambda = \frac{0.693}{T_{1/2}}$, t_d = decay time

measured from the finish of the neutron irradiation, and

$T_{1/2}$ = half life.

The relative activity of the W and Ta impurities will then be:

$$\frac{A_{187\text{W}}}{A_{182\text{Ta}}} = \frac{N_{186\text{W}}}{N_{181\text{Ta}}} \frac{\sigma_{186\text{W}}}{\sigma_{181\text{Ta}}} \frac{S_{187\text{W}}}{S_{182\text{Ta}}} \frac{D_{187\text{W}}}{D_{182\text{Ta}}}$$

For a 24 hr irradiation, the relative activity of W to Ta can be calculated for the material with the chemical analysis of Table 11.1 and using the nuclear data in Table 11.2. Immediately after the irradiation, the relative activity will be:

$$A_{187\text{ W}}/A_{182\text{ Ta}} = 23.97.$$

Thus, almost all of the activity (~96%) will be due to the W impurity in niobium. However, it is more practical to make the first autoradiograph at some time later. In this case, the first plate was a 15 min exposure some 25.6 hr after the end of the irradiation. Thus, the relative activity was reduced to $A_{187\text{ W}}/A_{182\text{ Ta}} = 11.43$. For this exposure, the activity was still largely due to the W impurity (~91%). The last radiograph in the series was a 2 hr exposure taken 9 days after the end of the irradiation. For this case, the relative activity is $A_{187\text{ W}}/A_{182\text{ Ta}} = 0.049$ which means that the Ta impurity now created most of the activity (~95%). Hence, by taking autoradiographs within 1 day and then about 10 days after the neutron irradiation, it is possible to record the distribution of W and Ta impurities, respectively, in niobium metal.

Another factor to consider is the range of penetration of the β^- particles in the photographic emulsion. The energies of the beta emission from W_{187} and Ta_{182} are 0.63 and 0.52 MeV, respectively.

These energies are similar enough to assume that the emulsion will respond in the same manner to each radionuclide. From penetration data for electrons into aluminum as a function of energy, it was estimated that about 80% of the β^- particles were stopped in the 100 μ thick nuclear emulsion on the autoradiograph plate.

Figure 11.2 is an enlarged print (~4X) of an autoradiograph taken 26.1 hr after the irradiation. Thus, the activity registered on this plate is due largely to β^- emission from W_{187} . There are several observations to be noted. First, the area which was the liquid zone prior to shutting off the electron beam is lighter on the plate (darker on print, Figure 11.2) indicating that the liquid contains less W than the solid. In fact, tentative calculations from the microphotometer readings give an effective distribution coefficient (concentration of W in solid divided by concentration in the liquid) $K = 1.06$. The Nb-W phase diagram predicts a K greater than 1. The Ta distribution coefficient calculated from later autoradiographs resulted in $K = 1.14$. It should be emphasized that these are preliminary figures. Second, the zoned section of the rod shows evidence that the W is inhomogeneously distributed. It occurs in alternate bands of impurity "rich" and "poor" layers that are roughly parallel to the liquid-solid moving interface. This was also true for the Ta impurity. Third, the liquid-solid

FIGURE 11.2. Print from Autoradiographic Plate of Zoned Niobium Containing W and Ta Impurities (4X)

interface is not planar but appreciably curved.

The distribution coefficients quoted here are tentative calculations due to problems caused by an inhomogeneous background on the autoradiograph plate. This was caused by γ radiation concentrations from the more massive zoned specimens compared to the thin standard disks. Future autoradiographs will use specimens and standards of the same thickness. The darkening due to the γ radiation is slight, but the inhomogeneous nature of this background "fog" did affect the microphotometer readings.

Conclusions

Tantalum and tungsten impurities are inhomogeneously distributed in as-zoned niobium rod. They lie in alternate layers of impurity "rich" and "poor" regions that are parallel to the moving liquid-solid interface during zoning. An autoradiographic technique shows promise in obtaining quantitative measurements on the Ta and W distribution.

THE EFFECT OF INTERSTITIAL IMPURITIES ON POSTIRRADIATION ANNEALING PHENOMENA IN NIOBIUM

J. M. Williams, J. T. Stanley, and W. E. Brundage

Introduction

In previously reported work,⁽⁷⁻¹¹⁾ we have been concerned with characterizing the interstitial impurity levels of several laboratory prepared niobium materials and determining the effects of these impurities on property changes during postirradiation annealing.

Interstitial impurity levels were estimated by internal friction and resistivity measurements; in two cases controlled additions of oxygen provided a further check on the analyses. Postirradiation annealing studies up to 200 °C revealed that resistivity decreases in the Stage III region (~150 °C) were attributable to segregation of oxygen in solid solution at radiation-produced dislocation loops.^(10,11) In the present work, postirradiation annealing

studies have been extended to about 550 °C for most of the samples. Also, chemical analyses of the interstitial impurities have been obtained for comparison with the results of the physical methods. This report concerns itself with materials previously labeled C, D, E, F, G, and P.^(7,8) We first present a brief recapitulation of the experimental procedures. Then a new summary and comparison of the interstitial impurity levels as determined by the various methods is presented. Finally, the results of annealing up to 550 °C are presented and discussed.

Experimental Procedures

Sample Preparation

Sample materials were prepared by swaging and drawing zone-refined niobium rods to 0.030 in. diameter wire suitable for both internal friction and resistivity measurements. The wires were then annealed and outgassed by resistive heating to a high temperature in a high-vacuum system. The annealing temperatures and final vacuum achieved during outgassing have been reported before.^(7,8)

After thorough outgassing, controlled quantities of oxygen were added to two of the materials, F and G. For material F, this was done by reheating the wire to about 1100 °C in a dynamic atmosphere of 5×10^{-6} torr of oxygen. From the approximate pumping speed of the sample at this temperature,

we estimated the amount of oxygen added as about 250 wt ppm. Approximately 50 wt ppm of oxygen was added to material G by anodizing the surface and dissolving the oxide as was described previously.⁽⁸⁾

Annealing and Measuring Procedures

Internal friction measurements were made in an inverted torsion pendulum placed inside an evacuated chamber. Grips and mounting fixtures were designed so that the samples could be mounted and dismounted without bending them. Thermocouples were mounted on both grips and separate heaters were used to maintain the grips at the same temperature. Except where otherwise noted, annealing of internal friction samples was done *in situ*.

Resistivity measurements were made with the samples immersed in liquid helium inside a superconducting magnet creating a field of 17.5 kG. It was necessary to use fine niobium wire for the primary potential contacts which establish the gage lengths of the samples because platinum leads would have become too radioactive during the irradiations. To make measurements, platinum wires were welded to these niobium contacts and copper leads were in turn soldered to the platinum.

Except for material G, all resistivity samples were isochronally annealed in a dynamically-evacuated tube furnace over the entire temperature range studied. The samples

were dismantled from the resistivity sample holder for each anneal, and they were thus subjected to considerable handling in this procedure. This resulted in the breaking of some of the Nb potential contacts after repeated mountings; some of the scatter in the annealing data at higher temperatures may be caused by slight changes in the sample geometry attendant upon replacing these leads. Resistivity samples from material G were annealed in a silicone oil bath up through 225 °C and were not dismantled between anneals. Sample holders used in these experiments left the samples unconstrained lengthwise so as to allow for thermal expansion and contraction during annealing and cooling to liquid helium.

Irradiation Procedures

Table 11.3 gives the ORNL irradiation facilities employed, the neutron fluences, and the irradiation temperatures for the irradiations performed on the various samples. The irradiation temperature was measured in each case by means of

a thermocouple spot-welded to a dummy sample placed inside the irradiation capsule with the rest of the samples.

Chemical Analyses

Oxygen and nitrogen analyses were done by the vacuum fusion technique and carbon analyses were done by Leco combustion. The analyses were carried out in ORNL's Analytical Chemistry Division by W. R. Laing, H. G. Davis, and J. L. Botts, and we are indebted to these workers for their cooperation. The number of significant figures used in reporting these results reflects the estimated accuracy.

Results

Material Characterization

Table 11.4 shows the comparison of the impurity levels as determined by the various techniques. In some cases, more than one analysis was obtained for a given material. The values determined by internal friction measurements and

TABLE 11.3. Information on Irradiations for the Niobium Samples

Sample	Facility	Neutron Fluence, n/cm ² (E > 1 MeV)	Irradiation Temp., °C
C,D	Low Intensity Test Reactor C-41	1 x 10 ¹⁸	45
E,F	Low Intensity Test Reactor C-41	2 x 10 ¹⁸	45
G	Bulk Shielding Reactor ⁽¹²⁾ Liquid Nitrogen Cryostat	2 x 10 ¹⁸	45

TABLE 11.4. Comparison of Interstitial Impurity Analyses for Niobium Impurity Concentrations in wt ppm

Material	Chemical Analysis			Internal Friction ^a			Resistivity ^b
	C	O	N	C	O	N	Total
C	20	17 60 10 40	9 16 <1 10	~3	<3	10	17
D	62	16	10	~30	<3	10	69
E	11	13 22	2 <1	<4	<4	<4	5
F	150	330	<1		260		250
G	15	96	<1	<4	85	<4	50
P ^c		2 3	2 1				3

a. Determined using internal friction data of Powers and Doyle.⁽¹⁴⁾

b. Determined by dividing residual resistivity by 0.0039 $\frac{\mu\Omega\text{-cm}}{\text{wt. ppm}}$ (13)

c. Parma-1 material used in deformation experiment, Reference 8.

the total impurity concentrations determined from resistivity measurements were reported previously.^(7,10) Unfortunately, as is indicated in Reference 7 (Figure 11.20), pre-irradiation internal friction measurements were not made above 200 °C on material F. Thus, we have no determination for C and N based on internal friction data in this material.

On surveying Table 11.4, it is apparent that there is general agreement among the various methods to the extent that they all rate the samples in the same order as concerns the total and individual impurity concentrations. However,

there is considerable disagreement in detail. To the limited extent that nitrogen is present, there is good agreement between the internal friction and vacuum fusion analyses. However, for carbon and oxygen the apparent discrepancies among the various methods warrant further discussion.

For carbon, we consider the agreement between internal friction and chemical analysis to be satisfactory in view of the present uncertainties associated with applying these methods to carbon. These uncertainties arise partly from the fact that carbon is scarcely soluble in niobium. The Snoek effect measures only the

amount of carbon retained in solution during cooling from the fairly high temperatures used in preparing the materials and is insensitive to precipitated carbon. Furthermore, as previously mentioned,⁽⁷⁾ the carbon retained in solution precipitates during the observation of the Snoek peak. Also, the resistivity is sensitive primarily to the carbon in solution. Leco combustion, on the other hand, measures all the carbon in the sample, although this method is considered inaccurate below 25 ppm for the sample weights employed. To summarize, we have physical evidence for carbon in materials C, D, and F. In materials C and D, this consists of internal friction evidence. As will be seen later, material F like material D exhibits a large annealing stage in the temperature region where carbon is mobile and we attribute this to precipitation of carbon. This lends credibility to the relatively high carbon analysis for this material.

In comparing the resistivity results with the internal friction results for materials F and G, we find that the internal friction results indicate somewhat larger oxygen concentrations than the total impurity concentration computed from the resistivity. It might be mentioned that the value of Tedmon, Rose, and Wulff⁽¹³⁾ of $0.0039 \frac{\mu\Omega - \text{cm}}{\text{ppm}}$ is the lowest value of the resistivity contribution we have found in the literature for any interstitial impurity in niobium. Thus, the total concentration determined from resistivity measurements should represent an

approximate upper limit. In computing the oxygen concentration from the Snoek peak, one assumes

$$C(\text{wt ppm Oxygen}) = KQ_p^{-1}$$

where Q_p^{-1} is the internal friction at the peak. The Powers and Doyle⁽¹⁴⁾ value for K is 3.64, but other values in the literature range from 2.65 to 4.09⁽¹⁵⁻¹⁹⁾ for low concentrations of oxygen. Thus, there is considerable latitude in estimating the concentration based on internal friction. In any case, the discrepancy between the internal friction and resistivity results appears worse on a percentage basis for material G than for material F. Even after precipitating the carbon in material F, the remaining resistivity corresponds to at least 200 wt ppm of oxygen. Possible sources of variations in damping values for interstitial impurities include the presence of substitutional impurities⁽¹⁷⁾ and preferred orientation of the recrystallized texture. The wires used in these experiments had a bamboo structure with grains ranging from 1 to 3 mm in length. Since wires F and G were drawn from different starting crystals, they could have different recrystallization textures and did, in fact, have different substitutional impurity contents. Material F had Ta and W totaling to about 369 wt ppm and material G had a total of 594 wt ppm of these impurities.⁽⁷⁾

The vacuum fusion results for oxygen are generally greater than those from the other methods, although they are quite reasonable for materials F, G, and P. For materials

C, D, and E, we believe the lower values indicated by internal friction are more correct. The internal friction values are consistent with the total impurity concentrations indicated by the resistivity values and also agree with those of other workers using similar sample preparation techniques.⁽²⁰⁻²²⁾

Postirradiation Annealing Studies

The isochronal annealing results are shown in Figure 11.3. Changes in resistivity are taken with respect to the preirradiation values. Annealing results up to 200 °C have been presented previously.⁽⁷⁻¹⁰⁾

Figure 11.3 shows that only samples F and G, which exhibited oxygen Snoek peaks, show a substantial resistivity decrease below 200 °C. Removal of oxygen from solid solution by association with radiation-produced defects causes the resistivity to drop below the preirradiation value in this temperature region. Sample G-6 (Figure 11.3) was first isothermally annealed at 150 °C and then isochronally annealed at higher temperatures. The entire resistivity decrease at 150 °C was previously correlated in time with the oxygen Snoek peak decrease for this material.⁽⁹⁾

Figure 11.4 shows a replot of the annealing results for material G (Figure 11.3) together with isochronal annealing results for an internal friction sample that had previously been isothermally annealed at 150 °C. The internal friction data were obtained by annealing at the temperatures shown and returning to 150 °C

to measure the Snoek peak height. Scales on the ordinates are adopted such that the resistivity decrease for sample G-6 at 150 °C is graphically equal to the internal friction decrease at 150 °C for G-3. Thus, the isothermal line at 150 °C represents both the internal friction decrease and the resistivity decrease at this temperature. The internal friction starts to increase by 300 °C, and by 550 °C it goes above the preirradiation value by about 20%. Upon isochronal annealing above 150 °C, sample G-6 exhibits a slight increase in resistivity and then decreases gradually between 200 and 250 °C, the temperature range where the internal friction starts to increase. Thus, although these two properties decreased in phase at 150 °C, they do not increase in phase, since the internal friction increases at a lower temperature than the resistivity and to a greater extent relative to the decrease at 150 °C.

Discussion

We attribute the large resistivity decrease between 200 and 350 °C in materials D and F (Figures 11.3a and b) to carbon precipitation. It occurs in both irradiated and unirradiated materials and, therefore, is primarily due to carbon in solution before irradiation. The effects of irradiation on carbon precipitation are not yet well understood because the precise state of the carbon in these materials before irradiation was not well defined. It is presumed that some of the carbon was

FIGURE 11.3. Isochronal Annealing Curves for Niobium Samples. One Hour Annealing Periods Except Specimen G-6 (2 hr Anneals)

precipitated before irradiation. Thus, in irradiated materials carbon might precipitate on radiation-produced defects or join existing nuclei. In material D, irradiation appears to have increased the amount of precipitation and lowered the tempera-

ture at which it occurs slightly, whereas in material F irradiation has had little effect. Possibly the oxygen, being more mobile than carbon, gets first claim on the dislocation loops in material F, but in oxygen-free material D the loops

ORNL-DWG 68-8338

FIGURE 11.4. Comparison of Postirradiation Annealing Behavior of Oxygen Snoek Peak and Resistivity for Niobium Material G. (For anneals above 400 °C the internal friction sample was dismantled from torsion pendulum and annealed in a manner similar to resistivity specimen. See text.)

are accessible to carbon. There is a suggestion of carbon precipitation in the irradiated C material (Figure 11.3a) but not in the control C material.

Material E (Figure 11.3b) exhibits only a small gradual decrease in resistivity until 350 °C after which there is a slight increase possibly due to contamination. Sample E control is not shown, but its resistivity curve increases slightly over the whole temperature range until it merges at about 450 °C with the one for the irradiated material E. Thus, for a niobium material containing only small amounts

of oxygen and carbon, no significant changes in resistivity are observed. This agrees with the previously reported measurements of internal friction for this material.⁽¹⁰⁾

We now turn our attention to the results for the high-oxygen, low-carbon material G, shown in Figure 11.4. It has been established⁽¹⁰⁾ that the annealing stage at about 150 °C in irradiated niobium is due to the trapping of oxygen at radiation-produced defect clusters or loops, which removes the oxygen from solid solution and decreases the resistivity and the height of the oxygen internal friction Snoek

peak. Furthermore, the kinetics of the oxygen removal is consistent with this interpretation.⁽¹¹⁾ It is to be expected that upon annealing at higher temperatures, the oxygen will dissociate from the radiation-produced loops and return to solid solution with an attendant increase in resistivity and internal friction. In a previous report, the increase in internal friction upon annealing up to about 500 °C was shown.⁽⁹⁾ This increase in internal friction was irreversible in that subsequent anneals at lower temperatures did not cause a return to the lower internal friction. This suggests that the radiation-produced loops anneal out when the oxygen dissociates from them. The irreversible dissociation of oxygen from the loops occurs upon annealing above about 300 °C. However, a reversible evaporation of oxygen may occur at lower temperatures as shown by Köthe,⁽²³⁾ and some evidence for this is seen in Figure 11.4 in the slight increase in resistivity at 175 to 200 °C for sample G-6.

A major problem exists in explaining the larger increase above 300 °C in Figure 11.4 for internal friction as compared to resistivity, when the two properties are normalized in the manner described above. One possibility is that the increase in resistivity caused by the return of oxygen to solid solution is partly compensated by a decrease in resistivity due to the annealing of the radiation-produced loops. Another possibility is that upon

annealing at the higher temperatures, a contribution to the internal friction (measured at 150 °C) is made from an additional source; other evidence for a "pseudo Snoek peak" is presented elsewhere in this report.*

PSEUDO SNOEK PEAK IN NIOBIUM

J. T. Stanley

Introduction

During the course of some recent investigations on the effect of oxygen on the annealing of neutron irradiated niobium, a number of specimens were prepared which exhibited an anomalous behavior of the oxygen Snoek peak. For the purposes of this discussion, the internal friction peak which appears in these specimens will be called the pseudo Snoek peak. This report describes the characteristics of the pseudo Snoek peak and presents the evidence that the pseudo Snoek peak is not a true Snoek peak.

Specimen Preparation

The specimens discussed in this report were all prepared from niobium stock obtained from Wah Chang Corporation containing approximately 367 ppm Ta and 227 ppm W as major metallic impurities. The bar stock was remelted in an electron-beam floating-zone apparatus by R. E. Reed. This treatment produced

* J. T. Stanley, next section, this report.

single crystals 3/16 in. diameter which were subsequently swaged and drawn to 0.030 in. diameter wire. Pieces of wire were pickled in a mixture of hydrofluoric and nitric acid, formed into a hair-pin shape, and then placed in a vacuum apparatus for annealing. The wires were heated by passing an electric current through them. A number of internal friction and resistivity samples were cut from each hair-pin. Specimens were given a three part designation; e.g., S5-A-1; where the first part, S5, is the number of starting single crystal; the second part, A, designates the hair-pin; and the third part, 1, designates the individual specimen. Table 11.5 lists the annealing conditions for the various specimens discussed in this report.

The wires after annealing at high temperature had a bamboo grain structure with grains 1 to 3 mm in length. The grain boundaries had an "etched" appearance, apparently due to preferential evaporation. In addition to the grain boundaries, microscopic examination of the specimens revealed another more faintly etched structure which might be sub-grain boundaries. Figure 11.5 is a photomicrograph of the surface of one of the specimens showing these markings.

Experimental Procedures and Results

Resistivity measurements were made with the samples immersed in liquid helium inside a supercon-

FIGURE 11.5. Photomicrograph of Surface of Niobium Wire (S5-A) Annealed at High Temperature. Wire Axis is Vertical in Photo and Approximately Horizontal Dark Band is a Grain Boundary. (200X)

ducting magnet. A field of approximately 17.5 kG was used to suppress the superconductivity of niobium. Small niobium wires were spot welded to the specimens to serve as potential leads and define a gage length about 1 in. long. The resistivity measurements were made by the potentiometer technique using a Rubicon Six Dial Microvolt Potentiometer. With these techniques, resistivity changes of a specimen as the result of annealing or irradiation could be measured with a precision of $\pm 0.01 \mu\text{ohm}$, but the absolute resistivity values are only accurate to $\pm 4\%$ because of the limited precision of the sample geometry measurements.

Internal friction measurements were made in an inverted torsion

TABLE 11.5. Annealing Conditions for Niobium Hairpins

Hairpin No.	Temperature, °C	Time, min	Vacuum Pressure, torr
S5-A	1800	5	9×10^{-7}
	2100	5	7×10^{-7}
	2100	12	1×10^{-9}
S5-B	2100	4	2×10^{-6}
	2100	8	1.5×10^{-7}
	2100	11	3×10^{-8}
	1700	30	5×10^{-8} (oxygen bled in to raise pressure from 2×10^{-8} to 5×10^{-8} torr)
27-A	2200	100	2×10^{-9}
57-A	1900	5	4×10^{-7}
	2100	15	1×10^{-8}

Note: In each case specimens were heated gradually to temperature shown. Where more than one temperature is given for a specimen it means that the specimen was heated more than once, the current being shut off between each heating.

pendulum which was placed inside an evacuated chamber. Grips and mounting fixtures were designed so that the samples could be mounted and dismounted without bending them. Thermocouples were mounted on both grips and separate heaters were used to maintain the grips at the same temperature.

The characteristic behavior of specimens having a pseudo Snoek peak is shown in Figure 11.6. The internal friction measured upon increasing temperature shows no sign of an oxygen Snoek peak, but upon making measurements with decreasing temperature a large internal friction peak appears at about the same tem-

perature as the oxygen Snoek peak. After this initial treatment, the pseudo Snoek-peak does not change with time at least for temperatures below 250 °C (the effect of further annealing at higher temperature has not yet been investigated). The pseudo Snoek peak shown in Figure 11.6 happens to be the largest observed. The height of the pseudo Snoek peak of various specimens is listed in Table 11.6. The small peak at 295 °C in Figure 11.6 is apparently a carbon Snoek peak since it is at the temperature of the carbon peak found by Powers and Doyle.⁽²⁴⁾ A carbon peak has been found in all but one of the

FIGURE 11.6. Internal Friction Versus Temperature of Specimen Nb-S5-A-3 Showing Characteristic Behavior of Pseudo Snoek Peak

specimens which exhibit the pseudo Snoek peak. The height of the carbon peak for the various specimens is given in Table 11.6.

The behavior outlined above is in sharp contrast to that of specimens exhibiting a normal Snoek peak. These specimens were prepared by giving first annealing treatments similar to those listed in Table 11.5 and then adding oxygen while annealing at about 1100 $^{\circ}\text{C}$. The methods used to add oxygen were described in a previous report.⁽⁸⁾

Specimens treated in this way showed oxygen peaks during the first run measurements and the height of the peaks remained constant for all runs on unirradiated specimens.

The effect of irradiation on the pseudo Snoek peak was investigated by irradiating specimen Nb-S5-A-4 to a fluence of 1×10^{18} n/cm^2 ($E > 1$ MeV) at 50 $^{\circ}\text{C}$. After irradiation and annealing at 145 $^{\circ}\text{C}$ for 24 hr, no change in the pseudo Snoek peak height was noted. The internal friction measurements on this

TABLE 11.6. Characteristics of Specimens Having Pseudo Snoek Peaks

Specimen	Pseudo Peak, Carbon Peak,		Residual Resistivity,		Chemical Analysis,		
	$Q_p^{-1} \times 10^4$	$Q_p^{-1} \times 10^4$	$\rho_o, \mu\Omega\text{-cm}$	$\rho_o/0.00068$	Carbon	Oxygen	Nitrogen
S5-A-1			0.071	104	155		
S5-A-2			0.058	86		98 230	59 66
S5-A-3	37.5	3.5	0.073	109		57	6
S5-A-4	20	3.5					
27-A-1	3.5	ND ^a			84	75	13
27-A-2			0.017	25			
27-A-3			0.016	24			
27-A-4						126	6
57-A-1	2.5	2.0					
57-A-2	7.2 ^b						
S5-B-1	4.5	12.5 ^c				92	66
S5-B-2			0.2684	346	480 ^d		
S5-B-3			0.1840	272			

a. ND = Not Detectable

b. Measured after irradiation

c. Carbon precipitates during measurement

d. S5-B-2 and S5-B-3 were combined for carbon analysis

specimen were made at three different frequencies with the results shown in Figure 11.7. The peak temperatures are plotted versus logarithm of frequency in Figure 11.8 for specimens showing pseudo Snoek peak behavior and normal Snoek peak behavior. There appears to be a small temperature difference between the temperature of the Snoek peak and the temperature of the pseudo Snoek peak for a given vibration frequency. This temperature difference is only 1.5 °C at

vibration frequency of 0.8 counts/sec, but we believe it is a real effect. The two straight lines in Figure 11.8 have the same slope corresponding to an activation energy of 26,800 cal/mole.

Discussion

The results presented in the previous section show that the effect which we are calling the pseudo Snoek peak has many characteristics that are the same as those of the

ORNL-DWG 68-7990

FIGURE 11.7. Internal Friction Versus Reciprocal Temperature of Specimen Nb-S5-A-4 for Different Vibration Frequencies

normal Snoek peak. Thus, the first task of this discussion is to present in detail the arguments that the pseudo Snoek peak is a different phenomenon than the real Snoek peak.

Clearly, the most distinguishing characteristic of the pseudo Snoek peak is the fact that it does not appear upon first measurement of specimens that were outgassed at high temperatures. However, after the initial series of measurements which are made up to about 340 °C, the pseudo Snoek peak appears fully developed. The initial reaction of many who have seen these data is that the specimen became contaminated with oxygen during the

measurements. However, contamination can be ruled out by a number of arguments. Specimen Nb-27-A-1, which showed only a small pseudo peak, was deliberately held at 350 °C for 16 hr in a vacuum of 20 μ to see if the peak would increase. A small increase from 3.5×10^{-4} was observed, but this is much smaller than the increase observed in specimens S5-A-4 and S5-A-3, which were held only a few hours at 350 °C in a vacuum of 10^{-4} torr.

Another argument against contaminations is the resistivity measurement that was made on Nb-S5A-3 after the pseudo Snoek peak

FIGURE 11.8. Arrhenius Plots for Snoek Peak (Open Points) and Pseudo Snoek Peaks (Filled Points) in Niobium

had been fully developed. The resistivity of this specimen was 0.073 $\mu\text{ohm-cm}$ at 4.2 °K. If we assume that this residual resistivity is entirely due to interstitial impurities and use the value of Tedmon, Rose and Wulff⁽¹³⁾ for the resistivity contribution of an oxygen atom, then the total interstitial impurity content would be 110 ppm (atomic). However, the height of the internal friction peak in this specimen corresponds to 570 ppm atomic oxygen if we use the value of Szkopiak⁽¹⁷⁾ for the

conversion of peak height to oxygen concentration. It should be noted that Szkopiak's value is the lowest value yet reported in the literature, while the resistivity conversion factor is also the largest reported in the literature so that if other reported values were used, the discrepancy between the two values would increase. There is also some evidence from internal friction and chemical analysis that sample Nb-S5A-3 contains some carbon; therefore, a fraction of the residual resistivity must be due

to carbon making the discrepancy between residual resistivity and oxygen peak height even greater.

The results of the vacuum fusion analysis made on specimen Nb-S5-A-3 after the internal friction measurements were somewhat inconclusive. The specimen was cut into two parts and one piece was etched in hydrofluoric-nitric acid to remove about 0.001 in. from the diameter before making the analysis. The other part was analyzed without etching. The etched half-specimen showed an oxygen content of 340 ppm (atomic), while the unetched half showed an oxygen content of 57 ppm (atomic). The same procedure was used on another specimen cut from the Nb-S5-A hair-pin which had not been used for any internal friction or resistivity measurements. In this case, the etched sample analyzed 230 ppm atomic and the unetched 97 ppm atomic. We believe that these results also indicate that contamination is not the cause of the large internal friction peak in specimen Nb-S5-A-3.

A second major difference between the behavior of the normal Snoek peak and the pseudo Snoek peak was observed for the irradiated specimens. Specimens having a normal Snoek peak show a decrease of the Snoek peak after irradiation and annealing at 150 °C.⁽⁹⁾ It has been shown that this effect is caused by migration of oxygen atoms to radiation-produced defects (primarily, small prismatic dislocation loops). The specimen having a pseudo Snoek peak, however, did not

show the decrease after irradiation and annealing. This result implies that these really are two different effects occurring in this temperature range.

At the present time, we are unable to offer a satisfactory explanation for the pseudo Snoek peak or even to state what conditions are necessary for its occurrence. However, from the limited experience available, it does appear that a large concentration of oxygen in solution suppresses the pseudo Snoek peak.

RADIATION ANNEAL HARDENING IN NIOBIUM

S. M. Ohr and R. P. Tucker*

We have reported previously⁽²⁵⁾ changes in the tensile properties and defect size distribution in neutron-irradiated polycrystalline niobium following postirradiation anneals in the temperature range between 200 and 800 °C. In addition to an increase in the yield stress of 5.8 kg/mm² following irradiation to a fluence of 2×10^{18} n/cm² ($E > 1$ MeV), a further increase of up to 8.5 kg/mm² in yield stress was observed upon annealing in the temperature range 200 to 400 °C. It was suggested that carbon atoms migrate to irradiation-produced defects during the anneal and subsequently are trapped by the defects. The trapped carbon atoms strengthen the defects as

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barriers to dislocation motion, thus causing further increase in the yield stress. This proposed strengthening mechanism was based in part on experimental evidence of Williams et al.,^(7,10) using internal friction and electrical resistivity techniques, which indicates that carbon atoms migrate in the temperature range where the hardening peak was observed. Here we describe the result of a further investigation into radiation anneal hardening and an analysis of the hardening process.

Figure 11.9 shows transmission electron micrographs of the deformation structures of as-irradiated and postirradiation annealed niobium. In the as-irradiated sample, the structure is characterized by numerous dislocation channels in which defect clusters have been removed by moving dislocations. In samples that were given postirradiation anneal at 400 °C for 2 hr and subsequently deformed to nearly identical strains, there is very little evidence of dislocation channeling. This observation is consistent with the conclusion reached previously that the defect clusters are strengthened as barriers by the trapped interstitial impurities.

Additional evidence in support of the impurity trapping mechanism of radiation anneal hardening is found in an experiment in which the carbon content in niobium samples was reduced from the previous level of 60 to 30 ppm by weight. In these low carbon samples, the

maximum hardening was observed following an anneal at 300 °C for 2 hr and the magnitude of the anneal hardening was reduced roughly by one-half. In addition, a small increase in the yield stress was also observed following a 2 hr anneal at 150 °C. According to Williams et al., this is the temperature range where interstitial oxygen migrates to defect clusters. Thus, all of the evidences accumulated so far indicate clearly that the radiation anneal hardening in niobium is impurity controlled.

Let us consider in detail the mechanism of radiation anneal hardening in niobium in terms of dispersed barrier interaction between trapped interstitial impurity atoms and moving dislocations. Following neutron irradiation to a fluence of $2 \times 10^{18} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$), defect clusters identified as dislocation loops at a density of $6.1 \times 10^{15} \text{ cm}^{-3}$ with an average diameter of 93 Å have been observed by transmission electron microscopy in niobium.⁽²⁶⁾ With 60 weight ppm carbon in niobium, the maximum number of carbon atoms available per loop defect is approximately 4000. During postirradiation anneals, some fraction of these carbon atoms are believed to migrate to the loops. The kinetics of impurity migration to dislocation loops has been discussed recently by Bullough et al.⁽¹¹⁾ for the case of oxygen migration in niobium. They found that in the early stage of impurity migration, the number of impurities, $N(t)$, that

(a) 2×10^{18} neutrons/cm² ($E > 1$ MeV)
 $\epsilon = 2.0\%$

(b) 2×10^{18} neutrons/cm² + 400 °C 2 hrs
 $\epsilon = 1.7\%$

FIGURE 11.9. Deformation Structure of (a) As-Irradiated and (b) Postirradiation Annealed Niobium

arrive at the loop in time t is given by

$$N(t) = 5.3 C_0 (ADt/T)^{3/5}, \quad (1)$$

where C_0 is the initial concentration of impurity in solution, A is the interaction parameter of Cottrell and Bilby, D is the diffusion constant, and T is the absolute temperature. The resistivity measurements of oxygen migration in niobium indicate that a $t^{3/5}$ - law is obeyed until approximately 20% of the available impurities have reached the loops. The rate of arrival of impurities then decreases rapidly and the impurity atmosphere around the loops is saturated. According to the available diffusion data of carbon in niobium,⁽¹⁴⁾ the saturation of impurities on dislocation loops is expected to occur after 2 hr at 300 °C. By this time, approximately 840 carbon atoms arrive at the loop, or roughly 8 carbon atoms per atomic site around the loop. Cochardt et al.⁽²⁷⁾ have shown theoretically that an edge dislocation in iron can accommodate approximately 8 carbon atoms per atomic site. Thus, the hardening maximum observed following anneals around 300 °C for 2 hr is consistent with the formation of saturated Cottrell atmospheres of carbon atoms around the loop as predicted by the kinetics of impurity migration.

It is not known at present as to how the trapped carbon atoms around the dislocation loops would interact

as a group with moving dislocations. It has been estimated in the past⁽²⁸⁾ that the interaction energy of a carbon atom with a dislocation in iron is approximately 0.5 eV. This interaction energy leads to the maximum restoring force per single carbon atom of

$$F_m = \mu b^2/16 \quad (2)$$

where μ is the shear modulus and b is the Burgers vector. For a group of carbon atoms, the maximum restoring force may be written as $n\mu b^2/16$, where n is the multiplication parameter. The increase in the yield stress is then given by

$$\begin{aligned} \Delta\sigma &= 2 F_m/b\ell \\ &= n\mu b^2/8 b\ell \end{aligned}$$

where ℓ is the planar spacing of the defects. Following neutron irradiation to a fluence of 2×10^{18} n/cm² ($E > 1$ MeV), the spacing ℓ is found to be 1.3×10^{-5} cm and the magnitude of anneal hardening $\Delta\sigma$ after 300 °C for 2 hr is 7.3 kg/mm². With these available data, the multiplication parameter n is 7.2. This is very close to the estimated number of carbon atoms trapped per atomic site following the postirradiation anneal. Since the moving dislocation intersects the defect at a point, it is reasonable to expect that only the carbon atoms present in a single atomic plane interact by contact with moving dislocations.

DISLOCATION CHANNELING IN NEUTRON-IRRADIATED NIOBIUM

R. P. Tucker* and M. S. Wechsler

Introduction

In recent years, there have been a considerable number of observations by transmission electron microscopy of defect clusters resulting from neutron irradiation, e.g., for bcc metals, Fe^(29,30), Mo⁽³¹⁻³⁵⁾, and Nb^(26,36). Plastic deformation following irradiation produces pathways cleared of defect clusters in a matrix which retains its as-irradiated appearance. The cleared pathways generally coincide with traces of planes upon which slip is expected. It then is reasonable to associate the formation of these pathways with a defect-sweeping action by dislocations moving on the slip plane. The term "dislocation channel" is used to describe the cleared pathway observable by transmission electron microscopy and "dislocation channeling," the process by which the pathway or channel forms. The dislocation channel as observed in TEM foils is bounded by the surfaces of the foil and by the walls of the channel. The "channel plane" is one of a set of planes parallel to the wall of the channel, and the "channel trace direction" is along the line of intersection of the channel wall and the foil surface.

Several years ago, Cottrell⁽³⁷⁾ proposed that each dislocation slipping on a plane upon which defect clusters are scattered would sweep away some of the defects, making the passage of subsequent dislocations on the swept plane easier and resulting in localized "run-away" slip. One of the first direct observations of dislocation channels was made by Greenfield and Wilsdorf⁽³⁸⁾ in irradiated-and-deformed copper. Seeger and Essmann^(39,40) indicated the correlation between dislocation channels and coarse, surface slip-markings in irradiated copper. Recently, Sharp⁽⁴¹⁾ studied the effects of deformation temperature, dose, and strain on the width of defect-free channels in neutron-irradiated copper single crystals and compared these results with those for surface slip bands. On the basis of the agreement between the channel width and channel spacing when compared with values for slip bands determined from surface replicas, Sharp's results strongly indicate that a correspondence exists between surface slip markings and dislocation channels.

In body-centered cubic metals, dislocation channels have been observed in irradiated-and-deformed iron,⁽³⁰⁾ molybdenum,^(31,42) and niobium.^(26,36) The dislocation channels in molybdenum were found to be consistent with slip on {110} and {112} planes.^(31,42) In the past, however, relatively little work on dislocation channeling has been reported in the bcc metals aside from observing that it exists.

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The present study was conducted to investigate in greater detail the crystal geometry of channeling in neutron-irradiated niobium. Particular configurations of dislocation channeling were observed using several diffraction vectors, which permitted the Burgers vectors of prominent dislocations hung-up along the walls of the channel to be deduced. Also, one case was examined under stereo conditions, which permitted an unambiguous determination of the channel geometry. As a result of the study, it was found that most of the channel planes were {110}, although a few cases of {112} channel planes were also observed. Some conclusions are drawn concerning the number of dislocations sweeping along the channel from observations of the offset of such markers as low-angle boundaries and dislocation helices when they are intersected by dislocation channels. Finally, some discussion is given of the mechanism of dislocation channeling.

Experimental Details

The niobium used in this study was obtained as dendritic powder from CIBA Limited. The powder was drop cast to yield a small ingot which was subsequently swaged into 3/16 in. diameter rod. The rod was seeded and given two electron-beam molten-zone passes, yielding a single crystal oriented for maximum resolved shear stress on the ($\bar{1}01$) [111] slip system (see References 43 and 44 for details on

the desired ideal orientation). The single crystal was cold-rolled to sheet about 0.13 mm thick from which tensile samples 4.2 mm wide and 12.7 mm in gage length were stamped out. The samples were given a recrystallization anneal at 1050 °C for 1 hr at a pressure of about 3×10^{-8} torr. This treatment resulted in an average grain diameter of 44 μ (see Figure 28 in Reference 45). The impurity content of the recrystallized material is shown in Table 11.7.

Although no systematic study has yet been conducted to characterize it, the grains exhibit a preferred orientation with [111] nearly perpendicular to the sample surface as determined by the electron diffraction patterns for electron microscope foils taken from the tensile samples.

The tensile samples from which the TEM samples were cut were two of those used in a fluence dependence study described in a previous report.⁽⁴⁵⁾ The irradiations were carried out in Tube 12 of the Hydraulic Facility of Position F-8 in the Oak Ridge Research Reactor where the neutron flux as determined by

TABLE 11.7. Impurity Content of Niobium in wt ppm

<u>Impurity</u>	<u>Content</u>	<u>Impurity</u>	<u>Content</u>
C	60	Ta	20
O	38	W	10
H	2	Zr	10
N	5	Hf	5
		Fe	1

nickel monitors was 2.0×10^{13} n/cm²/sec ($E > 1$ MeV) and the irradiation temperature was below 50 °C. The fluences received by the samples examined for dislocation channels were (Sample A) 1.8×10^{18} and (Sample B) 4.4×10^{18} n/cm². After a waiting period for radioactive decay, the sheet samples were deformed in tension in the laboratory at room temperature at a crosshead speed of 0.01 cm/min, which corresponds to a strain rate of 1.3×10^{-4} sec⁻¹. Samples A and B were examined in the electron microscope after being strained 2.7 and 6.6%, respectively. These strains correspond essentially to the strain at the lower yield stress, the lower yield stress being reached at a lower strain for Sample A. The stress-strain curves for the two samples show well-defined, sharp yield drops with a difference between the upper and lower yield stresses of 3.38 kg/mm² (Sample A) and 3.81 kg/mm² (Sample B). The curves are generally smooth in appearance with only the slightest evidence of "jerky flow." The lower fluence sample (Sample A) is above the fluence level where saturation in the fluence dependence of the yield stress is observed to set in (see Figure 30 in Reference 45). The particular sample examined as Sample A exhibits yield stresses somewhat higher than expected for its fluence level and actually even higher than the corresponding stresses for Sample B. For Sample A the upper and lower yield stresses were 14.44 and 11.06

kg/mm², respectively, and for Sample B, 14.16 and 10.35 kg/mm², respectively.

Pieces about 6 mm square were cut from the deformed sheet samples and electrolytically thinned to about 2500 Å in a solution 85 parts nitric acid and 15 parts hydrofluoric acid by volume. The foils were examined in a Hitachi 11A electron microscope operated at 100 kV. The microscope was equipped with a stage which could be tilted and rotated. The two degrees of freedom in the stage were used to obtain stereo pairs of photos and were convenient for varying the diffraction vector $\vec{g}$. For the stereo pairs, the normal to the foil surface was tilted approximately 8.5° from the electron beam. In correlating the micrographs and diffraction patterns, allowance was made for both the intermediate lens rotation and the extra inversion of the image by the objective lens for the micrograph. The micrographs were printed to give normal positive prints, i.e., with the emulsion side of the plate facing the emulsion side of the printing paper.

Results

In this section, we present a series of transmission electron micrographs illustrating various features of dislocation channeling in irradiated-and-plastically-deformed niobium. Table 11.8 lists the principle features illustrated in each case.

TABLE 11.8. Summary of Features Illustrated in Cases 1-7

<u>Case Number</u>	<u>Special Features*</u>
1	<ul style="list-style-type: none"> a. Regular channel array composed of two sets of channels, one set perpendicular to foil surface and one set inclined. b. Hung-up dislocations primarily on only one side of Channels 2 c. Tangles at channel intersections
2	<ul style="list-style-type: none"> a. Regular channel array b. Paired dislocation channels c. Channel-channel offset d. Channels in early stage e. Tangles at channel intersections
3	<ul style="list-style-type: none"> a. Triangular pattern of channels b. $\vec{g} \cdot \vec{b}$ criterion applied c. Channel-channel offset d. Conclude $\{110\}$ type channel planes
4	<ul style="list-style-type: none"> a. Dislocation array in channel b. Splitting-off of channels due to cross slip c. Channel not perpendicular to foil surface d. Edge-screw character analyzed e. $\vec{g} \cdot \vec{b}$ criterion applied f. Some tangle dislocations identified
5	<ul style="list-style-type: none"> a. Dislocation helix with depleted zone b. Channel-boundary offset c. Channel-helix offset
6	<ul style="list-style-type: none"> a. Channel-boundary offset b. Channel created at channel-boundary intersection c. Channel extending across boundary d. Depleted zone near boundary
7	<ul style="list-style-type: none"> a. Dislocation cell structure within channel b. Dislocations not rigidly confined to channel

* Cases 1-6, Sample B. Case 7, Sample A

Case 1 (Figure 11.10):

Figure 11.10 shows a nearly rectangular grid in Sample B composed primarily of two sets of channel traces, denoted by 1 and 2. The plane of the micrograph is (111)

and the operating reflection is $[\bar{1}10]$. As shown in the figure, the approximate trace directions for Channels 1 and 2 are $[\bar{1}\bar{1}2]$ and $[\bar{1}10]$, respectively. For Channels 1, the channel interface is sharp, the dislocation lines

FIGURE 11.10. Nearly Rectangular Pattern of Dislocation Channels in Sample B as Revealed by Tan

are largely out of contrast, and the faint images of the dislocation lines indicate that their projections in the (111) plane are parallel to the trace direction, $[\bar{1}\bar{1}2]$. This suggests, as discussed more fully below (e.g., see Case 4),

that the channel plane 1 is perpendicular to the foil surface and thus it is identified to be $(1\bar{1}0)$ and that the channeling dislocations seen in the micrograph are probably screw dislocations with Burgers vector $(a/2)[11\bar{1}]$. On the

other hand, the edges of Channels 2 are less well defined, the dislocations at their edges are in strong contrast, and the projections of the dislocations in (111) generally appear to lie across the channel trace (e.g., at A in Channel 2c). From this we conclude that Channels 2 are inclined to the foil surface, are identified as (110), and the channeling dislocations have $(a/2)[\bar{1}11]$ for their Burgers vector (see Case 4).

It is interesting that the dislocations in Channels 2 appear to be associated preferentially with one wall of the channel. We note also that Channels 1 are straight and of uniform width over rather long distances. The defect density near the channel walls is the same as it is well within the matrix. An increased defect density would be expected if channeling dislocations caused defect loops unfavorably oriented for the sweeping process to move to the channel wall say by prismatic slip. On the other hand, if many errant dislocations penetrated the channel walls say by cross slipping into the matrix, a region of gradually increasing defect density adjacent to the channel would be expected. For Channels 2, there is some suggestion of variation in defect density near the channel wall. This can, however, be accounted for on the basis of the wedge-shaped matrix region formed because of the inclined walls of Channels 2.

A feature noted repeatedly is the dislocation tangling at the in-

tersections of the channels. Another feature seen occasionally is the splitting and veering off of dislocation channels as seen in Figure 11.10 at B_1 and B_2 . This is discussed further for Case 4 below.

Case 2 (Figure 11.11):

Several interesting features are seen in the micrographs of Figure 11.11 which are of overlapping areas in Sample B. The plane of the foil is again near (111). Here the channels also form a rather orderly array, but the crystal geometry is less certain than for the channels in Figure 11.10. The channels which lie diagonally in the micrographs, e.g., A and A', are most likely $(\bar{1}01)$ planes channeled by $[1\bar{1}1]$ dislocations, based on the observed trace direction, $[1\bar{2}1]$. The determination of the indices of the generally vertical channels (Channels V) is less certain because of their lack of parallelism and changes of direction. The {110} plane which has a trace nearest the direction of these channels is $(\bar{1}10)$. In Figure 11.11, tangles are observed at the intersections of most channels and in some cases the local tilting of the lattice due to the tangle is sufficient to cause essentially complete diffraction of the beam, producing the dark regions at some intersections. Several dislocations can be seen in the matrix outside of the well-formed channels. These dislocations presumably are contributing to the

formation of the numerous ill-defined, narrow channels in Figure 11.11a, (e.g., at B), which are along the trace direction $[1\bar{2}1]$ for $(\bar{1}01)$ planes.

A noteworthy feature occurs where the nearly vertical channels $V_1 - V_4$ cross the diagonal ones $A_1 - A_4$ in Figure 11.11. At many of these intersections the diagonal channel experiences an offset, with the segment to the right of the vertical channel sheared upward with respect to the segment to the left. For example, such an offset is seen at C in Figure 11.11b. Another interesting feature is the pair of Channels A and A', whose entire length can be seen in Figure 11.11. Tangles are again seen at the ends of the channels and at most intersections along their lengths. The upper channel segments, $A'_1, A'_2,$ and A'_3 , are essentially straight and in line while the lower channel segments $A_1, A_2, A_3,$ and A_4 , are offset from one another at intersections with V channels. It seems reasonable to postulate the following sequence of events. Initially, a diagonal channel extended from S to T'_1 , possibly due to the operation of a source at S. Then, the Channels $V_2, V_3,$ and V_4 operated, causing the shear displacements indicated by the arrows in Figure 11.11. This caused the channel previously extending from S to T'_1 to consist of the offset segments $A_4, A_3, A_2,$ and A_1 . Finally, the source at S operated again which produced the in-line channel segments $A'_3, A'_2,$ and A'_1 .

Case 3 (Figure 11.12):

Figure 11.12 shows an equilateral-triangular pattern of channels on a (111) foil surface from Sample B. The sketch in Figure 11.12a gives the number designations of the channels, the channel trace directions as obtained from the electron diffraction patterns, and the slip systems operating in the channels. The operating slip systems (i.e., the planes of the channels) and the Burgers vectors of the slip dislocations, were deduced as described below. This determination was facilitated by the diffraction contrast analysis of micrographs of the same field taken with $[01\bar{1}]$, $[10\bar{1}]$, and $[1\bar{1}0]$ operating reflections ($\vec{g}$ vectors), as shown in Figures 11.12b, 11.12c, and 11.12d, respectively. The vertex angles of the triangle in Figure 11.12 were measured in the micrographs to be 60° to within $\pm 2.5^\circ$.

Now consider planes of the $\{110\}$, $\{112\}$, and $\{123\}$ types as possible slip planes. There are two sets of three $\{110\}$ -type planes which give traces 60° apart in a (111) plane. Of these two sets, only one, namely $(01\bar{1})$, $(10\bar{1})$, and $(1\bar{1}0)$, will give rise to channel traces in the observed $\langle 112 \rangle$ directions. The traces for the other set, (011) , (101) , and (110) , form an equilateral triangle rotated 60° about $[111]$ from the trace directions observed. The normals for the three $\{110\}$ planes corresponding to the observed channel traces lie in the

FIGURE 11.11. Transmission Electron Micrographs from Sample B in Which Paired Dislocation Channels Are Observed. (a) Left Field of View. (Case 2)

FIGURE 11.11. Transmission Electron Micrographs from Sample B in Which Paired Dislocation Channels Are Observed. (b) Right Field of View. (Case 2)

ORNL-DWG 68-8492

FIGURE 11.12. Triangular Pattern of Dislocation Channels from Sample B. (a) Sketch of Channel Geometry. (Case 3)

(111). Thus, the channeled $\{110\}$ planes would form a right equilateral-triangular prism whose axis is along $[111]$. Since there are no $\{112\}$ planes which give $\langle 112 \rangle$ traces in a (111) plane, $\{112\}$ channel planes may be discounted. As for $\{123\}$ planes, there are two planes of this type giving traces in each of the required $\langle 112 \rangle$ directions. In all cases, the angle between the normals to

these $\{123\}$ planes and $[111]$ is $22^\circ 12'$.

We shall now consider the contrast of the dislocations for the various operating reflections shown in Figure 11.12. Most of the dislocations are observed to lie parallel to, but not always in, the channel trace. In bcc metals, the $\langle 111 \rangle$ close packed direction is the normal slip direction and hence the Burgers vector direction for

FIGURE 11.12. Triangular Pattern of Dislocation Channels from Sample B. (b) Transmission Electron Micrograph for $\vec{g} = [01\bar{1}]$. (Case 3)

FIGURE 11.12. Triangular Pattern of Dislocation Channels from Sample B. (c) Transmission Electron Micrograph for $g = [10\bar{1}]$. (Case 3)

FIGURE 11.12. Triangular Pattern of Dislocation Channels from Sample B. (d) Transmission Electron Micrograph for $\vec{g} = [1\bar{1}0]$. (Case 3)

slip dislocations. Diffraction contrast theory⁽⁴⁶⁾ predicts that when the Burgers vector, $\vec{b}$, lies in the reflecting plane, the dislocation is invisible if pure screw and generally exhibits weak contrast or is invisible if pure edge. Thus $\vec{g} \cdot \vec{b} = 0$ means invisibility or or weak contrast for dislocations, where $\vec{g}$ is the diffraction vector. In Table 11.9 the contrast for dislocations under three operating reflections (see Figure 11.12) is compared to $\vec{g} \cdot \vec{b}$ for $\langle 111 \rangle$ Burgers vectors. From Table 11.9 we conclude that the Burgers vector for dislocations lying parallel to the traces of Channels 1, 2, and 3 are $(a/2)[\bar{1}11]$, $(a/2)[1\bar{1}1]$, and $(a/2)[11\bar{1}]$, respectively.

It should be noted, as shown in Table 11.9, that the $[111]$ dislocations are always invisible for reflections from planes of the 111 zone. However, since the tensile axis lies in (111) to within a few degrees, $[111]$ dislocations would be subjected to little or no shear stress under uniaxial tension for

TABLE 11.9. Contrast and Values of $\vec{g} \cdot \vec{b}$ for Dislocations in Case 3

Channel Number	Contrast for Dislocations Parallel to Channel		
	$\vec{g} = [011]$ Fig. 3b	$\vec{g} = [101]$ Fig. 3c	$\vec{g} = [1\bar{1}0]$ Fig. 3d
1	W	S	S
2, 2a	S	W	S
3	S	S	W
$\vec{b}$	$\vec{g} \cdot \vec{b}$		
$(a/2)[111]$	0	0	0
$(a/2)[\bar{1}11]$	0	-1	-1
$(a/2)[1\bar{1}1]$	-1	0	1
$(a/2)[11\bar{1}]$	1	1	0

S = Strong contrast, i.e., the dislocation is visible ($\vec{g} \cdot \vec{b} \neq 0$)

W = Weak contrast, i.e., the dislocation is invisible ($\vec{g} \cdot \vec{b} = 0$)

the sample geometry used. These dislocations could experience stress state which exists in polycrystalline material; nevertheless, $[111]$ dislocations are expected to be of relatively little importance in the deformation under consideration. Therefore, the three reflections in Figure 11.12 bring into contrast essentially all the channeling dislocations which remain "hung up" along the channel walls.

Table 11.10 lists the possible planes for each channel and indicates the direction of the projection on (111) of the $\langle 111 \rangle$ -type Burgers vector lying in the channel planes. We see that only the $\{110\}$ slip planes perpendicular to the (111) plane contain dislocations whose Burgers vectors would project on the (111) along the required $\langle 112 \rangle$ channel traces. Table 11.10 also shows that the projection of the Burgers vector for channeling dislocations on the $\{123\}$ type planes would not be parallel to their channel trace directions, but rather could be parallel to either of the other two channel directions in Figure 11.12. Since we do observe dislocations lying parallel to the direction of the channel in which they move, we conclude that the channel planes are the $\{110\}$ set indicated in Table 11.10. There are, however, dislocations in contrast in Figure 11.12b, for example, which do not lie parallel to Channel 1. However, these dislocations extend beyond the width of the channel and are not clearly associated

with the channel. Similar observations apply to a few dislocations crossing Channels 2 and 3.

It should be noted that Channel 3 does not appear to be cleared of defects to the same extent as Channels 1 and 2. This might suggest that Channel 3 is inclined to the foil surface and thus is (312) or (132) (see Table 11.10). Since dislocations lie along the trace of Channel 3 (Figures 11.12b and 11.12c), which is not possible for the $\langle 111 \rangle$ dislocations on these two $\{123\}$ planes, we believe that the appearance of Channel 3 can be accounted for on the basis that it is in an early stage of formation. Perhaps the dislocations forming Channel 3 originate from the dislocation tangle at the intersection of Channels 1 and 2a and thus were generated at a later stage in the deformation than those sweeping Channels 1, 2, and 2a.

Another equilateral-triangular channel pattern from Sample B, for which micrographs are not shown, was examined using the stereo technique. The channel trace directions in the (111) were the same as those for Case 3. From an examination of the stereo pairs, it appeared that all of the channel planes in this particular field were perpendicular to the plane of the (111) foil surface. Thus, the crystal geometry of these channels was determined uniquely to be the same as the set of $\{110\}$ planes indicated in Table 11.10.

It seems clear from the offset in Channel 1 that Channel 2a formed

after Channel 1 or, at the least, continued to serve as an active slip channel after Channel 1 formed. The magnitude of the offset in the direction of Channel 2a is about 1.5 cm in prints at a magnification of 50,500X. Thus, the actual magnitude of the offset in Channel 1 is about 3000 Å. For niobium, the Burgers vector for slip dislocations is 2.86 Å⁽⁴⁷⁾ and the magnitude of its component in the direction of Channel 2a is 2.70 Å. Therefore, we conclude that approximately 1100 dislocations have traveled along Channel 2a to produce the offset observed in Channel 1. It is interesting to note that the width of Channel 2a is also about 3000 Å. The spacing between $\{110\}$ planes in niobium is 2.33 Å and thus approximately 1300 ($\bar{1}01$) planes are needed to account for the width of Channel 2a. The ratio of the number of dislocations sweeping the channel to the number of adjacent ($\bar{1}01$) planes within the channel is 0.9. As will be discussed, this ratio need not be taken as evidence that only approximately one dislocation has moved down each plane within the channel.

Case 4 (Figure 11.13):

The field of view chosen for Case 4 is of particular interest for in it is displayed an array of channeling dislocations at an early stage of channel formation. The area discussed as Case 4 is shown in Figure 11.13, where, as before, the plane of the foil from Sample B

TABLE 11.10. Crystal Geometry of Channels in Case 3

Channel Number	Possible Channel Planes	$\vec{b}$ for Channeling Dislocations	Direction of Projection of $\vec{b}$ in (111)		Trace Direction	Angle Between [111] and Plane Normal
			Crystal Direction	Parallel to Channel		
1	(01 $\bar{1}$)*	(a/2)[$\bar{1}11$]	[$\bar{2}11$]	1	[$\bar{2}11$]	90°
1	(231)	(a/2)[$\bar{1}11$]	[$\bar{1}21$]	2	[$\bar{2}11$]	22° 12'
1	(213)	(a/2)[$\bar{1}11$]	[$\bar{1}\bar{1}2$]	3	[$\bar{2}11$]	22° 12'
2	($\bar{1}01$)*	(a/2)[$\bar{1}11$]	[$\bar{1}21$]	2	[$\bar{1}21$]	90°
2	(123)	(a/2)[$\bar{1}11$]	[$\bar{1}\bar{1}2$]	3	[$\bar{1}21$]	22° 12'
2	(321)	(a/2)[$\bar{1}11$]	[$\bar{2}11$]	1	[$\bar{1}21$]	22° 12'
3	($\bar{1}10$)*	(a/2)[$\bar{1}11$]	[$\bar{1}\bar{1}2$]	3	[$\bar{1}\bar{1}2$]	90°
3	(132)	(a/2)[$\bar{1}11$]	[$\bar{1}21$]	2	[$\bar{1}\bar{1}2$]	22° 12'
3	(312)	(a/2)[$\bar{1}11$]	[$\bar{2}11$]	1	[$\bar{1}\bar{1}2$]	22° 12'

* Planes deduced to be the operating one in Figure 11.12

is (111). As mentioned previously, this region overlaps slightly the area shown in Figure 11.10. The number designations of channels and the channel trace directions as obtained from the electron diffraction pattern are shown in Figure 11.13a. Figures 11.13a and 11.13b show the field under reflections $[0\bar{1}1]$ and $[\bar{1}10]$, respectively. Here we observe channel traces along $\langle 110 \rangle$ directions as well as in $\langle 112 \rangle$ directions. In particular, the traces of Channels 2 and 3 lie along $[0\bar{1}1]$ and $[\bar{1}10]$, respectively. There are no $\{123\}$ planes which give $[0\bar{1}1]$ or $[\bar{1}10]$ traces in a (111) plane; thus, we need consider only $\{110\}$ and $\{112\}$ planes for Channels 2 and 3. The ($\bar{2}11$) and the ($11\bar{2}$) planes which are perpendicular to the (111) plane can be ruled out as planes for Channels 2 and 3, respectively, on the basis that no dislocations seen in these channels lie along the channel trace direction, which would necessarily follow for channel planes perpendicular to the foil surface. Further-

more, the only $\langle 111 \rangle$ direction lying in these planes is $[111]$ which, as argued for Case 3, is not expected to be an important slip direction for the deformation geometry used. The (211) plane would give a trace in the $[0\bar{1}1]$ direction as required for Channel 2 and would contain dislocations with (a/2) $[\bar{1}11]$ Burgers vectors. However, $[\bar{1}11]$ dislocations would be in contrast for $\vec{g} = [1\bar{1}0]$ (see Figure 11.13b). Since the dislocations in Channel 2 are out of contrast in Figure 11.13b the (211) plane is ruled out as the channel plane. A similar argument when applied to Channel 3, which shows an absence of dislocation contrast in the channel for Figure 11.13a, rules out the (112) plane with $[11\bar{1}]$ dislocations. Therefore, we conclude that the channel planes for Channels 2 and 3 are (011) and (110), respectively, which are inclined 35° 16' to the foil surface (see Table 11.11). Following the analysis outlined in Case 3, we find the crystal geometry for the

TABLE 11.11. Crystal Geometry of Channels in Case 4

Channel Number	Channeled Plane	$\vec{b}$ for Channeling Dislocations	Channel Trace Direction
1a, 1b	($\bar{1}\bar{1}0$)	$(a/2)[11\bar{1}]$	$[11\bar{2}]$
2	(011)	$(a/2)[11\bar{1}]$	$[01\bar{1}]$
3	(110)	$(a/2)[\bar{1}11]$	$[\bar{1}10]$
4	(011)	$(a/2)[\bar{1}11]$	$[2\bar{1}\bar{1}]$

remaining channels in Case 4 to be as given in Table 11.11. Channel 5 is the relatively wide curved channel lying in the direction from lower left to upper right in the micrographs. Because of its curvature, we have not attempted to analyze Channel 5.

It is interesting to note from Table 11.11 that dislocations channeling along Channels 1 and 2 have the same Burgers vector. Thus, dislocations moving in Channel 1a can transfer to Channel 2 by cross slipping from ($\bar{1}\bar{1}0$) to (011). If the dislocations observed in Channel 2 were pure screw with a $(a/2)[11\bar{1}]$ Burgers vector, then their projection on the (111) plane would lie in the $[11\bar{2}]$ direction. If they were pure edge, then their projection would lie in the $[110]$ direction. The dislocations in Channel 2 actually make an angle of about 45° with the $[11\bar{2}]$ direction. Thus, the dislocations appear to lie almost exactly half-way between the pure edge and pure screw orientations. However, the edge character may have been introduced during thinning. In this event, the dislocations observed in Channel 2 could have cross slipped from the ($\bar{1}\bar{1}0$) plane; the fact that no

tangle is observed at the intersection of Channels 1a and 2 would lend support to this idea. Note, however, tangling is observed at the intersections of Channel 2 with Channels 3 and 5 for which the channeling dislocations have Burgers vectors different from those in Channel 2. There is some suggestion that many of the dislocations seen in Channel 2 are paired, with the two dislocations comprising the pair lying in opposite walls of the channel. These paired dislocations appear to have approximately the same length, but are displaced with respect to one another. This is precisely the displacement expected for the projection of dislocation lines lying on opposite walls of a channel inclined to the foil surface such as Channel 2.

In general, the tangles observed at the intersections of channels are rather complex three-dimensional dislocation arrays. However, focusing attention on the tangle at the intersection of Channels 2 and 5, comparison of Figure 11.13a and 11.13b indicates that $[11\bar{1}]$ dislocations which are the channeling dislocations for Channel 2 make up part of the tangle. In Figure 11.13b, dislocations with $(a/2)[\bar{1}11]$ and $(a/2)[11\bar{1}]$ Burgers vectors are expected to be in contrast. Since Channel 5 appears curved, its plane is somewhat uncertain. If, however, we take the directions of Channels 4 and 5 to be parallel, then $[\bar{1}11]$ dislocations would move in Channel 5 and would have a projection

FIGURE 11.13. Paired Dislocations Along Channel in Sample B.
 (a) Visible with $\vec{g} = [011]$. (Case 4)

FIGURE 11.13. Paired Dislocations Along Channel in Sample B.
(b) Invisible with $\vec{g} = [\bar{1}10]$. (Case 4)

in (111) along the channel direction. In Figure 11.13b, dislocations with this characteristic are observed in the tangle, as well as dislocations making an angle of approximately 60° with the direction of Channel 5. The dislocations lying at 60° to the channel direction are rather sharply kinked in at least two places. Although we are not able to determine the Burgers vector for these dislocations from the available reflections, dislocations with either $(a/2)[1\bar{1}1]$ or $(a)[010]$ Burgers vector would satisfy the geometry observed and should be in contrast in Figure 11.13b. The $(a)[010]$ dislocations could result from the following reaction:

Dislocation reactions of this kind in bcc metals have been discussed by Hartley⁽⁴⁸⁾ and are found to be energetically favorable.

Case 5 (Figure 11.14):

A striking feature observed in a (111)-foil from Sample B is the dislocation helix shown in Figure 11.14a and at a higher magnification in Figure 11.14b. Aside from the fact that helices are rather infrequently observed in high-purity metals,⁽⁴⁹⁾ its occurrence here is of particular interest since the offset produced where a channel crosses it provides a measure of the number of dislocations moving along the channel.

Furthermore, there is an opportunity for comparing the number of dislocations moving in a given channel within a grain with the number in the same channel crossing the boundary seen at the upper right in Figure 11.14a.

The helical character of the feature in Figure 11.14 is clearly seen in the segment to the left of the vertical channel trace (A', Figure 11.14b). Discrete closed prismatic loops exist at A, Figure 11.14b, with the same axis as the helix. The line feature lies along $[01\bar{1}]$, and lower magnification micrographs indicate that it extends over a distance of at least 9.5μ . The axis of the linear dislocation array is, in general, straight. However, it does exhibit some twisting as seen at B, Figure 11.14a. Notice also that the spot damage believed to be interstitial in nature,^(26,36) is depleted on either side of this array. The spacing between turns and loops is essentially uniform over the length of the feature and is about $280 \text{ \AA}$ as determined from Figure 11.14b. The radii of the helix and loops in Figure 11.14b are approximately 350 to 450 $\text{\AA}$.

Helical dislocations are generally considered (e.g., Mitchel⁽⁵⁰⁾) to form from screw dislocations by climb processes with the axis of the helix in the direction of the Burgers vector of the screw. The long length of the helix in Figure 11.14a along $[01\bar{1}]$ suggests that it originated from a grown-in $[01\bar{1}]$ screw dislocation.

The trace of the channel causing the offset in the helix (nearly vertical in Figure 11.14b) is along $[\bar{2}11]$ and the channel plane is probably $(01\bar{1})$. This is so because the $\langle 111 \rangle$ dislocations in the other possible channel planes, (231) and (213) , lie across the channel trace instead of along it as observed for example at C, Figure 11.14b. The offset of the helix in the $[\bar{2}11]$ direction as determined from Figure 11.14b is 9500 Å. The dislocations with $(a/2)[\bar{1}11]$ Burgers vectors moving in $(01\bar{1})$ produce the observed offset. The component of a $(a/2)[\bar{1}11]$ Burgers vector in the $[\bar{2}11]$ direction is 2.70 Å. Therefore, the number of dislocations required to give the observed offset of the helix is about 3500. From Figure 11.14a, the channel width is 4500 Å. Since the spacing between $\{110\}$ planes is 2.33 Å, approximately 1900 $(01\bar{1})$ planes are needed to account for the channel width. The ratio of the number of dislocations to the number of channeled planes is 1.8, which is twice the ratio found in Case 3.

From Figure 11.14a, the offset in the boundary is found to be about 6700 Å which corresponds to the passage of 2500 $[\bar{1}11]$ dislocations. Therefore, approximately 1000 or more dislocations have passed the helix than have crossed the boundary. This is consistent with the observed dislocation pile-up and tangling at the side of the boundary facing toward the helix (D, Figure 11.14a).

In Figure 11.14a, two channels cross the linear array at angles not perpendicular to its axis. The channel marked E lies along $[\bar{1}\bar{2}1]$ and is perpendicular to $[\bar{1}01]$. In this case, the channeled plane is most likely the $(\bar{1}01)$ with $(a/2)[\bar{1}11]$, the Burgers vector for the channeling dislocations. The trace of Channel F is in the $[\bar{1}01]$ direction with (101) as the most likely channel plane. The angle between the normal to the (101) plane and the $[111]$ direction is $35^\circ 16'$. Considerable dislocation structure is observed in Channel F. Very little, if any, offset is to be seen where these channels cross the linear feature.

Case 6 (Figure 11.15):

In contrast to previous cases for Sample B, the plane shown in Figure 11.15 is (112) . The diffraction patterns indicate that the offset feature is a low-angle grain boundary. The trace of Channel G (approximately horizontal) lies within 4° of $[3\bar{1}\bar{1}]$. The numerous dislocations seen nearly parallel to the edge of the channel are consistent with $(a/2)[\bar{1}11]$ screw dislocations moving on the channel plane $(0\bar{1}1)$. The projection of $[\bar{1}11]$ in (112) lies along $[\bar{4}21]$, which is about 9° from $[3\bar{1}\bar{1}]$, the trace direction of the channel plane, $(0\bar{1}1)$, in (112) . Although not clearly apparent in Figure 11.15, other micrographs show that the trace of Channel G continues along $[3\bar{1}\bar{1}]$.

FIGURE 11.14. Channel-Helix and Channel-Boundary Offsets in Sample B. (a) Low Magnification. (Case 5)

FIGURE 11.14. Channel-Helix and Channel-Boundary Offsets in Sample B. (b) High Magnification. (Case 5)

FIGURE 11.15. Large Channel-Boundary Offset in a (112) Foil from Sample B. (Case 6)

to the left of the boundary and the channel labelled K is another channel which terminates at the offset located in the region between K and G. It seems reasonable to suggest that Channel K was initiated as a result of stress concentrations at the offset.

The magnitude of the offset in the boundary in Figure 11.15 provides basis for a calculation of the number of channeling dislocations operating in Channel G. The offset displacement corresponds to 12,100 Å and the component of the Burgers vector $(a/2)[\bar{1}11]$ along $[\bar{4}21]$ is 2.52 Å. Therefore, about 4800 dislocations are required to produce the offset. The width of the trace of Channel G corresponds to 4000 Å, which is provided by a stack of about 1600 $(0\bar{1}1)$ planes intersecting $(11\bar{2})$. Thus, on the average about $4800/1600 = 3 [\bar{1}11]$ dislocations have been channeled down each $(0\bar{1}1)$ plane, which is about three times that calculated for Case 3. The magnitude of the channeling shear due to Channel G in Figure 11.15 is also seen by the displacement of the channel trace at H to its position at H'. It is apparent that Channel H-H' must have formed prior to Channel G.

The offset, almost horizontal, segment of the boundary in Figure 11.15 is remarkably straight, which suggests that the channeling dislocations were distributed uniformly across Channel G. It is interesting, also, that the boundary is lengthened in the region of the offset. Another feature in Figure 11.15 is

the depleted region adjacent to the boundary, for example at J. The defect clusters in these samples are considered to be interstitial in nature.^(26,36)

Case 7 (Figure 11.16):

In comparison to Sample B, which has been the focus of attention in the preceding discussion, the area of the $(11\bar{1})$ foil from Sample A seen in Figure 11.16 exhibits some unexpected features. Both the fluence and the strain in Sample A were less than in Sample B, namely, $1.8 \times 10^{18} \text{ n/cm}^2$ and 2.7% as compared to $4.4 \times 10^{18} \text{ n/cm}^2$ and 6.6%. Although the yield stresses in Sample A were slightly greater than those in Sample B, the density of radiation-produced defect clusters in Sample A is noticeably lower than in Sample B.

A striking feature seen at M within the main channel in Figure 11.16 is the rather well-developed cell structure. The cell walls generally lie along crystallographic directions which are important in the crystal slip geometry. The dark tangle lying across the main channel to the left of M is in the $[11\bar{2}]$ direction, and to its right the cell walls are aligned with the channel trace direction, $[\bar{1}2\bar{1}]$. On the other hand, to the left of this dark tangle the uppermost cell wall is in the $[0\bar{1}1]$ direction. When viewed under the $[\bar{1}10]$ reflection, in a micrograph not

FIGURE 11.16. Cell Structure Within Channel Observed in Sample A After 2.7% Nominal Tensile Strain. (Case 7)

shown here, an entirely different set of more or less parallel, joggy dislocations are brought into contrast which form an interlaced network with those seen to the right of M in Figure 11.16 where $\vec{g} = [0\bar{1}1]$. The high dislocation concentration in the cell walls is in marked contrast to the cell interior (e.g., at M) which is almost entirely free of dislocations. The fact that a cell structure is observed after only 2.7% nominal strain in the sample emphasizes that the strain is concentrated in the channel.

Discussion

The observation of an offset where microstructural features are crossed by dislocation channels is quite significant, because it provides a measure of the direction and the magnitude of the shear displacements produced by the channeling dislocations. Three types of offsets are illustrated above: channel-channel (Cases 2 and 3), channel-helix (Case 5), and channel-low angle boundary offsets. Table 11.12 summarizes the results. We conclude from the column on the extreme right that, on the average, 1 to 3 dislocations move along the channel per atomic plane. This agrees with Sharp's estimates of 2 to 3 dislocations per plane for neutron-irradiated copper.⁽⁴¹⁾ Sharp estimated this quantity by dividing the shear displacement as observed from slip markings

by the channel width observed by transmission electron microscopy. Since our determination is based on the shear displacement and channel width for the same channel, the contention that surface slip lines correspond to dislocation channels is strengthened.

The formation of a cell structure within a dislocation channel for a sample given only 2.7% nominal strain (Figure 11.16) emphasizes, as we have said, the fact that strain is heavily concentrated in the channel. Nevertheless, within the channel the strain appears to be distributed uniformly. This is seen quite clearly in Figure 11.15 since the offset segment of the low-angle boundary is straight. This, again, is a conclusion reached by Sharp⁽⁴¹⁾ based on surface replicas. The uniformity of shear within the channel is consistent with the observation that there generally appear to be no defect clusters within the cleared channel when the channel plane is perpendicular to the plane of view. If, on the average, two dislocations per plane are considered to sweep down the channel, it is not necessary to assume that each plane receives two dislocations. Macroscopic uniformity of slip within the channel would also be achieved if packets of, say, 20 dislocations sweep down single planes spaced about 10 planes apart. However, for complete annihilation of the damage clusters within the channel the spacing between the

TABLE 11.12. Offsets Observed in Sample B

Case	Type Offset	Channel Plane	Channel Width	No. Planes Channel Width	Offset Magnitude ^c	Offset Direction ^c	$\frac{b}{a}$ for Channeling Dislocations	No. Dislocations/Offset	Ratio: (i)/(e)
2 ^a	Channel-Channel	(110)	2500-3330 Å	1070-1430	3330 Å	[112]	(a/2)[111]	1230	0.9-1.2
3 ^a	Channel-Channel	(101)	3300 Å	1290	3000 Å	[121]	(a/2)[111]	1110	0.9
5 ^a	Channel-Helix	(011)	4500 Å	1930	9500 Å	[211]	(a/2)[111]	3520	1.8
5 ^a	Channel-Boundary	(011)	4500 Å	1930	6710 Å	[211]	(a/2)[111]	2490	1.3
6 ^b	Channel Boundary	(011)	3960 Å	1600	12,080 Å	[421]	(a/2)[111]	4790	3.0
Not Discussed ^a	Channel-Boundary	(110)	5550 Å	2380	5940 Å	[112]	(a/2)[111]	2200	0.9

a. Plane of view: (111)

b. Plane of view: (112)

c. As observed in plane of view

dislocations packets should not exceed the diameter of the clusters which corresponds to about 35 planes.

The precise mechanism of the removal of the defect clusters by the slip dislocations is still a matter of conjecture. One possibility is that the motion of the channeling dislocation produces an input of energy that is largely converted into heat. This "heat of plastic deformation" will cause an increase in temperature possibly capable of annealing the radiation-produced damage clusters. Each $(a/2) \langle 111 \rangle$ dislocation produces a shear displacement of $b = \sqrt{3} a/2$ and the $\{110\}$ spacing is $\sqrt{2} a/2$, where a is the lattice parameter. Therefore, each dislocation gives rise to a shear strain of $\sqrt{3}/\sqrt{2}$. If, from Table 11.12 we consider about 2 dislocations per plane to operate in the channeling shear, the shear strain becomes $2\sqrt{3}/\sqrt{2} = 2.5$. This shear occurs at a shear stress level corresponding to the yield stress, or about $\tau = 7.1 \text{ kg/mm}^2$. The energy of plastic deformation per unit volume is, therefore,

$$E_{PD} = 18 \text{ kg/mm}^2 = 42 \text{ cal/cm}^3.$$

Niobium has a specific heat at room temperature of about $0.06 \text{ cal/g-deg} = 0.54 \text{ cal/cm}^3\text{-deg}$. Therefore, under adiabatic conditions if all the energy of plastic deformation were converted into heat, the temperature rise would be about 80°C . This would appear to be an upper estimate, since some heat diffuses away from the incipient channel and some energy is stored in dislocations and defects produced during the deformation. On the other hand, the above simple calculation assumes that the energy of plastic deformation is distributed equally among all the atoms in the channel. It is quite possible that, since the dislocations are impeded by the defect clusters, the energy is expended in localized bursts at the defect clusters. This would cause a large local increase in temperature of the defect cluster, possibly for a sufficiently long period to permit the clusters to anneal.

A second mechanism is the chopping up of damage clusters as slip dislocations pierce through them.

An analysis of the size distribution of damage clusters in as-irradiated niobium (Figure 11.25 in Reference 25) showed the distribution to be sharply peaked at a cluster size of 80 Å. The number of clusters at 60 Å was about 6% of the number at the peak size, and at 40 Å the number was negligibly small. This suggests that a reduction in the effective dimension of the cluster may cause it to become unstable.

A third approach to an understanding of how the defect clusters are removed in the channeling process is the sweeping up of the clusters by the slip dislocations. It is clear that in a process of this type the clusters must somehow be incorporated into the slip dislocations and not simply swept aside. This is suggested by the fact that the concentration of clusters is not higher at the walls of the channels.

Finally, it is possible that the defect clusters are annihilated by virtue of the production of anti-defects during the motion of the channeling dislocations. The damage clusters in as-irradiated niobium have been found to be mostly of interstitial type, and thus vacancies created by the nonconservative motion of jogs on the channeling dislocations may annihilate the clusters. Earlier work has shown (Figure 11.27 in Reference 25) that small defects are observed in irradiated niobium annealed to 600 °C, which may be due to the aggregation of radiation-produced vacancies. In the as-irradiated niobium, these vacancies

dispersed as isolated defects or small-number aggregates. It may be that the slip dislocations tend to drag vacancies to the interstitial clusters where mutual annihilation takes place. In another section of this report,* evidence is presented for the absence of channeling in the radiation-anneal-hardened niobium (i.e., irradiated niobium annealed at about 400 °C which exhibits a further increase in yield stress; see Figure 11.23 in Reference 25). The radiation-anneal-hardening may be due to the trapping of interstitial impurities at radiation-produced vacancies. The trapping of oxygen in irradiated and annealed niobium has been revealed by internal friction and resistivity measurements by Williams, Brundage, and Stanley.⁽¹⁰⁾ Thus, the absence of channeling in radiation-anneal-hardened niobium may be an indication that the radiation-produced vacancies are associated with interstitial impurities and are therefore not available for the role of anti-defects for the interstitial-type defect clusters.

The above discussion has focussed attention on the mechanism of the removal of defect clusters from the cleared channels. A related question is how the dislocation channeling process is initiated. Figures 11.10 and 11.11 illustrate that the channels are often widely and regularly spaced, suggesting that the presence

* S. M. Ohr and R. P. Tucker, "Radiation Anneal Hardening in Niobium," this report.

of one channel may trigger the formation of another one. As pointed out by Sharp,⁽⁴¹⁾ this is evidence for a long-range internal stress in the irradiated-and-deformed material. Thus, it seems likely that a group of dislocation channels, corresponding to a coarse slip band on the surface, is initiated at a single point of stress concentration, activating the formation of one dislocation channel which rapidly produces others in its neighborhood. The dislocation tangles at the intersection of channels or where a channel crosses another microstructural feature may well serve as concentrations of stress from which additional channels emanate. An example of the first of these is seen in Figure 11.12, where Channel 3 appears to be generated by the tangle at the intersection of Channels 1 and 2a; the second is illustrated in Figure 11.15 where a channel appears to be generated at the intersection of another channel and a low-angle boundary. Also, a number of narrow channels apparently in an early state of development is seen at B in Figure 11.11a. These appear to have originated at small dislocation tangles in the channel segment above T'_1 to the left of B.

The phenomenon of dislocation channeling probably plays a central role in the plastic deformation of irradiated metals. The yield stress is likely governed by the stress to initiate the formation of dislocation channels, and the work-hardening behavior depends upon the manner in

which strain is concentrated in the channels and in which the channels propagate to encompass the entire gage section of the sample.

THE TEMPERATURE DEPENDENCE OF RADIATION HARDENING

S. M. Ohr

In formulating a theory of dispersed barrier hardening, Fleischer^(51,52) considered the interaction between a straight dislocation and a center of tetragonal distortion. He derived a force-distance relationship by assuming that the stress from the dislocation is uniform over the volume of the defect. However, to predict the temperature dependence of the yield stress in analytical form, a modified force function was adopted which exhibits substantial deviation from the expression originating directly from the dislocation-defect interaction model. The modified force function predicts that the square root of the yield stress, τ , is directly proportional to the square root of the absolute test temperature, T . Despite the lack of the exactness in the Fleischer theory as discussed recently by Ono,⁽⁵³⁾ temperature dependence of this type has been observed for hardening due to impurities, quenching, and irradiation in crystals of various structures. This report discusses on the basis of continuum theory the applicability of the linear relationship between $\tau^{1/2}$ and $T^{1/2}$ for contact interaction

between dislocation loops and straight dislocations.

In the continuum theory of defect interaction, the interaction energy is obtained as the work required to form a defect in the presence of the stress field of another defect. On the basis of isotropic elasticity theory, Kroupa⁽⁵⁴⁾ has calculated the interaction energy between a straight dislocation and a dislocation loop. The expression for the interaction energy is given by

$$E_{\text{int.}} = b^{\text{OG}} \iint_A \sigma_{13} dA + b^{\text{OP}} \iint_A \sigma_{33} dA, \quad (1)$$

where b^{OG} and b^{OP} are the glide and prismatic components, respectively, of the loop Burgers vector and σ_{ij} is the stress tensor component for the straight dislocation. The integrals are performed over the area of the circular loop with radius R . The retarding force is obtained through $\vec{F} = -\nabla E_{\text{int.}}$.

In moving the dislocation over the loop, a portion of the work is done by the applied stress which exerts a force on the dislocation $F_a = \tau b \ell$, where ℓ is the length of dislocation segment involved in the process. The remaining work in forcing the dislocation toward the loop is supplied by thermal fluctuations. The activation free energy for surmounting the loop as a barrier is obtained from

$$\Delta G = \int_{r_m}^{r_a} (F_r - F_a) dr, \quad (2)$$

where F_r is the radial component

of the retarding force, r_a is the distance from the center of the loop to the dislocation where the applied force is equal to the retarding force, and r_m is the value of r where the retarding force is maximum.

The interaction energy has been calculated for various relative orientations between the dislocation and the loop exactly from Equation (1).⁽⁵⁴⁾ The results indicate that the force-distance curves exhibit a sharp discontinuity at $r = R$. The force on the dislocation is found to be independent of r when the dislocation is inside the loop. Outside the loop, the force is in general a complex function of r , and hence a simple temperature dependence of hardening cannot be obtained. However, when the dislocation is normal to the loop the interaction obeys the inverse square force law, i.e.,

$$\begin{aligned} F_r &= F_m R^2/r^2, \text{ for } r \geq R \\ &= -F_m, \text{ for } r \leq R \end{aligned} \quad (3)$$

where F_m is the maximum retarding force and is given by $f_1(\theta)\mu b^{\text{OG}}/2$ for a screw dislocation and by $f_2(\theta)\mu v b^{\text{OP}}/(1-v)$ for an edge dislocation. The function $f(\theta)$ represents the angular dependence of the retarding force, while b is the Burgers vector of the straight dislocation, μ is the shear modulus, and v is Poisson's ratio. Such a force-distance curve is shown schematically in Figure 11.17, where the shaded area corresponds

ORNL-DWG 68-7472

FIGURE 11.17. Schematic Force-Distance Curve for a Circular Dislocation Loop and a Straight Dislocation that Lies Normal to the Loop. (There is a sharp discontinuity at $r = R$, i.e., when the dislocation comes in contact with the loop. The shaded area corresponds to the activation energy of the process.)

to the activation free energy of the process as defined by Equation (2). F_m occurs where the dislocation comes in contact with the loop, i.e., $r_m = R$. When the dislocation is parallel to the loop, F_m is infinite so that the process of overcoming the loop cannot take place by thermal activation.

The interaction energy has also been calculated for the approximation of an infinitesimal loop.^(51,54) In this approximation, the stress field of the dislocation is assumed not to vary appreciably over the area of the loop and the integrals in Equ-

ation (1) are replaced by the area of the loop times the values of σ_{ij} at the center of the loop. The approximation is generally considered to apply when the dislocation is at distances greater than $2R$ from the center of the loop. In this case, the inverse square force law is always found, since $E_{int.}$ is directly proportional to σ_{ij} and σ_{ij} is always proportional to $1/r$. In view of the sharp discontinuity found for the exact calculation discussed above, the force-distance curves obtained using the infinitesimal loop approximation are cutoff at $r = R$.

In cases where the inverse square force law is obeyed, the activation free energy is found from Equations (2) and (3) as

$$\Delta G = F_m R \left[1 - (\tau/\tau_0)^{1/2} \right]^2, \quad (4)$$

where $\tau_0 = F_m/b\ell$ is the applied stress needed to overcome the barrier in the absence of thermal fluctuation. From the rate equation, $\dot{\epsilon} = \dot{\epsilon}_0 \exp(-\Delta G/kT)$, where $\dot{\epsilon}$ is the strain rate and $\dot{\epsilon}_0$ is the frequency factor, the activation free energy may be set equal to a multiple of kT ,

$$\Delta G = \alpha kT, \quad (5)$$

since $\alpha = \ln(\dot{\epsilon}_0/\dot{\epsilon})$ may be assumed to be constant in a given test. From Equations (4) and (5) we obtain

$$(\tau/\tau_0)^{1/2} = 1 - (T/T_0)^{1/2}, \quad (6)$$

where $T_0 = F_m R / \alpha k$ is the critical temperature above which thermal activation alone is sufficient to move the dislocation past the loop. Thus, the linear relationship between $\tau^{1/2}$ and $T^{1/2}$ is obtained directly from the theory of defect interaction. The difference between the present result and that of Fleischer is in the expressions for the activation free energy and T_0 , where the Burgers vector in Fleischer's theory is here replaced by the loop radius.

In conclusion, the continuum theory of defect interaction does not in general predict a simple temperature dependence of yield stress. However, in cases where the inter-

action is governed by the inverse square force law, the linear relationship between $\tau^{1/2}$ and $T^{1/2}$ is expected.

METALS AND CERAMICS DIVISION

IRRADIATION DAMAGE TO TYPE 316 AND TITANIUM-MODIFIED TYPE 316 SS

E. E. Bloom and J. R. Weir

Type 316 and 316L SS are potential cladding materials for liquid-metal cooled fast-breeder reactors and, as such, it is necessary to gain an understanding of the effects of irradiation on the tensile and creep-rupture properties. The alloys are similar in composition to the most commonly used Type 304 SS alloy (18 wt% Cr, 10 wt% Ni), with the exception that they contain approximately 2.0 to 3.0 wt% Mo. The addition of molybdenum produces an improvement in the elevated-temperature creep-rupture properties. Barnby⁽⁵⁵⁾ has attributed the improved strength to the precipitation of small $(Fe, Cr)_{23}C_6$ particles on dislocations during creep and has shown that creep strength was decreased by aging treatments which removed carbon from solution prior to testing. Our investigation of the effects of neutron irradiation on the mechanical properties involves tensile and creep-rupture tests on specimens in three conditions: 1) annealed, 2) annealed and irradiated, and 3) annealed and aged at the irradiation temperature for the irradiation time. Variables under

investigation include: 1) preirradiation heat treatment, 2) irradiation temperature and 3) postirradiation test conditions, such as test temperature and strain rate.

A second area of this program is an attempt to improve the mechanical properties of an alloy having the basic composition of Type 316 SS by the addition of small amounts of titanium. Initially, this involves an investigation of the effects of up to 0.5 wt% titanium on the tensile and creep-rupture properties in both the unirradiated and irradiated conditions.

Results on Standard Type 316 and 316L SS

The chemical composition of the two heats of material presently being investigated are listed in Table 11.13. Results of tensile tests on standard Type 316 SS were previously reported.⁽⁵⁶⁾ For specimens irradiated to approximately 9×10^{20} n/cm² (thermal) and 7×10^{20} n/cm² ($E > 1$ MeV) at tem-

peratures between 550 and 750 °C and tested at 550, 650, and 750 °C, there was a significant decrease in ductility. The decrease in tensile ductility was independent of irradiation temperature over the range investigated.

A summary of the 650 °C creep-rupture data for standard Type 316 and 316L SS is listed in Table 11.14. An examination of the data indicates that for Type 316 SS there is a reduction in rupture life and ductility due to irradiation and that the changes in properties are sensitive to the irradiation temperature. For the specimens irradiated at 550 °C and tested at 650 °C, there appears to be no effect on the creep rate, but a large reduction in ductility and thus rupture life. An exception is the specimen which was postirradiation tested at 30,000 psi. In this case, the rupture life was 15 hr as compared to 180 hr for the specimen tested in the annealed condition and 437 hr for the thermal control specimen. It is possible that this specimen failed before second stage creep was established and thus the measured creep rate is not a true second stage creep rate. Note that the creep rate of the specimen aged 2000 hr at 550 °C is essentially the same as that of the specimen which was tested in the annealed condition, thus indicating that thermal aging effects are negligible. As the irradiation temperature is increased to 650 or 750 °C, there is an increase in the postirradiation creep rate and ductility. In this case, however, it is observed

TABLE 11.13. Chemical Composition of Type 316 and 316L SS

Element	Composition, wt%	
	316	316L
C	0.068	0.02
Cr	17.2	16.9
Ni	12.5	12.7
Mo	2.06	2.7
Mg	1.9	2.3
Si	0.5	0.58
P	0.039	0.013
S	0.004	0.015
B	0.0005	---

TABLE 11.14. Creep-Rupture Properties of Type 316 and 316L SS at 650 °C

Heat Treatment and/ or Irradiation Conditions	Stress, psi	Rupture Life, hr	Creep Rate, hr	Total Elongation, %	Reduction in Area, %
<u>Type 316 SS</u>					
Annealed 1 hr at 1050 °C	40,000	28	0.35	36.2	39.2
	35,000	91	0.14	29.6	50.4
	32,500	117	0.029	25.1	44.9
	30,000	180	0.024	54.5	32.9
	27,500	1732	0.0046	31.4	52.6
	25,000	1311	0.0069	30.9	57.8
Annealed 1 hr at 1050 °C, irradiated ~2000 hr at 550 °C	30,000	15	0.085	3.2	9.0
	25,000	252	~0.005	2.7	14.3
	20,000	5127	~0.00012	8.7	13.4
Annealed 1 hr at 1050 °C, plus 2000 hr at 550 °C	30,000	437	0.030	33.0	62.0
	25,000	2305	0.0017	29.2	55.2
Annealed 1 hr at 1050 °C, irradiated 2000 hr at 650 °C	35,000	25	0.58	26.7	32.9
	30,000	199	0.032	13.8	26.4
Annealed 1 hr at 1050 °C plus 2000 hr at 650 °C	35,000	20	1.13	52.2	71.0
	30,000	146	0.14	37.5	75.2
Annealed 1 hr at 1050 °C, irradiated ~2000 hr at 750 °C	25,000	92	0.12	14.8	17.4
	20,000	137	0.064	9.3	6.7
	20,000	524	0.024	16.7	22.1
<u>Type 316L SS</u>					
Annealed 1 hr at 1050 °C, irradiated ~2000 hr at 650 °C	30,000	14	0.13	2.4	11.6
	25,000	216	0.037	9.5	12.9
	22,500	48	0.027	2.5	5.8
	20,000	370	0.009	5.4	8.4

that there is also an increase in the creep rate of thermal control specimens which indicates that thermal aging phenomena are occurring.

Stress-rupture results for Type 316L SS irradiated and tested at 650 °C are also listed in Table 11.14. It appears that the 0.02 wt% carbon alloy is significantly weaker, both in terms of rupture life and creep rate, than the 0.068 wt% carbon alloy. Since a boron analysis has not been performed on the low-

carbon alloy, a direct comparison of ductility values is difficult.

Results on Titanium-Modified Type 316 and 316L SS

Specimens of several vacuum-induction melted heats of Type 316 and 316L SS containing various amounts of titanium (up to 0.5 wt%) have been irradiated at 650 °C to approximately 9×10^{20} n/cm² (thermal) and 7×10^{20} n/cm²

($E > 1$ MeV). Boron content ranged between 4 and 10 ppm and the carbon content was approximately 0.02 for the low-carbon series and 0.05 for the high-carbon series. The effect of titanium on the stress-rupture ductility at 650 °C and 30,000 psi stress is shown in Figure 11.18. For each carbon content there is a significant improvement in the ductility of both thermal control and irradiated specimens for all titanium contents. In the case of the 0.05 wt% carbon series, the highest ductility occurred at 0.23 wt% titanium. However, since no alloy containing between 0.05 and 0.15 wt% titanium was available, it is not certain that maximum ductility does not occur at a somewhat lower titanium level. Results on the 0.02 wt% carbon series are not complete in that specimens containing 0.45 wt% titanium remain to be tested. All of the low-carbon alloys containing between 0.1 and 0.3 wt% titanium have exhibited postirradiation ductilities in excess of 30% elongation.

Metallographic examination of tested specimens is presently underway. Figure 11.19 shows the fracture which occurred in Type 316 SS which was annealed 1 hr at 1050 °C and tested at 650 °C and 30,000 psi. The fracture is predominantly intergranular and large voids or cracks have formed on grain boundaries normal to the applied load. In comparison, Figure 11.20 shows the fracture which occurred in Type 316 plus 0.23 wt% titanium SS after annealing at 1050 °C and testing at 650 °C and 35,000 psi. In this case,

the fracture is a transgranular shear fracture and there was essentially no void formation during the test. Similar observations have been made in titanium-modified Type 304 SS and possible mechanism for the change in fracture mode have been discussed. (Reference 56, pp. 22-24)

MECHANICAL PROPERTIES OF TITANIUM-MODIFIED TYPE 304 SS

E. E. Bloom and J. R. Weir, Jr.

Evaluation of the mechanical properties to two 1500 lb vacuum-induction melted heats of titanium-modified Type 304 and 304L SS is continuing. The compositions of these two alloys are listed in Table 11.15.

Specimens of Heat No. 60508 which is Type 304 SS containing 0.2 wt% titanium have been irradiated for approximately 1000 hr at 650 °C. Neutron fluences for this experiment were approximately 2.6×10^{20} n/cm² (thermal) and 2×10^{19} n/cm² ($E > 1$ MeV). On the basis of the initial boron content (0.0027%), it is calculated that approximately 19×10^{-6} atom fraction helium were produced during the irradiation. Figure 11.21 shows the effect of this irradiation on the stress-rupture properties at 704 °C. The results available at this time indicate that in the unirradiated condition the rupture life of specimens annealed 1 hr at 925 °C is approximately a factor of 8 lower than the rupture life of specimens annealed 1 hr at 1040 °C. For both annealing treatments, there is a

- △ ANNEALED 1 HR AT 1050° C, AGED 2000 HRS AT 650° C
 ▲ ANNEALED 1 HR AT 1050° C, AND IRRADIATED APPROXIMATELY
 2000 HRS AT 650° C 9×10^{20} NEUTRONS/CM² (TH) AND 7×10^{20} NEUTRONS/CM²
 (E > 1 Mev)

FIGURE 11.18. Effect of Titanium on the Creep-Rupture Ductility of Type 316 SS Tested at 650 °C and 30,000 psi Stress

FIGURE 11.19. Fracture of Type 316 SS Annealed 1 hr at 1050 °C and Tested at 650 °C and 30,000 psi; Rupture Life 91 hr, Elongation 29.6%

FIGURE 11.20. Fracture of Type 316 SS Plus 0.23 wt% Ti SS Annealed 1 hr at 1050 °C and Tested at 650 °C and 35,000 psi; Rupture Life 582 hr, Elongation 47.0%

TABLE 11.15. Composition of Titanium-Modified Type 304 SS

Element	Composition, wt%	
	316	316L
C	0.012	0.05
Cr	19.5	19.0
Ni	11.2	10.0
Ti	0.19	0.21
Mn	1.0	0.65
Si	0.05	0.8
P	0.006	0.013
S	0.013	0.002
B	0.0003	0.0027
N		
Total	0.006	0.009
Soluble	--	0.003

reduction in rupture life due to irradiation. For the 1040 °C anneal, the rupture life appears to be reduced by approximately a factor of 4 to 5. For the 925 °C anneal, the reduction in rupture life is less than a factor of 2. Elongation as a function of rupture life is shown in Figure 11.22. In both the annealed

and irradiated condition, the ductilities are independent of annealing temperature. Irradiation causes a large reduction in ductility; however, the ductility is approximately constant at 9 to 12% for rupture lives between 10 and 1500 hr.

ELECTRON MICROSCOPE EVALUATION OF TITANIUM-MODIFIED TYPE 304L SS

G. T. Newman

A Type 304L SS with improved resistance to irradiation embrittlement has been developed by making small additions of titanium to the standard alloy. Optimum improvement in ductility has been recorded with alloys containing 0.2 wt% titanium, solution treated at 925 °C for 1 hr. The role of titanium in improving the postirradiation ductility is being studied by electron microscopy. The solution treatment resulted in a fine matrix precipitation, in contrast to the grain boundary precipitation obtained at higher temperatures

FIGURE 11.21. Effect of Irradiation on the Stress-Rupture Properties of a 1500 lb Vacuum-Induction Melted Heat of Type 304 + 0.2 wt% Ti SS at 704 °C

FIGURE 11.22. Effect of Irradiation on the Ductility of Type 304 + 0.2 wt% Ti SS at 704 °C

(> 1000 °C). Larger angular precipitates ($1/2$ to $1\ \mu$) were also present and were assumed to be the Ti(C, N) complexes observed as a yellow phase by optical metallography. Aging studies showed that the density of the fine precipitate increased with increase in aging time, but the size of the individual precipitates ($\sim 500\ \text{\AA}$ after aging 100 hr at 700 °C) appeared relatively stable. Electron microprobe analysis of the larger precipitates obtained by carbon extraction techniques revealed evidence of sulfur as well as titanium (carbon and nitrogen cannot be detected by this microprobe) in alloys aged at 850 °C and higher, suggesting that resolution of the precipitate and segregation of trace impurities may be occurring.

Carbon extraction techniques are also being applied to a series of alloys containing nominally 0.1, 0.2, 0.3, 0.4, and 0.5 wt% titanium. To date, work on the alloy containing 0.2 wt% titanium has revealed precipitates rich in titanium, titanium plus aluminum, and chromium plus iron. Electron diffraction data on the latter particles (0.2 to $0.4\ \mu$ in size) do not identify with that of the most frequently observed carbide in austenitic stainless steels (Fe, Cr)₂₃C₆.

DRAWING OF STAINLESS STEEL TUBING

G. A. Reimann

Work has continued on the investigation of the production of tubing from titanium-modified grades of Types 304 and 304L SS. Tubing has

been fabricated from two heats (Heat Nos. 3756 and 60508) of the modified stainless steel.

The primary and secondary breakdown techniques have been described⁽⁵⁷⁾ and any subsequent variations applied to the mandrel drawing approach have been limited to the amount of reduction per pass, the amount of reduction between anneals, and the substitution of plug drawing for the final pass.

Approximately 150 ft of 0.250 in. OD x 0.016 in. wall tubing from Heat No. 3756 has been submitted for non-destructive testing and property evaluation. About half of this tubing was subjected to a drawing schedule with two passes (25 to 29% reduction) between anneals, and the balance was drawn with anneals between each pass (14 to 16% reduction). Annealing conditions were $1/3$ hr at 1200 °C. No significant differences have been observed which may be attributed to the drawing schedule.

The drawing schedule has been modified so that the finished tubing contained 5% cold work after a 1 hr anneal at 925 °C prior to the last pass. The tubing is rather soft at this level of cold work and must be handled carefully to avoid bending and denting. If the tube emerges from the final pass with a slight bend, it is difficult to straighten on existing equipment without losing control of the diameter dimensions. Even though plug drawing is used on the last pass to minimize this problem, a small amount of spiral

from reeling the previous pass from the mandrel is still evident.

Several tube shells from Latrobe Heat No. 60508 are in process for conversion into 0.250 in. OD x 0.016 in. wall tubing. This heat differs from Heat No. 3756 in several respects. The chemistry is compared in Table 11.15 and the microstructure is shown in Figure 11.23. Attempts were made to dissolve the second phase by heating in hydrogen for 16 hr at 1200 °C, but at least 25% of the second phase remained after this treatment (Figure 11.23). Drawing the tubing from the as-extruded condition without attempting to remove the second phase produced no adverse effects and the second phase disappeared after an equivalent of 6 hr at 1200 °C during drawing schedule.

Presently, we have drawn this heat down to 0.387 in. OD x 0.019 in. wall tubes.

Several more passes remain before the final size is achieved; however, this heat of tubing has shown itself to be more susceptible to longitudinal cracking than Heat No. 3756. The grain size of Heat No. 60508 is somewhat smaller than Heat No. 3756 and more nitride particles are present in the Latrobe heat (Heat No. 60508). The longitudinal cracking has been confined to the front of the tubes where swaging is done repeatedly for pointing.

THE EFFECT OF CYCLOTRON-INJECTED
HELIUM ON THE CREEP-RUPTURE PROP-
ERTIES OF TYPE 304 AND 0.2%
TITANIUM-TYPE 304L SS

R. T. King and J. R. Weir, Jr.

The effect of cyclotron-injected helium on the tensile properties of AISI Type 304 SS has previously been shown to be qualitatively similar to that of helium produced by $^{10}\text{B}(n,\alpha)^7\text{Li}$ reactions.^(58,59) It was also shown that a 0.2% titanium-modified Type 304L SS, which is resistant to radiation-induced high-temperature loss of ductility,⁽⁶⁰⁾ is not as severely affected by cyclotron-injected helium as the Type 304 SS alloy. However, the elevated-temperature mechanical properties of these materials are sensitive to the rate of deformation, and hence it is necessary to know the effects of helium on the creep-rupture properties of these materials. We have, therefore, injected 2.0×10^{-5} atom fraction of helium into sets of specimens of both materials for creep-rupture testing.

Both materials were finish cold-rolled 50% to a 0.010 in. thickness; specimens were machined having a 0.25 in. gage width and a 0.75 in. gage length, and then annealed for 1 hr at either 1038 or 925 °C. The grain sizes produced in the Type 304 SS were ASTM 5 and 6, and in the

As-Extruded *After 16hr at 1200 °C*

FIGURE 11.23. Microstructures of Heat No. 60508, As-Extruded, Comparing Structure Before and After 16 hr at 1200 °C Heat Treatment

0.2% titanium-type 304L SS the grain sizes were ASTM 7 and 8 to 8-1/2, respectively. Specimens of both materials, in both annealed conditions, were then bombarded with α -particles at less than 200 °C to contain a uniform concentration of 2.0×10^{-5} atom fraction of helium.

Stress-rupture tests were then performed under 10 psig of static argon at 600 and 700 °C. The stresses were selected to yield rupture times ranging from approximately 1 to 1000 hr. The results of these tests are shown as graphs of strain versus rupture life, and the stresses at which individual tests were run are indicated on the plots (Figures 11.24 through 11.27).

At 600 °C, the Type 304 SS control specimens annealed at 925 °C have consistently higher ductility than those annealed at 1038 °C, although the different annealing temperatures do not cause much difference in rupture life (Figure 11.24). At 700 °C, the Type 304 SS control specimens annealed at 925 °C have consistently lower rupture life, but higher ductility than those annealed at 1038 °C (Figure 11.25).*

The cyclotron-injected 2.0×10^{-5} atom fraction of helium causes substantial reductions in both the rupture life and ductility of Type 304 SS (Figures 11.24, 11.25). The loss of ductility is generally greatest at 700 °C, since the tendency for

intergranular fracture increases with increasing temperature.

The behavior of the control specimens of the 0.2% Ti-Type 304L SS is qualitatively similar to the Type 304 SS, although at high strain rates there is little effect of annealing on rupture life at 700 °C (Figures 11.26, 11.27). The rupture life of the 0.2% Ti-Type 304L SS alloy is consistently lower than that of the Type 304 SS. This may be explained on the basis of reduced solid solution strengthening and carbide precipitate formation in the 0.2% Ti-Type 304L SS, which causes more rapid second-stage creep rates and hence shorter rupture life.

In contrast to the behavior of the Type 304 SS, the rupture life of the titanium-modified alloy is not significantly reduced by 2.0×10^{-5} atom fraction of helium at either test temperature. Furthermore, the ductility of the titanium-modified alloy tested at high strain rates at 600 °C is not altered by this amount of helium; however, at the slower strain rates the ductility is reduced, although not as greatly as that of Type 304 SS. At 700 °C, the ductility loss due to helium is greater than at 600 °C, but the ductility of the modified alloy annealed for 1 hr at 925 °C (~ 30%) remains significantly higher than that of Type 304 SS, and does not vary greatly with strain rate in tests lasting up to 100 hr.

Electron microscopy and optical metallographic studies are in progress to aid in interpreting these results. We are also continuing to

* With the exception of specimens tested at 600 °C after annealing at 925 °C, the strain-to-rupture tends to decrease with decreasing strain rate, or applied stress.

FIGURE 11.24. Strain Versus Rupture Time of Type 304 SS at 600 °C

FIGURE 11.25. Strain Versus Rupture Time of Type 304 SS at 700 °C

FIGURE 11.26. Strain Versus Rupture Time of 0.2% Ti-Type 304L SS at 600 °C

FIGURE 11.27. Strain Versus Rupture Time of 0.2% Ti-Type 304L SS at 700 °C

test cyclotron-bombarded specimens at lower stresses, to obtain more information on the behavior of these materials at rupture times approaching 1000 hr.

ELECTRON MICROSCOPY OF IRRADIATED EBR-II FUEL CLADDING

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

During the past quarter, we concluded our study of the effects of irradiation temperature and fluence on the void population in a fuel pin irradiated in EBR-II and also examined the annealing behavior of the various void distributions present.

Irradiation temperatures varied continuously along the pin from the inlet coolant temperature of 370 °C to a maximum temperature of 472 °C, and the fast neutron fluence ranged from a maximum of 1.4×10^{22} n/cm² at the center of the pin to a minimum of 0.8×10^{22} n/cm² at its extremes. Sections were taken at intervals of 3 in. along the length of the cladding which allowed us to examine the influence of irradiation temperature and fluence on the microstructure.

The preliminary examination showed that the void density decreased and the average void size increased irradiation temperature. Irradiation conditions, measured void densities and calculated volume changes based on experimentally determined void distributions are listed in Table 11.16. For the conditions examined here, we see that the increased growth at higher temperatures compensates for the reduced void den-

sity to render the overall swelling relatively weakly dependent on irradiation temperature.

Although the voids formed at the higher irradiation temperatures had crystallographic shapes, a variety of forms occurred within a single grain. We believe that complex shapes result from collisions of migrating voids and that surface diffusion reduces these to crystallographic shapes, but not all to the lowest energy octahedral form.

Precipitate particles, presumably carbides, on the order of 0.1 μ in diameter were dispersed along grain boundaries of the specimen irradiated at 472 °C. Dislocation loops persisted up to the boundary, but a region about 500 Å wide adjacent to the boundary was denuded of voids. The denuded zone decreased with decreasing temperature and at 438 °C was only about 200 Å thick. No grain boundary precipitate particles were observed. The denuded zone increased with a further decrease in irradiation temperature, reaching a value of about 1500 Å at 370 °C, as is shown in Figure 11.28. Again, no precipitate particles were present in the boundary, but the dislocation loop distribution showed peculiar behavior in the vicinity of the boundary as can also be seen in Figure 11.28. The area between about 750 and 1500 Å from the grain boundary was denuded of loops, but within 750 Å a very high density of loops was present. We suspect that impurities play an important part in nucleating the loops.

TABLE 11.16. Irradiation Conditions and Void Density Measurements in Type 304L SS

Specimen Number	Irradiation Temperature, °C	Fast Neutron Fluence, $n/cm^2 \times 10^{22}$	Voids/ $cm^3 \times 10^{15}$	% $\Delta V/V$
1	370	0.8	1.4	0.07
2	398	1.2	1.3	0.15
3	430	1.4	1.3	0.17
4	460	1.3	0.9	0.16
5	472	0.9	0.4	0.08

It is not readily apparent from Figure 11.28 that the defects do exist as loops because of overlap of their images. However, but by tilting the foil to reduce contrast, loops are clearly evident ranging up to 500 Å in diameter.

Annealing of Defects

The annealing of the irradiation damage in Specimen No. 3 (irradiated at 430 °C to $1.4 \times 10^{22} n/cm^2$) was studied by isochronal anneals of 1 hr at temperatures between 475 and

FIGURE 11.28. Grain Boundary in Type 304L SS Irradiated at 370 °C to $0.8 \times 10^{22} n/cm^2$. The Grain at Left Is Oriented Such that the Region Denuded of Voids Is Visible. The Grain on the Right Is Oriented for Diffraction and Shows a Region Denuded of Dislocation Loops.

900 °C and by isothermal anneals at 700 °C for times between 1 and 100 hr.

Annealing 1 hr at 475 °C resulted in no change in the character of the dislocation loops nor in the size distribution of the observable voids. After 1 hr at 600 °C, the dislocation loops had converted to a dislocation network and the total void density had decreased from 12.7×10^{14} voids/cm³ (as-irradiated) to 9.6×10^{14} voids/cm³. With increasing annealing temperature, there was a continuous decrease in both dislocation and void density as can be seen from the values listed in Table 11.17. Histograms of void density as a function of void size after various anneals are shown in Figure 11.29. The decrease in void density occurs because of the annealing of the smaller voids, those having initial diameters less than approximately 150 Å, while the density of larger voids remains essentially unchanged. Thus, after 1 hr at 800 °C, the only remaining voids had diameters between 150 and 300 Å. After 1 hr at 900 °C, the voids had completely disappeared.

Similar results (i.e., rapid annealing of the smaller voids with little change in the density of larger voids) were obtained from the isothermal anneals.

Specimen No. 1, in the as-irradiated condition, contained voids with diameters between 67 and 187 Å. In this case, all voids were eliminated after annealing 1 hr at 700 °C. In comparison, Specimen No. 5 contained voids having diameters as large as 300 Å and annealing temperature of 800 °C was required for complete elimination of the voids.

The results of the annealing treatments are consistent with the annealing behavior of quenched voids in aluminum reported by Volin and Balluffi.⁽⁶¹⁾ In their work, void dissolution was controlled by the diffusion of vacancies away from the void. They felt that exchange of vacancies between the void and the lattice occurred sufficiently rapidly that the equilibrium vacancy concentration was maintained at the surface of the void at all times. Since this concentration varies exponentially with the reciprocal of the void radius, the dissolution rate increases rapidly as the void size decreases. Consequently, small voids are eliminated quickly, while large ones barely change size at all. As the small voids dissolve, the vacancy concentration in the lattice increases over the equilibrium value and may be sufficient to cause some large voids to grow. The curves presented in Figure 11.29 show that the tendency for this is slight except possibly for the anneal of 1 hr at

TABLE 11.17. Dislocation and Void Densities for Various Postirradiation* Annealing Treatments

Annealing Treatment	Dislocation Density cm/cm ³ x 10 ⁹	Void Density/cm ³ x 10 ¹⁴
1 hr at 600 °C	31	9.6
1 hr at 700 °C	7.6	5.0
1 hr at 800 °C	1.7	0.8
1 hr at 900 °C	<0.1	0
10 hr at 700 °C	2.1	2.3
100 hr at 700 °C	0.8	0.3

* Irradiated at 430 °C to 1.4×10^{22} n/cm²

FIGURE 11.29. The Effects of Annealing on the Void Size Distribution in Type 304L SS

800 °C. The increased lattice concentration of vacancies does slow the dissolution rate of all voids over that predicted by the treatment given by Volin and Balluffi.

The shapes of the void distribution curves for the annealed specimens reflect this process. The density of small voids does not go to zero, however, for larger ones as they shrink must pass through the range of small sizes. The tails on the high size of the distribution are hardly influenced until either high-temperature or long-time annealing treatments are encountered.

The observation that small voids developed in Section 1 annealed at a significantly lower temperature than the larger ones in Section 3 and 5 is in agreement with this picture of the annealing process. It suggests that periodic annealing may provide a means for controlling swell-

ing of reactor components if it is done frequently enough that a population consisting only of small voids is present, otherwise excessively high temperatures will be required to eliminate the voids in reasonable times.

SOME EFFECTS OF GRAIN SIZE ON THE RADIATION DAMAGE OF INCOLOY 800

D. G. Harman

Initial testing of irradiated Incoloy 800 showed the grain size to be an important variable regarding creep ductility. Greater creep strains were noted for the finer grain sizes.

We selected four heats of materials with carbon levels of 0.03 and 0.12 wt% and titanium levels of 0.1 and 0.3 wt% and heat-treated cold-worked material to obtain a range of grain sizes, as is shown in Figure 11.30. Button-head tensile specimens were then irradiated in the ORR at 700 °C 2 to 3×10^{20} n/cm² (thermal), while identical control specimens were held at 700 °C in a laboratory furnace for the duration of the reactor cycle (~1100 hr).

We have evaluated the irradiated specimens at 700 °C: tensile tested at 0.002 and 0.05 min⁻¹ and creep-rupture tested at 10,000 and 12,000 psi. The tensile testing of the control specimens has been completed; however, the corresponding creep-rupture tests are just getting underway and will be reported at a later date.

The tensile results are listed in Table 11.18. We have not completed

FIGURE 11.30. Microstructure of Experimental Incoloy 800. The Four Alloys Were Held for 30 min at Indicated Temperatures to Obtain Desired Grain Size.

TABLE 11.18. Short-Time Tensile Test Results for Incoloy 800 Irradiated and Tested at 700 °C*

Anneal Temp, °C	Strain Rate, min ⁻¹	0.2% Yield Strength, 10 ³ psi		Ultimate Tensile Strength, 10 ³ psi		Elongation, %			
		Control	Irrad.	Control	Irrad.	Uniform Control	Irrad.	Total Control	Irrad.
0.1% Ti, 0.03% C									
900	0.05	22.9	23.6	36.4	34.5	19.0	12.4	58.0	22.9
925	0.05	22.3	24.2	37.7	35.9	15.0	11.8	51.0	13.8
950	0.05	21.5	21.8	37.4	35.4	17.0	12.5	59.0	17.8
980	0.05	17.5	19.1	41.1	38.5	17.0	11.9	53.0	14.8
900	0.002	17.3	21.1	24.3	23.4	8.6	2.7	61.0	18.4
925	0.002	17.5	19.4	24.4	23.0	10.8	6.1	64.6	22.0
950	0.002	18.0	20.6	25.3	24.6	9.3	7.2	59.3	19.8
980	0.002	16.0	18.5	27.9	26.9	10.5	7.3	56.7	13.7
1000	0.002	13.9	15.5	28.9	27.5	13.0	7.7	53.5	10.9
0.3% Ti, 0.3% C									
900	0.05	23.0	--	40.0	--	17.0	--	53.0	--
925	0.05	22.8	--	40.7	--	17.0	--	59.0	--
950	0.05	21.8	36.7	42.0	40.6	18.0	9.5	62.0	11.3
980	0.05	17.2	--	43.4	--	18.0	--	47.0	--
900	0.002	19.3	20.8	25.2	24.2	12.5	4.7	68.9	11.9
925	0.002	20.8	21.4	30.6	26.3	12.2	5.4	53.6	10.0
950	0.002	18.3	20.3	27.8	25.6	7.6	5.1	48.1	10.1
980	0.002	15.7	19.5	30.0	28.4	11.7	6.0	50.8	7.7
1000	0.002	15.0	18.2	31.8	29.3	9.5	5.8	52.4	8.1
0.1% Ti, 0.12% C									
975	0.05	29.6	26.7	42.1	35.0	14.8	11.6	58.6	27.5
1000	0.05	30.0	26.1	41.8	35.5	14.7	10.5	55.9	20.0
1025	0.05	28.1	24.6	42.6	35.4	14.0	11.4	44.3	20.3
1040	0.05	27.2	24.0	43.7	37.1	16.0	12.1	58.0	21.3
975	0.002	17.7	19.5	22.8	21.3	8.5	5.4	67.9	38.4
1000	0.002	20.4	18.4	25.6	21.6	7.5	5.6	76.6	31.5
1025	0.002	17.9	19.5	26.5	22.6	7.7	5.9	70.5	24.7
1040	0.002	20.0	19.2	27.5	24.0	8.0	6.9	54.7	26.0
1100	0.002	18.0	18.9	32.3	26.6	10.5	8.4	49.6	20.8
0.3% Ti, 0.12% C									
975	0.05	28.0	29.2	38.1	36.6	12.1	7.4	52.0	13.6
1000	0.05	27.2	28.4	38.7	37.4	13.5	8.1	53.7	13.6
1025	0.05	25.9	27.9	40.0	38.5	15.0	7.9	49.5	11.6
1040	0.05	25.5	27.1	39.9	42.6	15.0	10.9	49.2	13.6
975	0.002	22.8	20.8	27.5	22.3	7.3	4.7	69.7	13.1
1000	0.002	22.5	22.3	28.0	24.1	8.0	4.4	68.4	12.8
1025	0.002	21.1	22.4	29.5	25.1	7.1	5.1	57.9	11.3
1040	0.002	21.4	22.3	29.8	25.1	10.0	5.9	60.2	12.1
1100	0.002	19.9	21.3	35.9	28.7	11.2	6.1	53.8	12.1

* Specimens irradiated to 2 to 3 x 10²⁰ n/cm² (thermal) in the ORR and control specimens held at 700 °C for duration of irradiation.

our evaluation of these data; however, certain trends are evident. All four heats of material show an increasing ultimate tensile strength with increasing grain size (i.e., increasing annealing temperature) for both the irradiated and control specimens and this is more pronounced at the slower strain rate. In most cases, the

0.2% yield strength decreases with increasing grain size.

The uniform elongations for both the irradiated and the control specimens seem to be quite independent of grain size, but are lower for the slower strain rate. Radiation reduced the uniform elongation by about 30% of the control value at the

faster strain rate and by about 50% at the slower strain rate with the 0.3% titanium heats showing slightly higher losses as compared with the 0.1% titanium heats.

Total elongation values of 50 to 70% are noted for the control specimens. Radiation reduced these to 11 to 23% for the two 0.1% titanium alloys and 7 to 14% for the two with 0.3% titanium. The finer grain size material showed the better postirradiation ductility and this was more pronounced for the 0.1% titanium alloys. The high-carbon heats showed less radiation damage to the total tensile elongation, but this was probably due to the finer grain sizes as was shown in Figure 11.30.

Figures 11.31 and 11.32 show the postirradiation creep elongations and rupture times, respectively. As expected from previous results (e.g., Fuels and Materials Quarterly Progress Report ORNL-TM-2217, June 1968.), the 0.1% titanium heats showed much higher creep elongations than did the 0.3% titanium heats. Also apparent from Figure 11.31 is the significant effect of grain size and stress level. Creep elongations as high as 100% were measured for both carbon levels when tested at the lower stress. As can be noted from the figure, this is possible only for the finer grain size material. It is also significant that the creep elongation values are independent of carbon level when the grain size is considered.

As expected, the larger grain size provided longer rupture times as can be seen in Figure 11.32. Also, the

0.1% titanium heats show slightly longer rupture lives than the 0.3% titanium heats with the same carbon content. This is probably a result of the higher ductilities of these alloys. A tendency for longer rupture times is evident for the high carbon as compared with the low-carbon alloys when tested in the large grained condition. Whether or not this is an aging effect will be determined upon completion of the control tests.

This experiment, we feel, shows that one may be able to obtain the desired combination of strength and ductility by carefully controlling the grain size of Incoloy 800 of a particular composition. It should be remembered that these are small vacuum-melted heats of material, but many of our observations should indicate important variables for commercial application.

VOIDS IN EBR-II IRRADIATED VANADIUM

F. W. Wiffen and J. O. Stiegler

Unalloyed vanadium that had been used as cladding in a fuel irradiation experiment was recently obtained from United Nuclear Corporation (UNC). This material was from the cladding of the fueled portion of a (U, PU)C fuel pin irradiated in Row 4 of EBR-II for 2850 hr at power. The cladding was stripped from the fuel by BMI and was received at ORNL in the form of segments of tubing. The original tube had an inner diameter of 0.257 in. and a wall thickness of 0.022 in. Irradiation conditions reported by UNC⁽⁶²⁾ are a calculated

FIGURE 11.31. Postirradiation Creep Elongation of Incoloy 800. Specimens Were Irradiated in the ORR at 700 °C to a Thermal Fluence of 2 to 3×10^{22} n/cm² and Tested in Air at 700 °C

FIGURE 11.32. Postirradiation Creep Rupture of Incoloy 800. Specimens Were Irradiated in the ORR to a Thermal Fluence of 2 to 3 $\times 10^{20}$ n/cm² and Creep Tested in Air at 700 °C

clad surface temperature of 630 °C and peak fluence of 1.7×10^{22} n/cm² ($E > 0$). Postirradiation examination by BMI indicated an increase in the clad outer diameter of 0.4% and a change in the Knoop microhardness from 136 for the unirradiated material to 151 after irradiation. No effect due to the irradiation was seen in metallographic examination. The irradiated vanadium was rated as "moderately ductile" as a result of fuel decladding operations and room temperature bend tests.

Transmission electron microscopy of segments of the cladding revealed a high density of irradiation-produced defects. The most obvious feature, as shown in Figure 11.33, is the uniform distribution of voids. In a typical section of foil, there were 508 countable voids per 10^{-8} cm² of foil area. The size distribution of these voids, classified by diameters measured with a Zeiss Particle Size Analyzer on prints at 200,000 X, is shown in Figure 11.34. The average diameter of the 1700 voids represented in Figure 11.34 is 117 Å. Assuming a value for the foil thickness of 1500 Å gives a total density of 3.4×10^{15} voids/cm³ and a total void volume of 3.7×10^{-3} cm³/cm³ of foil. Some of the larger voids were faceted in a regular manner, but the crystallography of the bounding planes has not yet been determined. A high density of platelets was also observed in the irradiated material. A few of these can be seen in Figure 11.33, but most are not visible under the contrast conditions of this photograph. This system of

precipitate platelets is on planes of the form {110} and has not yet been studied in detail.

COMPARISON OF LABORATORY AND COMMERCIAL HEATS OF TITANIUM- MODIFIED HASTELLOY N

C. E. Sessions and H. E. McCoy

We have found that small additions of titanium significantly improve the mechanical behavior of Hastelloy N under irradiation and are currently evaluating several small heats.

In our development work, we have irradiated both laboratory (2 lb) and commercial heats (100 lb) of Hastelloy N with varying concentrations of titanium and carbon. The alloy base composition is Ni-12% Mo-7% Cr-0.2% Mn. These alloys have been irradiated in several different experiments to thermal neutron fluences of 2 to 5×10^{20} n/cm² using the Oak Ridge Research Reactor (ORR) and the Experiment Test Reactor (ETR). Although several irradiation temperatures have been investigated, we shall consider only the results for specimens irradiated at 500 to 650 °C in order to assess the effects of our titanium modification on high-temperature irradiation damage. We shall compare the behavior of the small laboratory melts with that of larger commercial melts of this material.

Commercial Melts

A partial list of 100-lb commercial heats on which we have accumulated preliminary data is given in

FIGURE 11.33. Transmission Electron Photomicrograph of Vanadium Irradiated in EBR-II to a Fluence of $\sim 1.7 \times 10^{22}$ n/cm² ($E > 0$) at a Temperature Near 630 °C. The Void Population Has an Average Diameter of 117 Å and a Density of 3.4×10^{15} /cm³.

FIGURE 11.34. Void Size Distribution for Vanadium Specimen Show in Figure 11.33.

Table 11.19. The titanium concentrations varies from 0 to 1.2% with only a small variation in carbon content (0.06 to 0.9%). The ranges of stress-rupture properties listed in Table 11.19 were obtained from an approximate fit of a stress-rupture life plot of the few data points that are currently available for each heat. All data considered were for a comparable pretest heat treatment of a 1 hr anneal at 1177 °C in argon, but undoubtedly reflect differences in 1) grain size and 2) thermal aging during the irradiation cycle due to variations in titanium content.

The properties of these same alloys in the unirradiated condition are also given in Table 11.19. The ranges result from uncertainties because of the small number of tests available. The 40,000 psi rupture life at 650 °C for unirradiated material containing zero titanium is

approximately 200 hr and creep-fracture occurs at total strains of between 15 and 25%. A small, but apparently insignificant, increase in both rupture life and fracture strain is found for titanium levels up to 0.5% titanium. A large increase in both strength and ductility is found, however, at the 0.99% titanium level where fracture would be predicted in 1500 hr after 35 to 40% strain.

The postirradiation stress-rupture properties are also given in Table 11.19 and the variation of the rupture life with titanium content is shown in Figure 11.35. These samples were irradiated at temperatures from 500 to 650 °C in a thermal neutron fluence of from 2 to 5 x 10²⁰ n/cm². Again, all data are for the same preirradiation heat treatment of 1 hr at 1177 °C. The postirradiation creep-rupture life at 650 °C and 40,000 psi increased markedly with

TABLE 11.19. Effect of Titanium Additions on Creep Properties of Commercial Heats of Hastelloy N Tested at 650 °C and 40,000 psi

Heat No.	Titanium, wt%	Carbon, wt%	Creep-Rupture Properties			
			Unirradiated		Irradiated	
			Rupture Life, hr	Strain,* %	Rupture Life, hr	Strain,* %
21546	0	0.07	200	15 - 25	10	2.5 - 5
65535	0.15	0.07	250	10 - 22	70	2.5 - 4
66541	0.27	0.08	320	13 - 23	200	2 - 3
21545	0.52	0.06	700	20 - 30	400	3 - 19
66548	0.45	0.06	300	14 - 27	400	4 - 8
67550	0.71	0.08	Test Incomplete		2000	4 - 12
67549	0.99	0.08	1500	35 - 40	2000	2 - 8
67548	1.20	0.09	Test Incomplete		2500	6 - 17

* Range for three different stress levels

FIGURE 11.35. Variation in Stress-Rupture Life with Titanium Content for Irradiated Hastelloy N

titanium level from approximately 5 hr at zero titanium to 3000 hr at 1.2% titanium. At 760 °C where the creep strength is much lower, we also find a beneficial strengthening effect of titanium; the rupture life at 20,000 psi increased from approximately 0.2 hr at zero titanium to 200 hr at 1.2% titanium. At 760 °C, a smaller improvement in rupture life is found for increases in titanium content about 0.2% as compared with the progressive increase in life with titanium content at 650 °C. The postirradiation fracture strain for

the commercial heats varied from 2 to 5% for the heats containing less than 0.27% titanium to 3 to 19% for alloys containing 0.5 to 1.2% titanium.

Laboratory Melts

For comparative purposes, although the data are not tabulated, we observed that 2 lb laboratory heats of unirradiated Hastelloy N containing increasing titanium concentrations at 4,000 psi and 650 °C exhibited rupture lives from 45 to 1050 hr. This

increase in rupture life was progressive with increasing titanium from 0 to 1.0%. The fracture strain for the laboratory melts showed an increase with titanium level from 10 to 15% up to 22 to 27% over the range of titanium investigated.

In small laboratory heats following irradiation, we observed an increase in the rupture life at 40,000 psi and 650 °C from 70 to 1000 hr for titanium levels from 0 to 1.0%, which is somewhat less than that observed for the commercial alloys. We also found a progressive increase in the fracture strain from 2 to 13% for additions of up to 1.0% titanium.

In summary, the observed properties indicate that the commercial heats of titanium-modified Hastelloy N almost exactly parallel the out-of-pile results for laboratory melts. However, a small difference between laboratory and commercial heats resulted for the irradiation condition in that greater strengthening is found with titanium for commercial heats than would have been predicted from the laboratory heat results. Also, the fracture strain following irradiation is slightly less than for the higher purity laboratory heats.

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IRRADIATION DAMAGE TO REACTOR METALS

A. L. Bement

REACTOR METALS RESEARCH

IRRADIATION FACILITIES OPERATION

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, and special testing techniques, applicable to either irradiated or unirradiated specimens, will be developed.

The experiments discharged from the various Engineering Test Reactor (ETR) Core positions for Cycle 95 are given in Table 12.1, along with the estimated neutron fluxes and exposures.

The operating history of the ETR and the G-7 hot water loop for Cycle 94 is given in Table 12.2. Loop

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

Operating History	ETR Cycle Number
1. Reactor History	Cycle 94
Start up ^(a)	2-28-68
End of Cycle	4-1-68
Megawatt Days	5309
Effective Days at 175 MW	30.34
Number of Scrams ^(b)	16
Number of Shutdowns	1
2. G-7 Hot Water Loop History	
Maximum Temperature	526 °F
Effective Days Above 200 °F	31.2
Effective Days at Oper. Temp.	29.5
Operating Efficiency ^(c)	100.3%
Temperature Efficiency ^(d)	94.5%

(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 12.1. Summary of Experiments Discharged at the Conclusion of ETR Cycle 95

Experiment Designation	No. of Specimens	Type of Specimens	Estimated Flux, n/cm ² /sec ^(a)	Estimated Exposure, n/cm ² ^{2(a)}
GEH-20-17	18	Zr-2 Tensiles	10.6×10^{13}	1.97×10^{22}
18	18	Zr-2 Tensiles	11.2×10^{13}	2.08×10^{22}
179	18	Zr-2 Corrosion Blanks	2.0×10^{13}	8.6×10^{20}
290	6	18-18-2 Tensiles	3.7×10^{13}	1.1×10^{20}
291	6	18-18-2 Tensiles	5.5×10^{13}	1.5×10^{20}
285 ^(b)	36	304 Tensile & Creep	10.4×10^{13}	8.5×10^{20}
286 ^(b)	36	316 Tensile & Creep	8.0×10^{13}	6.0×10^{20}

Note: Total specimens discharged, ETR G-7 Loop, Cycle 95 = 138
Total specimens discharged, ETR G-7 Loop, to date = 3524

(a) Neutrons with energies greater than 1 MeV.

(b) Denotes double length quadrant.

operation was normal for this cycle. The history for the out-of-reactor loop is given in Table 12.3.

TABLE 12.3. Summary of Operating History and Discharge Schedule for Out-of-Reactor Loop, ETR Cycle 94

1. TIME-TEMPERATURE SUMMARY

<u>Temperature</u>	<u>Cycle 94 Time, hr</u>
200 °C (392 °F)	9.6
218 °C (424 °F)	7.1
246 °C (475 °F)	26.4
282 °C (540 °F)	<u>707.1</u>
TOTAL TIME	750.2

2. SPECIMEN DISCHARGES: Material, Specimen Type and Number.

JS-700 and 18-18-2 SS Tensiles	24
AM-350 SS Tensiles	<u>18</u>
TOTAL	42

Total Specimens discharged to date = 1503.

Data Processing

Ninety-one tensile data reports have been added to the Radiation Effects on Metals (REM) computer program during the quarter, raising the accumulative total to 5597.

IRRADIATION DAMAGE TO STAINLESS STEEL

A. J. Lovell

The purpose of this program is to determine the combined effects of neutron irradiation and environment on stainless steels. Irradiation-induced mechanical property changes are determined as a function of fast and thermal neutron fluence, temperature, and environment of irradiation and postirradiation test temperature and environments.

Emphases are placed on tensile and creep investigations and on studies of the mechanisms of irradiation-induced property changes.

The tensile properties of annealed and 25% cold-worked, AISI Type 348 specimens irradiated in the ETR G-7 hot water loop to a fast neutron fluence of 8.1×10^{21} ($E > 1$ MeV) have been evaluated. The strength data for tests at RT, 300, 500, 650, and 750 °C are presented in Figure 12.1 for the annealed specimens. The irradiated specimens have increased strength over as-fabricated and out-of-reactor controls for all test temperatures. The ductility, as indicated by total and uniform elongation (Figure 12.2), has been reduced markedly by irradiation. The close approach of the yield strength to the ultimate strength (Figure 12.1) and the low uniform elongation (Figure 12.2) indicate inability of the specimens to work harden and also their inherent instability.

The small difference between uniform and total elongation (Figure 12.2) for the irradiated specimens and the low values is evidence of the gross embrittlement observed at all test temperatures.

Creep-rupture testing of a group of AISI Type 304 specimens irradiated in the ETR hot water loop has been initiated, with one test completed. The results show that for the specimen irradiated at 290 °C in the hot water loop to a fast fluence of about 8×10^{20}

Neg 0682124-1

FIGURE 12.1. The Effect of Test Temperature on the Strength of Irradiated AISI 348 Flat Stainless Steel Specimens in the Annealed Condition.

Neg 0680963-1

FIGURE 12.2. The Effect of Test Temperature on the Ductility of Irradiated AISI 348 Flat Stainless Steel Specimens in the Annealed Condition.

($E > 1$ MeV), the 540 °C (1004 °F) rupture life decreases from 88 to 19.2 hr. The creep-rupture test program is continuing.

Creep-rupture testing on as-fabricated specimens in support of the EBR-II irradiations is in progress. The results for 650 °C (1202 °F) tests on AISI Type 304 SS (program heat No. 55697) in the 1066 °C (1950 °F)/1-hour, solution-treated condition are shown in Table 12.4. The creep strength of this heat of material in the 1066 °C

(1950 °F)/1-hour, solution-treated condition is below the average of 304 heats reported previously.⁽¹⁾

TABLE 12.4. Creep-Rupture Data for Type 304 SS Tested at 650 °C in Air

Specimen No.	Stress, psi	Rupture Life, hr	Minimum Creep Rate in./in./hr	Total Elongation, %	Reduction of Area, %
M-13	35,000	2.2	0.1257	52.6	44.7
M-12	28,000	22.0	0.0125	51.1	47.1
M-7	25,000	44.7	0.0064	52.3	43.8
M-4	24,000	67.2	0.0033	38.6	34.9
M-6	23,000	89.0	0.0023	36.3	32.5
M-11	22,000	147.7	0.0017	41.9	24.4
M-10	21,000	239.3	0.0011	45.4	31.6
M-9	20,000	257.7	0.0009	36.1	37.4

TRANSMISSION ELECTRON MICROSCOPY OF IR-RADIATED AUSTENITIC STAINLESS STEEL

H. R. Brager

Transmission electron microscopy studies are being conducted on austenitic stainless steel to provide information for interpreting fast reactor irradiation effects, and to assist in both the formulation of material irradiation behavior models and the improvement of austenitic stainless steel performance in reactor environments.

EBR-II Irradiated AISI Type 304 SS

Transmission electron microscopy examination has been completed on specimens cut from an EBR-II control rod thimble. Quantitative analysis of the micrographs has just started. The irradiation history of the AISI Type 304 SS samples is as follows:

Sample No.	Calculated Irradiation Temperature, °F	Calculated Fluence (E > 0.1 MeV)
A	704	1.2×10^{22}
B	704	2.8×10^{22}
C	728	4.0×10^{22}
D	792	4.8×10^{22}
E	866	3.1×10^{22}
F	866	0.7×10^{22}
G	866	0.03×10^{22}

Sample G, Figures 12.3 and 12.4, which received about 3×10^{20} n/cm² (E > 0.1 MeV) at 866 °F, should exhibit the condition of the microstructure closest to that of the unirradiated steel. Three distinct features should be noted about this section of the steel:

1. Preliminary stereoscopic examination showed about 10^{12} /cm³ of 100 to 200 Å diameter cavities within the foil. Two of these cavities are shown near the center in Figure 12.3. The number and size of the observed cavities are consistent with the amount of helium that would be produced by transmutation of a nominal boron impurity level found in commercial austenitic stainless steel. Therefore, the cavities are probably helium-filled bubbles.
2. Stereo-microscopic results have shown that 1000 to 2000 Å precipitates lie within the foil. These precipitates produce an electron diffraction pattern consistent with Cr₂₃C₆.
3. The dislocation density of Sample G is significantly greater than fully-annealed material. This observation indicated that the unirradiated steel had at least a small amount of cold work--about 5-10% reduction of area--in its unirradiated condition.

The microstructure of Sample G was significantly different than Samples A through F which contained:

- Large cavities whose size and concentration was a strong function of irradiation temperature and fluence.
- A lower density of large precipitates and dislocations than Sample G.
- Displacement damage whose morphology, over the fluence range investigated, was a strong function of irradiation temperature.

Neg 5778-D

96,000X

FIGURE 12.3. Sample G, AISI Type 304 SS Irradiated in EBR-II
at 866 °F to 3×10^{20} n/cm² (>0.1 MeV).

Neg 5774-C

24,000X

FIGURE 12.4. Sample G, AISI Type 304 SS Irradiated in EBR-II at 866 °F to 3×10^{20} n/cm² (>0.1 MeV).

Samples B, Figure 12.5, and Sample E, Figure 12.6, received about the same fluence [3×10^{22} n/cm² ($E > 0.1$ MeV)] and were irradiated at the lowest and highest temperature investigated, 704 and 866 °F, respectively. The thickness of the electron microscopy foils have not yet been determined. By assuming a 1500 Å foil thickness, a preliminary analysis indicates that Sample B contained about 2×10^{15} /cm³ of 100 Å average-diameter cavities, while Sample E contained about 7×10^{14} /cm³ of 250 Å average-diameter cavities. This trend toward a lower concentration of larger cavities at higher irradiation temperatures, over the temperature range investigated, is also shown by Sample D, Figure 12.7. This steel, which received the highest fluence, 5×10^{22} n/cm² ($E > 0.1$ MeV) at 792 °F, contains about 10^{15} /cm³ of 275 Å average-diameter cavities. The microscopic observation showed that Sample D had the greatest amount of irradiation swelling of the samples examined and, thus, is consistent with the macroscopic density measurements.

Samples A through F, which received high fluences, have a lower concentration of both large carbide precipitates and dislocations than Sample G, which received a relatively low fluence of about 3×10^{20} n/cm² ($E > 0.1$ MeV). Very preliminary X-ray analysis of Samples G and D substantiate the observed difference in precipitate morphology of

the specimens. Examination of Sample G produced two strong M₂₃C₆ X-ray peaks while Sample D produced weak X-ray signals just above the background noise level. The metal carbide precipitate concentration could have been reduced by displacement damage. The damage mechanism could displace carbide atoms into the matrix, and thereby increase the effective solubility of the carbon in the steel at the irradiation temperature. Similar results have been previously observed by dissolving and re-forming small bubbles of inert gas in irradiated and postirradiation annealed UO₂.⁽²⁾

It appears that the microstructure of the examined austenitic stainless steel irradiated to high fluence (about 10^{22} n/cm²) is effectively independent of the preirradiation dislocation network, but is strongly dependent on irradiation temperature. Sample G (Figures 12.3 and 12.4), irradiated to the relatively low fluence of 3×10^{20} n/cm² ($E > 0.1$ MeV), contained a higher dislocation density than annealed material. However, no dislocations were observed in the microstructure of Sample C, irradiated to a high fluence of about 4×10^{22} n/cm² ($E > 0.1$ MeV) and contrasted in Figures 12.8 and 12.9. Either the displacement damage of the steel irradiated to high fluence is great enough to mask the dislocations, or the existing dislocations were able to climb and be annihilated by interaction with other dislocations and/or grain boundaries during the irradiation. In either event, the

Neg 5742-F

96,000X

FIGURE 12.5. Sample B, AISI Type 304 SS, Irradiated in EBR-II at 704 °F to 2.8×10^{22} n/cm² (>0.1 MeV), Microstructure Out of Contrast.

Neg 5729-D

96,000X

FIGURE 12.6. Sample E, AISI Type 304 SS, Irradiated in EBR-II at 866 °F to 3.1×10^{22} n/cm² (>0.1 MeV), Microstructure Out of Contrast.

Neg 5668-B

96,000X

FIGURE 12.7. Sample D, AISI Type 304 SS, Irradiated in EBR-II at 792 °F to 4.8×10^{22} n/cm² (>0.1 MeV), Microstructure Out of Contrast.

Neg 5825-E

96,000X

FIGURE 12.8. Sample C, AISI Type 304 SS, Irradiated in EBR-II at 728 °F to 4.0×10^{22} n/cm² (>0.1 MeV), Microstructure in Contrast.

Neg 5824-C

24,000X

FIGURE 12.9. Sample C, AISI Type 304 SS, Irradiated in EBR-II at 728 °F to 4.0×10^{22} n/cm² (>0.1 MeV), Microstructure in Contrast.

dislocation structure of Sample G was not observable in the microstructure of the other samples irradiated to a high fluence [about 10^{22} n/cm² ($E > 0.1$ MeV)] over the temperature range investigated.

The microstructure of these high neutron fluence specimens varied with irradiation temperature. At the 704 °F, the microstructure consisted of a small amount of ordered structure along crystallographic planes in addition to the normal low temperature displacement damage. At 728 °F irradiation temperature, a low concentration of ordered structure is observed which appears to be small faulted loops (Figure 12.9). At 866 °F irradiation temperature, the size and concentration of the faulted loops have significantly increased with a corresponding decrease of the low temperature displacement damage (Sample F, Figure 12.10).

ETR Irradiated AISI Type 316 SS

Transmission electron microscopy examination was completed on AISI Type 316 SS irradiated in a thermal reactor (ETR) at 1000 °F in a sodium environment to 1.4×10^{21} n/cm² ($E > 0.1$ MeV). Compared to its control material, the irradiated steel had shown a nearly constant amount of hardening over a test temperature range from 70 to 1380 °F. This type of high temperature hardening has been correlated with irradiation-produced cavities.⁽³⁾

Three of the tested steel specimens were examined. One specimen was from the button head section of the irradiated material tested at room temperature (Figure 12.11). This micrograph should exhibit the as-irradiated microstructure. The second specimen was of the button head section of the irradiated material tested at 1380 °F (Figure 12.12). This micrograph should exhibit the condition of the irradiated material after about a 1 hr postirradiation anneal at 1380 °F. The third specimen was an unirradiated thermal control specimen tested at 1380 °F (Figure 12.13). The as-irradiated and the postirradiation-annealed steel contained large pores. The unirradiated control sample did not exhibit any pores. The pores observed in the irradiated Type 316 SS were not similar to irradiation-produced cavities previously found in irradiated austenitic stainless steels examined at PNL. Sixteen pairs of stereo electron micrographs of the as-irradiated material were examined. In all cases, the distribution of the observed pores was not uniform within the foil. About half of the pores were surface pits while the balance appeared to be within the foil, but were positioned near both foil surfaces. To determine if the pores were irradiation cavities or surface pits, an irradiated tensile specimen that had not been tested was sectioned for electron microscope examination. The same conditions were used to prepare the thin foils of all the Type 316 SS

Neg 5654-E

96,000X

FIGURE 12.10. Sample F, AISI Type 304 SS, Irradiated in EBR-II at 866 °F to 7×10^{21} n/cm² (>0.1 MeV), Microstructure in Contrast.

Neg 5632-B

96,000X

FIGURE 12.11. Microstructure of Button-Head from AISI Type 316 SS Irradiated in the ETR at 1000 °F to 1.4×10^{21} n/cm² ($E > 0.1$ MeV) and Tested at Room Temperature.

Neg 5575-A

96,000X

FIGURE 12.12. Microstructure of Button-Head from AISI Type 316 SS Irradiated in the ETR at 1000 °F to 1.4×10^{21} n/cm² ($E > 1$ MeV) and Tested at 1380 °F.

specimens. The as-irradiated steel sample did not contain cavities. The microstructure, which consisted

of a high density of precipitates, was very similar to the unirradiated control sample (Figure 12.13).

Neg 5589-E

96,000X

FIGURE 12.13. Microstructure of Button-Head from AISI Type 316 SS Annealed 86 hr at 1110 °F and Tested at 1380 °F.

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. When these alloys are irradiated at service temperatures, however, they generally suffer embrittlement and reduction in rupture life. The purpose of this program is to determine the effects of modified microstructures upon the irradiation stability of nickel-base alloys. These structural modifications are made by preirradiation thermal or thermomechanical treatments and are evaluated by tensile and stress-rupture tests and by microstructural examinations.

The work to date has shown that improvements can be achieved for Hastelloy X-280 and Inconel 600 in postirradiation tensile and stress-rupture properties after irradiation at 540 °F^(4,5) and in postirradiation tensile properties after irradiation at 1250 °F to fluence levels of approximately 1×10^{20} n/cm² (E > 1 MeV).⁽⁶⁾

These improvements have been related to the improved radiation-damage resistance and thermal stability afforded by the microstructures produced by these treatments. However, these treatments also produced (unirradiated) ductilities that were often lower than those in the respective standard solution treatments. Due to this, an investigation of new

treatments that could produce thermally stable microstructures without causing important ductility losses was started.

Because of a switch in program emphasis from nickel-base to stainless steels alloys, these investigations were initiated upon 304 SS. The treatments considered were to be thermomechanical in nature also, but without cold-working. Instead, they were to involve a technique referred to as "stress-aging".⁽⁷⁾

Stress-aging of steels has been shown to yield increases (up to 3-fold) in room temperature proportional elastic limits (PEL) and 0.2% YS (over 50%) while maintaining, and sometimes increasing, elongation.⁽⁷⁾ These treatments consist of placing the specimen under constant load at temperature. (Optimum load, temperature, and duration of loading are all parameters that must be determined for each alloy.)

By increasing the PEL (assuming service temperature PEL is increased), these treatments could improve the ability of an alloy to withstand a given stress at lower strain levels and, thus, diminish the requirement of good grain boundary ductility during irradiation. Also since these treatments may impart good ductility to the alloy, and because the microstructural changes occurring during the treatments may contribute to improved thermal and radiation stability, they were considered worthy of further investigation.

The initial phase of the program was the determination of the effect

of various simple stress-aging treatments on the room- and elevated-temperature tensile properties. Results to date are from tensile tests at room temperature and at 1000 °F.

Stress levels ranged from about 11,400 to 30,800 psi. Specimens were held at these stresses for 1/2 hr at temperatures ranging from room temperature to 1400 °F; they were then air cooled.

Room temperature tests showed that for a treatment consisting of a 30,800 psi stress at 1200 °F, the PEL could be increased 450% (50,600 versus 11,430 psi) and the 0.2% YS could be increased 87% (57,050 versus 30,450 psi) while maintaining 49% total elongation (versus 77% for the annealed condition). A better ductility of 71.5% total elongation was produced by another treatment (30,800 psi at 800 °F) with lesser improvements in strength levels; namely, a 300% increase in PEL (33,500 versus 11,430 psi) and a 53% increase in 0.2% YS (48,800 versus 30,450 psi).

Tensile tests at 1000 °F showed that these treatments could also have beneficial effects upon intermediate temperature properties. A treatment consisting of a 33,800 psi stress at 800 °F increased the PEL by 180% (22,450 versus 8,070 psi) and the 0.2% YS by 100% (35,470 versus 17,550 psi) while maintaining good ductility, 35.3% total elongation versus 42.2% for the annealed condition.

The effect of these simple treatments on high temperature tensile properties (1200 °F) will

be determined next; then, the effect of additional parameters will be examined. The results of these experiments will be reported next quarter.

IN-REACTOR MEASUREMENTS OF MECHANICAL PROPERTIES

E. R. Gilbert and N. E. Harding

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

Hastelloy-X

A correlation by use of the Larsen-Miller rupture-life parameter was made for in-reactor-and unirradiated control creep-tests on Hastelloy-X tested at 1000 °C, with lower temperature published postirradiation and unirradiated control creep data. A plot of the creep stress versus Larsen-Miller rupture-life parameter for these data is shown in Figure 12.14. The data of Robertshaw, et al.⁽⁸⁾ at 540 and 650 °C and the data of Lovell⁽⁹⁾ at 650 and 730 °C provide a good correlation between the low-temperature postirradiation

Neg 0682018-1

FIGURE 12.14. Stress Versus Larson-Miller Parameter, Temperature Compensated Rupture Life, for Stress-Rupture of Hastelloy-X.

data and the 1000 °C in-reactor and unirradiated control data. A change of slope is observed in both the postirradiation and unirradiated control data at 650 °C, with the 1000 °C data in agreement with the steeper slope segment of the curve.

Zircaloy-2

An in-reactor and an unirradiated control creep curve is shown in Figure 12.15 for Zircaloy-2, cold-worked 16% and stress relieved 72 hr at 400 °C. The strain measured in-reactor during neutron irradiation is greater than that measured in the unirradiated control test. This be-

havior is consistent with that reported by Fidleris and Williams⁽¹⁰⁾ on similar material tested under similar conditions of stress and temperature.

Zr-2.5 Nb

An in-reactor and an unirradiated control creep curve is shown in Figure 12.16 for Zr-2.5 Nb quenched from 870 °C and aged at 500 °C. The strain measured in-reactor during neutron irradiation is lower than that measured in the unirradiated control test. This behavior reflects irradiation strengthening during the in-reactor creep.

GRAIN BOUNDARY STABILIZATION

E. R. Bradley

The purpose of this program is to study the recrystallization and grain growth characteristics of metals containing small concentrations of inert gases.

During this period, nickel powders, with and without helium-ion bombardment, have been compacted hydrostatically at pressures of 10, 30, and 50 kbars and sintered at 600 and 800 °C for 2 hr. The density and microhardness of these samples are given in Table 12.5.

The density increased with increasing compacting pressure in both materials in the as-compacted condition. The material containing helium had lower densities than the untreated powder compacts at all pressures. The density increased with increasing sintering

Strain-Time Curves for Zr-2 at 300C and 11.6 kgf/mm².

Neg 0682018-2

FIGURE 12.15. Strain-Time Curves for Zry-2 at 300 °C and 11.6 kg/mm².

Neg 0682018-3

FIGURE 12.16. Strain-Time Curves for Zr-2.5 Nb at 300 °C and 58.3 kg/mm².

TABLE 12.5. *The Density and Microhardness of Nickel Compacts, with and Without Helium, After Various Pressure and Temperature Treatments.*

Helium Injected	Compacting Pressure, kbars	Sintering Temperature, °C-2 hr	Density, gm/cm ³	Microhard- ness, Vickers number
No	10	-	7.380	163
No	30	-	8.359	297
No	50	-	8.506	311
No	10	600	7.538	95
No	30	600	8.468	138
No	50	600	8.511	139
No	10	800	8.029	105
No	30	800	7.817	85
No	50	800	6.418	-
Yes	10	-	6.898	97
Yes	30	-	7.332	224
Yes	50	-	7.983	233
Yes	10	600	7.022	69
Yes	30	600	8.519	139
Yes	50	600	8.072	137
Yes	10	800	7.461	92
Yes	30	800	4.861	140
Yes	50	800	8.240	136

temperatures in both materials compacted at 10 kbars. With higher compacting pressure, the density decreased drastically after sintering at 800 °C in the samples not containing helium. Large holes were formed in the sample compacted at 50 kbars and sintered at 800 °C (Figure 12.17). These holes were probably caused by entrapped gases unable to diffuse out of the compacts during sintering.

The compacts containing helium reacted more favorably when sintered at 800 °C (Figure 12.18). The sample compacted at 30 kbars increased in density after 800 °C, but not as great as after 600 °C. The 50 kbar sample increased in density with increasing sintering temperature. Apparently, the ion bombardment conditioned the surface of the powder, enabled better bonding, and thus improved the sintering characteristics of the compacts.

Neg 468-303A

80X

FIGURE 12.17. *Microstructure of Nickel Powder Without Helium, Compacted at 50 kbars and Sintered at 800 °C for 2 hr.*

Neg 468-383A

80X

FIGURE 12.18. *Microstructure of Nickel Powder Containing Helium Compacted at 50 kbars and Sintered at 800 °C for 2 hr.*

Recrystallization occurred in both materials as indicated by the decrease in hardness and microscopic examination. Considerable porosity and twin markings prevented accurate grain size measurements, and the porosity influenced the accuracy of the microhardness measurements. Further sintering at higher temperatures is being examined to determine if differences in the grain size between the two materials can be detected.

Thin nickel foils of different purities and grain size are being prepared for irradiation at elevated temperatures. The helium concentration can be varied by the boron concentration due to the thermal reactions $^{10}\text{B} + \text{n}^1 \rightarrow \alpha$. Fine grained materials will be examined to determine the effect of grain size on the void formation recently observed in high purity nickel irradiated at elevated temperatures.⁽¹¹⁾

FRACTURE STUDIES

R. G. Hoagland and J. A. Williams

Heavy Section Steel Technology

The Heavy Section Steel Technology program objective is to obtain a quantitative engineering evaluation of the effects of irradiation on heavy-section, pressure vessel steels. The effects of irradiation on the plain strain-fracture toughness are of primary concern. The development of suitable fracture mechanics specimens and the development of valid correlations and surveillance methods

for analyzing service integrity of heavy-section vessels from fracture mechanics data are included in the overall program objective.

The initial fracture toughness (K_{IC}) measurements have been made by using the double cantilever beam (DCB) specimen. The objective was to define its limits of measurement and the validity of K_{IC} for the range of material strengths currently under consideration in the fabrication of heavy-section reactor pressure vessels. The material currently being investigated is A 533-B, quenched and tempered, 12 in. thick steel plate with a room temperature yield strength of 70 ksi.

The fracture toughness of A 533-B as a function of temperature is shown in Figure 12.19. All data are from the half thickness position of the plate, with the specimen crack plane oriented parallel to the rolling direction and normal to the plate surfaces. The plotted data points are taken from 2-in. gross thickness (B) specimens with a net to gross thickness ratio of 0.35. Validity of the data points is based on the ASTM criteria that $B/(K_{IC}/\sigma_{ys})^2 \geq 2.5$. Yield strength data are taken from 8 in. A 533-B plate⁽¹⁾. All K_{IC} values, with the exception of the 2-in. DCB data points (Figure 1), meet the ASTM validity criteria.

Tensile studies of the HSST program plate (12-in. thick, A 533-B) and current fracture studies may establish different validity criteria for the DCB specimen.

Neg 0681604-9

FIGURE 12.19. Fracture Toughness - Temperature Data for A 533-B HSST Program Plate at 1/2T Position

Fracture Studies

The current interpretations of radiation embrittlement are based essentially on the increase in flow stress and loss of strain hardenability from defect impedance to dislocation generation and motion. This simply amounts to a reduction in the principal mode of absorbing energy during crack propagation. In alpha-zirconium alloys both irradiation hardening and hydrogen pickup are responsible for the embrittlement effects observed after irradiation in hot water. However, as shown in previous reports, the loss of fracture toughness of Zircaloy-2 with exposure to in-reactor conditions saturates at moderate

exposures levels, i.e. approximately 3×10^{20} nvt for both annealed and cold-worked material. This saturation effect in fracture toughness is inconsistent with mechanisms based on either defect impedance to slip or formation of a brittle hydride phase; flow stress and weight gain continue to increase with no apparent tendency for saturation.

Deformation twinning, which has commonly been associated as a mechanism for initiating cracks in ferrous alloys, is presently being explored for its contribution to the fracture toughness of Zircaloy-2 for the purpose of explaining saturation effects. The initiation of twinning, unlike slip, can not be associated with a critical resolved shear stress;

instead, it is thought that twins are formed at stress concentration sites. As a crack tip should provide a logical region for forming twins, it is reasonable to expect that the stress intensity level required to initiate a given amount of twinning would be much more insensitive to the defect state than that required to initiate a given amount of slip. Thus, if we consider the fracture toughness in terms of the effective surface energy, γ_e , γ_e can be thought of as a composition of primarily two plastic work terms-- a contribution from slip, γ_d , and a contribution from twinning, γ_t . From the simplest viewpoint, γ_e and, hence, the fracture toughness, K_{IC} , will be reduced during irradiation because of defect impedance effects on γ_d ; γ_t will remain essentially unaltered. As γ_d is reduced with irradiation, γ_e should asymptotically approach a value determined by γ_t . While this describes the essential elements for saturation, the actual mechanisms must certainly be much more involved. For example, the amount of twinning occurring at initiation of crack extension must depend on the applied stress intensity. Secondly, twinning will often result in the rotation of the lattice into an orientation more favorable for slip. There also seems to be no direct manner in which one could separate the contributions from the two deformation modes.

An indirect approach to analyze the contribution of twinning to the fracture toughness of Zircaloy-2, presently being explored, utilizes the inherent anisotropy of Zircaloy-2 in rolled plate. The rolling texture

in Zircaloy-2 plate gives rise to a lower toughness for cracks propagating parallel to the rolling direction (RD) relative to the long transverse direction (TD). This anisotropy in toughness is retained after fluence levels of 1×10^{21} nvt and, therefore, may reflect a difference in the number of grains favorably oriented for the growth of twins between the two plate orientations. A test of this hypothesis requires an examination of the magnitudes of the shear stress from the crack tip stress field (resolved on the active twin and slip planes) and in the direction of shear. This must be done for various orientations of the hexagonal close packed (hcp) unit cell with respect to the crack plane. The following paragraphs outline the method for determining the orientation factors for twinning about a crack in a hcp material. The results, however, have not been completed.

We are given the crack tip stress field defined in terms of a right-handed rectilinear system where x is parallel to the crack propagation direction, y is perpendicular to the crack plane and z is parallel to the crack front. To identify the shear stress acting in the shear direction; it is necessary to rotate the original coordinate system. After rotation, the convention chosen is one where the x -axis coincides with the shear direction and the y -axis is perpendicular to the plane of shear. To obtain the transformation matrix, which determines this rotation, is necessary to describe the orientation of the shear plane and shear direction in terms of the original

rectilinear coordinate system. If $\vec{i}$, $\vec{j}$, and $\vec{k}$ are unit vectors along the x, y, z axes, respectively, then the four unit vectors in the hex Miller system are

$$\begin{aligned}\vec{a}_1 &= \ell_1 \vec{i} + m_1 \vec{j} + n_1 \vec{k} \\ \vec{a}_2 &= \ell_2 \vec{i} + m_2 \vec{j} + n_2 \vec{k} \\ \vec{a}_3 &= \ell_3 \vec{i} + m_3 \vec{j} + n_3 \vec{k} \\ \vec{c} &= \ell_4 \vec{i} + m_4 \vec{j} + n_4 \vec{k}\end{aligned}$$

where the ℓ 's, m 's, and n 's are the direction cosines between the axes of the two coordinate systems. To simplify the notation, define the following matrices as:

$$\hat{T} = \begin{bmatrix} \ell_1 & m_1 & n_1 \\ \ell_2 & m_2 & n_2 \\ \ell_3 & m_3 & n_3 \\ \ell_4 & m_4 & n_4 \end{bmatrix}$$

the matrix of the direction cosines,

$$\tilde{v} = \begin{bmatrix} u & o & o & o \\ o & v & o & o \\ o & o & -(u+t) & o \\ o & o & o & w \end{bmatrix}$$

where u , v , t , w are the indices of a vector in the hex system [for which $t = -(u+v)$],

$$\tilde{h} = \begin{bmatrix} a_1 \\ a_2 \\ a_3 \\ c \end{bmatrix}$$

a column matrix of the hex unit vectors, and

$$\tilde{r} = \begin{bmatrix} i \\ j \\ k \end{bmatrix}$$

a column matrix of the rectilinear unit vectors. By using the above notation, Equation (1) becomes

$$\tilde{h} = \hat{T} \tilde{r}.$$

A vector described by the indices of one system may be described by the indices of the other system by means of the relation

$$\tilde{v} \tilde{h} = \tilde{v} \hat{T} \tilde{r}.$$

A plane, given as $(h \ k \ i \ \ell)$ in the hex system, has intercepts on the a_1 , a_2 , a_3 , and c axes at $1/h$, $1/k$, $1/i$, $1/\ell$, respectively. In the rectilinear system these points are at

$$\begin{aligned}1/h &(\ell_1, m_1, n_1) \\ 1/k &(\ell_2, m_2, n_2) \\ 1/i &(\ell_3, m_3, n_3) \\ 1/\ell &(\ell_4, m_4, n_4)\end{aligned}$$

where, of course, only three of these points are needed to define the plane.

Let the matrix $\hat{H}$ be given by

$$\hat{H} = \begin{bmatrix} \ell_1/h & m_1/h & n_1/h \\ \ell_2/k & m_2/k & n_2/k \\ \ell_4/\ell & m_4/\ell & n_4/\ell \end{bmatrix}$$

This matrix allows the determination of the rectilinear indices of the $(h \ k \ i \ \ell)$ plane. Thus, to define the orientation of the three equations, the form $Ax + By + Cz = 1$ must be solved to determine the coefficients, A , B , and C . In matrix notation these equations are

$$\hat{H} \begin{matrix} A \\ B \\ C \end{matrix} = \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix}.$$

The solution of the above equation gives A as the summation of the terms in the first row in the inverse of $\hat{H}$, etc. As A , B , and C are the indices of the plane in the rectilinear system, they are also the direction cosines of the plane's normal. Once the matrices $\hat{H}$ and $\hat{T}$

are known, the problem of determining the proper transformation matrix for rotating the x, y, z , axes is essentially solved.

Let I represent a matrix whose components are $I_1 (= A)$, $I_2 (= B)$, and $I_3 (= C)$; the direction cosines between the y -axis in the rotated system; and the x, y, z axes, respectively, in the original system. Let II represent the x -axis in the rotated system. The components II_1 , II_2 , and II_3 are the direction cosines between x' and x, y, z , respectively, and are given by the summations of the terms in the columns of the matrix $\hat{V}^T$. The third axis, z' , represented by III , is defined in terms of the direction cosines determined from the vector product of II and I . Therefore, the transformation matrix is

$$\hat{R} = \begin{bmatrix} II_1 & II_2 & II_3 \\ I_1 & I_2 & I_3 \\ III_1 & III_2 & III_3 \end{bmatrix}.$$

The stress tensor in the transformed system is

$$\sigma' = \hat{R}^T \sigma \hat{R}$$

where σ is the plane strain crack tip stress tensor

$$\sigma = \begin{bmatrix} \sigma_{xx} & \sigma_{xy} & 0 \\ \sigma_{xy} & \sigma_{yy} & 0 \\ 0 & 0 & \nu(\sigma_{xx} + \sigma_{yy}) \end{bmatrix}.$$

The shear stress σ_{xy} in the rotated system is the shear stress acting on the shear plane and in the shear direction. Letting $\tau = \sigma_{xy}$, this stress is given by

$$\begin{aligned} \tau = & II_1 II_2 \sigma_{xx} + I_2 I_1 \sigma_{yy} \\ & + (II_1 I_2 + I_1 II_2) \sigma_{xy} \\ & + \nu III_1 III_2 (\sigma_{xx} + \sigma_{yy}) \end{aligned}$$

where

$$\begin{aligned} III_1 III_2 = & (I_2 II_3 - I_3 II_2) \\ & (I_3 II_1 - I_1 II_3). \end{aligned}$$

The variation in τ must be examined both in terms of the orientation of the hcp unit cell with respect to the crack plane and the angular variation in the stresses around the crack tip. The orientation of the hex cell and, therefore, the rotation of the rectilinear system is specified by the angle, λ (between the a_1 - and x -axes), and the angle, ϕ (between the c - and z -axes). The angle, θ , is the angle between the radius from the crack tip vector to a point in the body and the x -axis, and it expresses the angular dependence of the crack tip stresses given by

$$\sigma_{xx} = \frac{K}{\sqrt{2\pi r}} \cos \frac{\theta}{2} \left(1 - \sin \frac{\theta}{2} \sin \frac{3\theta}{2} \right),$$

$$\sigma_{yy} = \frac{K}{\sqrt{2\pi r}} \cos \frac{\theta}{2} \left(1 + \sin \frac{\theta}{2} \sin \frac{3\theta}{2} \right),$$

and

$$\sigma_{xy} = \frac{K}{\sqrt{2\pi r}} \cos \frac{\theta}{2} \sin \frac{\theta}{2} \cos \frac{3\theta}{2}.$$

The factor introducing anisotropy in τ comes from a consideration of the rolling texture. Of Zircalloy-2, analysis of pole figure shows that the most predominant feature of the rolling texture is the very low density of basal poles aligned parallel to the rolling direction. The basal poles are fairly uniformly distributed between the short and long transverse directions. Essentially then, the analysis of the orientation dependence of τ becomes one of rotating the basal poles about the x -axis in the yz -plane for RD cracking and about the z -axis in the zy -plane for TD cracking.

The experimental data showing the saturation and anisotropy effects was collected at temperature ranging from slightly above room temperature down to -190°C . At these temperatures, the only active twin systems are $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$; these, in addition to the $\{10\bar{1}0\} \langle 11\bar{2}0 \rangle$ slip system are presently being examined. Since the terms comprising τ are quite lengthy, this study is being accomplished by means of an 1108 computer.

HIGH PRESSURE STUDIES

G. L. Kulcinski

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

Irradiation Stability of High Pressure Polymorphs

The high pressure forms of Zr, InTe, P, SiO_2 (coesite), PbO_2 , and B_2O_3 have been irradiated to modest fluences at 1 atm and the resulting crystal structures have been examined. The detailed information about the production of these polymorphs has been described earlier,⁽¹²⁾ and the diffraction patterns of the irradiated materials were obtained on a shielded X-ray double diffraction unit.⁽¹³⁾

The irradiations were carried out at reactor ambient conditions ($<20^{\circ}\text{C}$), and the samples were contained in helium-filled, water-cooled, aluminum capsules. All the samples except B_2O_3 were irradiated to $2 \times 10^{19} \text{ n/cm}^2$ ($>0.1 \text{ MeV}$). The B_2O_3 was irradiated to $3.6 \times 10^{20} \text{ n/cm}^2$ (thermal). Gamma heating was negligible for the sample size and geometry used, but the heat generated by the decay of some radioisotopes with high absorption cross sections and short half lives makes the estimate of irradiation temperature somewhat qualitative. It is thought that the samples were irradiated at less than 80°C except for B_2O_3 , which, because of the fissioning of B^{10} atoms, may have reached higher temperatures.

The results of the X-ray analysis are listed in Table 12.6. To the limit of the X-ray measurements ($\pm 0.2\%$), omega zirconium, orthorhombic PbO_2 , and monoclinic SiO_2 show no change in crystal structure or lattice parameter. The cubic form of InTe showed a slight ($\sim 30\%$) reversion to its stable semiconducting form, while the monoclinic structure of B_2O_3 and the orthorhombic phase phosphorous were completely destroyed.

The omega zirconium sample, which contained trace amounts of the alpha starting material before irradiation, showed no tendency to form any more alpha during irradiation. In fact, the relative intensities of the alpha X-ray lines were somewhat diminished when compared to the omega diffraction pattern. Experiments at higher fluences are being conducted to determine the stability of this

TABLE 12.6. Effect of Fast Neutron (> 0.1 MeV) Irradiation on the Crystal Structure of Various High Pressure Polymorphs^(a)

Material	Initial Structure	Final Structure	Comments
Zr	Omega & Trace Alpha	Omega & Trace Alpha	<0.2% Change in Lattice Parameter
InTe	Cubic	~70% Cubic ~30% Tetragonal	Possibility of Thermal Conversion at 80 °C
PbO ₂	Orthorhombic	Orthorhombic	<0.2% Change in Lattice Parameter
SiO ₂	Monoclinic	Monoclinic	<0.2% Change in Lattice Parameter
P	Orthorhombic	Amorphous	--
B ₂ O ₃	Monoclinic	Amorphous ^(b)	Faint Lines at 3.20 Å and 2.25 Å Detected

(a) Irradiated to 2×10^{19} n/cm² at $T < 80$ °C.

(b) Irradiated in separate capsule to 3.6×10^{20} n/cm² (thermal).

metastable structure with respect to neutron irradiation.

The amount of cubic InTe transformed was estimated by comparing the relative intensities of the (220) lines of both structures. The crystal structure factors were calculated and used to normalize the X-ray peak heights. The issue of an irradiation-promoted change is somewhat clouded by the extreme sensitivity of the cubic InTe to thermal decomposition. Banus et al.⁽¹⁴⁾ has shown that 75% reversion will occur in 1 hr at 100 °C, with the cubic to tetragonal transformation very rapid above 125 °C. The irradiation capsules used were not instrumented, and the sample temperature had to be calculated from the gamma heating rate, and decay heat generated by

such isotopes as ¹¹⁶In. Assuming that the sample remained in intimate contact with the aluminum capsule during the irradiation and cool down period, it is calculated that the sample temperature never exceeded 80 °C. Although an unirradiated control sample maintained at 25 °C for the duration of the experiment showed no conversion, a long-term 80 °C test is required to confirm the cause of the 30% conversion (thermal- or irradiation-induced). However, one can definitely state that fast neutron irradiation does not completely destroy the metastable cubic InTe structure, because 70% of it remained after 2×10^{19} n/cm².

The results for SiO₂ (coesite) compare very favorably with those of

Whittles,⁽¹⁵⁾ who found no change (to within 0.1%) in the lattice parameter of coesite after exposure to 2×10^{20} fast neutrons/cm². This resistance to damage by fast neutron is interpreted as the difficulty of rearrangement of the SiO₂ tetrahedra due to closely packed structure.

The fact that orthorhombic PbO₂ has about the same degree of conic bonded as SiO₂, and also displays considerable stability to radiation damage, is being studied in more detail. Both of these compounds have characteristic reconstructive phase transitions which undoubtedly assist in retaining the metastable phase.

The conversion of black phosphorous (orthorhombic form) to an amorphous structure is in opposition to the results of Chipman et al,⁽¹⁶⁾ who found that it was relatively unaffected by fast neutron bombardment. However, it is difficult to compare the present results to that experiment because the total fast fluence in the latter is not given. The authors⁽¹⁶⁾ only state that "the samples were irradiated in the Brookhaven reactor for one month at a temperature of 50 °C". They do indicate that a small amount of red phosphorous may have been produced, and more recent communication⁽¹⁷⁾ revealed that they may have had a factor-of-10 less fast fluence on their samples.

The conversion of B₂O₃ from its dense monoclinic form to an essentially amorphous structure (two lines were detected at 3.20 Å and 2.25 Å, but neither fit into the monoclinic pattern) can undoubtedly be attributed

to the large number of B¹⁰ fissions. Preliminary calculations show that practically all of the B¹⁰ atoms were burned up under a thermal exposure of 3.6×10^{20} n/cm². This means that one out of every six boron atoms had fissioned, and that one out of every three B₂O₃ molecules had been disrupted. With such internal damage, it is not surprising that long-range order was effectively removed.

These preliminary results show that the metastable metallic, ionic, and covalent nonfissionable high pressure polymorphs are not affected by fast neutron irradiation to 2×10^{19} n/cm². However, the more weakly bound P does not revert, which indicates a dependence of metastability on the strength of the interatomic bonds. The reversion in B₂O₃ represents an extreme case and must be treated separately. Future experiments will be conducted at higher fluences for those unaffected by the present irradiations, and lower fluences for those whose metastability was removed.

IN-REACTOR CORROSION OF ZIRCONIUM ALLOYS

A. B. Johnson, Jr.

ATR "Clean Loop" Test

A "clean loop" corrosion test was described in a previous Quarterly Progress Report.⁽¹⁸⁾ The objective of the test is to differentiate between water chemistry and fissionable contamination factors in explanations that have been offered

for flux-enhanced corrosion of zirconium alloys. The test will be conducted in the North loop of the Advanced Test Reactor (ATR) in a system that has not been exposed to fissionable contamination. Coolant conditions will simulate the most aggressive corrosion environment from a series of tests in the G-7 Loop of the Engineering Test Reactor (ETR).⁽¹⁹⁾ The proposed coolant conditions are as follows:

Temperature	280 °C
Pressure	2150 psi
Dissolved Oxygen	>0.5 ppm <4.0 ppm
Tentative Goal	1 ppm
Coolant	pH 10 LiOH
Fast Fluence	>1 x 10 ²⁰ , >1 MeV
Exposure	ATR Core 2 and Core 3

Three specimen holders and associated hardware were supplied by Knolls Atomic Power Laboratory (KAPL). Specimens were charged into the holders and delivered to the National Reactor Testing Station in April 1968. Specimens include Zircaloy-2, crystal bar zirconium, and specimens from sample stock characterized in ETR tests--all supplied by Battelle-Northwest; Zircaloy-4 supplied by KAPL; and Zr-2.5 Nb supplied by Atomic Energy of Canada. Each of the specimen holders contains 42 coupons with dimensions 1.94 by 0.94 by 0.040 in. (nominal thickness). Zircaloy-2 and Zr-2.5 Nb specimens, having platinum stubs to act as shorts through the oxide, are included in each holder. Two holders will be positioned at high-flux positions in the loop, with the third holder positioned above the core at the exit end of the loop.

Out-of-reactor control specimens will be exposed in an autoclave at Battelle-Northwest.

The three specimen holders will be charged for exposure during ATR Core 2 and Core 3 operation. One of the two in-flux holders will be discharged at the end of Core 2; the two remaining holders will be discharged at the end of Core 3. Water chemistry control under irradiation will be investigated during Core 1 operation, prior to insertion of the specimens.

Following the "clean loop" test, the oxygen level in the pH-10 LiOH coolant will be reduced to <0.05 ppm. Specimens similar to the "clean loop" specimens will be exposed in the low-oxygen coolant to investigate dissolved oxygen level as an in-flux corrosion parameter.

The primary objective of the test is to differentiate between water chemistry and fission fragment effects on in-flux corrosion. Additional goals of the test include the following:

Comparison of Alloy Response to a Combination of Flux and Dissolved Oxygen

The Zr-2.5 Nb alloy is sensitive to dissolved oxygen in out-of-reactor environments, while Zircaloy-2 corrosion is essentially unaffected by dissolved oxygen.⁽²⁰⁾ However, Zircaloy-2 shows a sensitivity to dissolved oxygen in reactor environments.^(19,20) The "clean loop" test provides an opportunity to compare the combined effects of dissolved oxygen and flux on the test alloys and crystal bar zirconium.

Effect of Pretreatment

Experience in the ETR G-7 loop indicates that prefilming accelerates the in-flux corrosion of Zircaloy-2 and Zircaloy-4.⁽¹⁹⁾ The "clean loop" and "post clean loop" tests will compare the prefilm effect as a function of alloy, film thickness and oxygen level. The test includes as-etched and prefilmed specimens of each type. The prefilm conditions are indicated below:

Zircaloy-2	-	300 °C water 7 days
		400 °C steam 8 hr
		400 °C steam 21 days
Zircaloy-4*	-	360 °C water 3 days
		360 °C water 168 days
		330 °C water 28 days
Crystal Bar Zirconium	-	300 °C water 7 days
Zr-2.5 Nb	-	400 °C steam 24 hr
		400 °C steam 14 days

The prefilm treatments produced film thicknesses of ~ 10 mg/dm² and 40 to 50 mg/dm². Zircaloy-2 was prefilmed at 300 °C to compare with G-7 loop prefilm treatments; the prefilms at 400 °C provide comparisons of prefilm condition (300 °C versus 400 °C) and film thickness (~ 10 versus ~ 40 mg/dm²). Zr-2.5 Nb prefilm treatments will provide film thickness comparisons (~ 10 versus ~ 40 mg/dm²). The Zircaloy-4 specimens were supplied from KAPL stock prepared for a broad series of in-reactor and out-of-reactor tests. The prefilm conditions vary from the remaining "clean loop" alloys, but the film thickness comparison is

similar. The effect of prefilm conditions on in-flux corrosion is under investigation in the G-7 loop on Zircaloy-2 exposed in nine prefilm conditions.

Effects of Electron Conductivity Through the Oxide

Zircaloy-2 and Zr-2.5 Nb specimens have been prepared with platinum stubs to act as electrical shorts through the oxide. Behavior of the shorted specimens will be investigated in high-oxygen, low-oxygen and out-of-reactor environments.

Postirradiation examination of the specimens will be conducted at Battelle-Northwest, Richland, Washington.

ETR G-7 Loop Tests

Corrosion specimens of several materials have been prepared and assembled for in-flux corrosion studies in the ETR G-7 loop. The materials are summarized below:

- NPR Zircaloy-2 Lots AT, CT and HT
 - As-etched
 - Prefilmed 400 °C steam, 360 days, followed by 280 °C pH-10 NH₄OH, 140 days.
- Zircaloy-2/Inconel 600 welded joints
- Inconel 600
- 304 SS
- 348 SS

The three lots of NPR Zircaloy-2 have been extensively characterized in the ETR G-7 loop in pH-10 LiOH ~ 1 ppm O₂.⁽²¹⁾ In-flux corrosion

* The sample array includes two Zircaloy-4 lots. One lot was exposed in three metallurgical conditions.

rates were accelerated, but the degree of acceleration was lot dependent. A more recent G-7 loop exposure on the three Zircaloy-2 lots in pH-10 NH_4OH , <0.05 ppm O_2 resulted in almost complete absence of accelerated corrosion on specimens exposed as-etched.⁽²²⁾ Accelerated corrosion appeared on prefilm specimens, but the degree of acceleration varied with prefilm treatment. The current test series has been prepared to further characterize the in-flux corrosion behavior of the three Zircaloy-2 lots in pH-10 NH_4OH .

Zircaloy-2/Inconel transition joints exposed in the ETR G-7 loop in pH-10 LiOH showed evidence of preferential hydride accumulation near the welded interface.⁽²³⁾ Exposure of the joints in pH-10 NH_4OH is undertaken to further our understanding of the corrosion and hydriding behavior of metal couples in reactor fluxes. Uncoupled Inconel 600 is exposed as a control material for the metal couples. In-reactor corrosion rates on stainless steels have not been extensively characterized, although the data are of considerable significance to the analysis of crud generation and deposition in reactor systems containing stainless steel. The corrosion data also will serve to characterize corrosion effects on mechanical property specimens exposed in the G-7 loop.

FAST REACTOR SUPPORTING STUDIES

FAST NEUTRON MECHANISMS

R. P. Allen

The objective of this work 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. Special hydrogen permeation and gas evolution techniques are currently being developed and used to investigate the effects of irradiation, composition, deformation, stress gradients, phase instabilities, surface condition, and desorption environment on the normal hydrogen diffusion, solubility, and effusion characteristics of the austenitic stainless steels.

The use of a nonisothermal gas-evolution technique for rapid, qualitative, screening-type investigations of hydrogen behavior in fast reactor alloys was described in the preceding quarterly progress report.⁽²⁴⁾ With this method, the effects of various material and environmental parameters on the combined hydrogen diffusion, occlusion, and effusion characteristics of reactor alloys can be readily determined, and any anomalies or temperature intervals of particular interest can be identified for further investigation.

The results of a preliminary investigation, using this technique, of

the effect of cold work on the hydrogen evolution characteristics of AISI 348 SS were also reported.⁽²⁴⁾ Room temperature deformation was shown to have an appreciable accelerating effect on the elevated-temperature deuterium diffusion- and effusion-behavior of this alloy. The three maxima present in the effusion rate-versus-temperature curve for the annealed condition were eliminated, and the position of the new curve was displaced to a lower temperature.

In the following section, some additional work, which include the use of AISI 304L SS and both isothermal and nonisothermal gas-evolution techniques, is discussed that confirms this cold-work effect and establishes a correlation between the enhanced hydrogen mobility in cold-worked, metastable, austenitic stainless steel and the deformation-induced martensite content.

Cold Work Effects

The observation of a beneficial effect of room temperature plastic deformation on the elevated-temperature deuterium diffusion- and effusion-characteristics of AISI 348 SS⁽²⁴⁾ is contrary to the type of behavior noted for hydrogen in iron and the ferritic steels. For these materials, the apparent activation energy for hydrogen diffusion is substantially increased by cold working, so even at relatively high temperatures the hydrogen diffusivities in cold-worked samples are appreciably less than those for the annealed condition.⁽²⁵⁾ Normally, then, the behavior of

hydrogen in cold-worked material and the interaction of the interstitially-diffusing atoms with deformation-produced defects would be described more in terms of "trapping" mechanisms than any enhancement of atomic mobility or possible provision of preferred paths for diffusion.

For the metastable austenitic stainless steels, however, plastic deformation at lower temperatures ($<M_D$) is characterized by the introduction of body-centered cubic (bcc) martensite into the parent face-centered cubic (fcc) austenite matrix. This martensitic mode of deformation competes with the normal slip mechanism and becomes dominant at low temperatures.⁽²⁶⁾ Furthermore, the diffusivity of hydrogen in bcc iron is four times that for fcc iron at the allotropic transformation temperature (910 °C), the extrapolated difference increases markedly with decreasing temperature; this effect of crystal structure on diffusivity appears to predominate over the influence of steel composition.⁽²⁷⁾

These facts suggest that the enhanced deuterium evolution behavior noted for cold-worked AISI 348 SS is caused by the presence of deformation-induced martensite within the austenite matrix, and that the diffusivity of hydrogen in this bcc martensite is substantially greater at the test temperatures employed than in the fcc austenite. To test this hypothesis, specimens of AISI 304L SS were thermally charged with

deuterium, cold worked at room temperature (RT) or liquid nitrogen temperature (LN), and their deuterium effusion characteristics determined by using both isothermal and nonisothermal gas-evolution techniques. Thus, by varying the deformation temperature, the amount of martensite formed could be substantially altered for the same amount of cold work. Angel,⁽³⁾ for example, has shown that a true plastic strain of 0.20 yields less than 10% martensite at room temperature and approximately 80% martensite at liquid nitrogen temperature for the typical 18/8 Type SS.

The discoid, AISI 304L SS specimens were saturated with deuterium at 900 °C, oil quenched, deformed by compression at the indicated temperature, ground to a uniform thickness (1 mm), and

electropolished. The amount of martensite formed in each of the cold-worked samples was estimated by comparing the magnetic response, as given by an Aminxco-Brenner Magne-Gage, with the value for a sample deformed sufficiently at liquid nitrogen temperature. This gave the limiting martensite content (90%) for this alloy.⁽²⁶⁾

The results of the nonisothermal gas evolution tests are given in Figure 12.20. The heating rate for both specimens was 10 °C/min. Although the amount of cold work was greater for the (RT) specimen (18% versus 13%), the (LN) sample exhibited by far the greater effect of deformation on deuterium evolution behavior. Effusion occurred over a much narrower temperature interval and at a mean

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FIGURE 12.20. Effect of Deformation Temperature on the Nonisothermal Deuterium Effusion Characteristics of AISI Type 304L SS

temperature more than 200 °C less than for the (RT) specimen, which indicates a significantly higher deuterium diffusivity at lower temperatures for this deformation treatment. The estimated martensite contents were 10% and 65% for the (RT) and (LN) samples, respectively.

The isothermal test results are illustrated in Figure 12.21. The slope of the linear portion of the effusion rate-versus-time curves is directly proportional to the diffusivity of deuterium in the test material at 400 °C. Here also, the diffusivity for the (LN) sample (2.5×10^{-6} cm²/sec) was substantially greater than that for the (RT) sample (9×10^{-7} cm²/sec) even though the amount of cold work was appreciably less (5% versus 15%). The es-

timated martensite contents, however, had the same relationship as the observed diffusivities (40% versus 10%). The corresponding 400 °C diffusivity value for an as-annealed control sample (not shown) was 5×10^{-7} cm²/sec. Thus, 5% cold work at (LN) temperature increased the diffusivity of deuterium in the test alloy at 400 °C by a factor of 5, and the non-isothermal test results indicate that this difference would increase substantially with decreasing isothermal test temperatures.

These isothermal and nonisothermal test results thus show

- That cold working enhances the deuterium evolution characteristics of AISI 304L SS
- That this enhancement reflects a

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FIGURE 12.21. Effect of Deformation Temperature on the Isothermal Deuterium Effusion Characteristics of AISI Type 304L SS

significant increase in the elevated temperature deuterium diffusivity characteristics of the cold worked material

- That this diffusivity increase correlates with the deformation-induced martensite content of the test specimens rather than with the actual amount of plastic deformation.

This effect of cold work on hydrogen mobility should be general, then, for all metastable austenitic stainless steels deformed below their M_D temperatures.

Further work is in progress using a semiaustenitic stainless steel (AM 355) that can be heat-treated to give up to 100% martensite with either room temperature deformation or sub-zero cooling. This work is to:

- Determine the quantitative aspects of the diffusivity enhancement versus martensite content relationship
- Examine the effects of martensite tempering and reversion on this relationship
- Compare the hydrogen diffusion behavior for martensite formed spontaneously by cooling below the M_S temperature with that for equivalent deformation-induced martensite contents.

These results will then be related to the probable neutronic hydrogen retention, diffusion, and effusion characteristics of annealed versus cold-worked fast-reactor core components, with particular emphasis on the possible service-induced formation of preferred paths for hydrogen diffusion through hydrogen-sensitive, non-austenitic material.

TEST FACILITIES

R. H. Todd

The comprehensive investigation of the effects of irradiation and environment on candidate structural and cladding materials for use in the FFTF has created a need for a facility that can provide the necessary test environments. Such a facility is now under construction by Battelle-Northwest. It will provide the means for testing control and irradiated specimens in sodium and controlled gas atmospheres. The final phases of construction have been delayed by late delivery of heating and ventilating equipment originally scheduled for delivery in March. This equipment is now on site and installation is in progress. The interior of the building has been finished sufficiently to allow some test equipment to be installed. The relocation of the main transformer for this facility has been completed.

The test stands temporarily located in the mezzanine workshop completed 11 tests on uniaxial specimens during the past three months; 2 tests in static sodium and 9 tests in air. These tests totaled 465 hr of test time for sodium tests and 1345 hr of test time for air tests. One series of tests at 1202 °F is near completion, and another series at 1000 °F has been started. These data are given in a previous section of this report.

The support section of the liquid metal loop was deficient in some respects on the original acceptance test operational run, but the vendor

has agreed to supply those parts needed to correct these deficiencies and they are being shipped. Although minor changes and repairs necessary in instrumentation and electrical controls were made and tested by operating the loop for a short period, other minor repairs are still needed. Present plans call for insertion of a single test station in the test leg of the loop. The test leg will contain five biaxial unirradiated specimens for the first series of tests. After the prototype equipment has been proven in actual service, a gamma shield will be provided, which will allow screening tests of irradiated biaxial specimens in flowing sodium. These tests are expected to reveal the qualitative effects of flowing sodium on the candidate materials. More complete test programs will then be developed from that information.

Present program needs on irradiated, uniaxial specimens emphasize tests in inert gas. Plans are now being made to place the maximum number of uniaxial test units in the remote cell to meet this need. Two prototypical test units are now undergoing operational bench tests prior to being placed in remote operation.

The inert atmosphere glove-box and vacuum oven for handling high purity alkali metals have been received and are now being set up for use. The Sodium Measuring Unit will be positioned alongside the glove-box. Thus high-purity sodium can be capably delivered to a closed vessel or through the glove-box into an open vessel, which can then be

sealed while still in the glove-box.

A chamber for the calibration of thermocouples has been placed in service for cross-checking the numerous control thermocouples required in this facility. This chamber consists of a stainless steel container with thermocouple wells, which is filled with sodium and a small expansion chamber and sealed by welding. All thermocouple wells are surrounded by sodium which has excellent heat transfer properties. The tips of the thermowells are close together in the middle of the sodium mass. Temperature deviation between thermowells is less than can be detected with equipment used in this laboratory (1 F°). This chamber checks thermocouples used in test against a calibrated standard to detect gross deviations or thermocouple breakdown. Frequent cross-checking of this type is necessary to insure specified temperature control of the tests being performed in this facility.

LMFBR FUEL CLADDING INFORMATION CENTER

F. B. Shober

The LMFBR Fuel Cladding Information Center has been established at Battelle-Northwest. This center utilizes a computerized data storage and retrieval system and was designed in conjunction with the REM Program (Radiation Effects to Materials). The purpose of this center is to provide complete information and data for LMFBR cladding materials from the major areas of materials related to primary processes, composition,

fabrication, physical and mechanical properties, quality, fuel element fabrication, and irradiation history. The center provides an analysis of cladding performance in reactor service by first establishing any lot-to-lot variation in properties that may result from melting and fabrication processes, second determining the dependence of any variation on a single manufacturing process variable or combination of variables, and finally providing the analysis of information to evaluate the performance characteristics of a given lot or lots of cladding in specific reactor environments. The latter will provide the information needed for reliability analysis of FTR cladding. The information and data initially will consist of that obtained for

austenitic stainless steels in direct support of FFTF fuel pin experiments and FTR core fabrication.

The initiation point for the design of the LMFBR Information system was the REM Program, which is a functioning multipurpose program designed to store, process, and retrieve large volumes of mechanical test data. A schematic representation of the program is shown in Figure 12.22.⁽²⁸⁾ The program has been used extensively at Battelle-Northwest for the past five years and presently contains data for about 6000 tensile tests stored on tape in five master files. A program incorporating REM was defined and designed to meet the following objectives:

1. To provide for the convenient

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FIGURE 12.22. Schematic Representation of the REM Computer Program

storage of data related to primary material processes and composition, fabrication, physical and mechanical properties, and qualities of LMFBR fuel cladding material;

2. To allow for expansion through the inclusion of similar data concerning fabrication of fuel pins as well as irradiation history data;
3. To provide an interface with the REM Program;
4. To allow for the future incorporation of data analysis and retrieval passes.

The initial phase of the work consisted of identifying and listing all terms associated with melting processes, primary and secondary fabrication, and heat treatment in the manufacture of tubing. A flow diagram was prepared depicting each step in the manufacturing process from melting to the accepted tubes ready for fabrication into fuel pins. It further included listing all tests and inspections to which the tubing will be subjected prior to acceptance by the fuel pin fabricator. This phase has been completed and a computer program written for the storage of these data. A set of data have been prepared for a hypothetical fabrication sequence and will be stored. This will provide a test case situation to determine the effectiveness of the system for storing fabrication information. A questionnaire based on information required has been prepared.

The second phase of the system design will include completion of the section to store physical and mechanical properties data. It is necessary

to modify the original design to provide space to store information on average values and standard deviation pertaining to lot-to-lot comparison and analysis. A retrieval program will be written after these changes have been affected and the system tested.

The effectiveness of the information system depends upon the analysis of stored data as well as data processing. Therefore, a program for analysis of data was prepared by the Statistical Design and Analysis Group of the Mathematics Department. This group further prepared a test case by using test data stored in REM with the Biomed Subroutine (BMD-02V), analysis of variance for factorial design. Analyses were performed on 304 SS data to investigate the effects of heat treatment (cold-worked and annealed) and sample orientation (rolling direction and transverse direction) on the yield strength, ultimate strength, uniform elongation, and total elongation. Data from unirradiated and irradiated samples were analyzed. The results from the test case are not presented, since the selection of data for analysis ignored all differences among samples except material condition and sample orientation and therefore lead to erroneous conclusions. The test case, however, demonstrated the potential capabilities of the Biomed Subroutine Series for mechanical properties analysis and comparison. Additional test cases using REM data will be made to demonstrate further the capabilities of these subroutines to answer questions

related to the dependence and interdependence of manufacturing variables on the lot-to-lot variation in properties of fabricated reactor fuel cladding. A partial list of BMD programs that may be useful for data analysis in the Fuel Cladding Information Center Program is as follows:

1. Multiple Regression with Case Combinations
(BMD 03R). Linear regression analysis of a dependent variable on 2 to 50 independent variables.
2. Analysis of Variance for Factorial Design
(BMD 02V). Tests the effects (on a variable) of up to 999 levels of each of 8 factors with a maximum of 18,000 combinations.
3. Analysis of Covariance for Factorial Design
(BMD 03V). Combines features of regression and analysis of variance. Up to 8 covariates with 6 factors at less than 1000 levels and not to exceed 1500 combinations.
4. Multiple Range Tests
(BMD 07V). An among-within analysis of variance and comparison of means of up to 100 groups of 20,000 or less data each.

Analysis of these types will be made on LMFBR cladding data when sufficient data are available to Battelle-Northwest, or they can be obtained from off-site laboratories.

It is planned to use results obtained from statistical analysis of lot-to-lot variations to aid in the preparation of future manufacturing specifications. These results also can be used to evaluate validity of mathematical models for cladding

deformation and/or fuel swelling restraint capabilities of the cladding during reactor service.

RADIATION EFFECTS ON METALS -

05 PROGRAM

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ELECTRON MICROSCOPY

The presence of cavities in nickel irradiated at elevated temperatures was described in the last Quarterly Progress Report (BNWL-790). The postirradiation annealing behavior of the cavities has been studied in detail. Bulk samples were annealed isothermally for 2, 20 and 96 hr at 600 °C in vacuum. Figure 12.23 is a plot showing the size distribution of the cavities as a function of the annealing time. The average size increases with annealing time, while the density decreases as indicated in the figure. The volume fraction remains constant during annealing within experimental accuracy. These observations indicate that the smaller cavities are breaking up and the vacancies are migrating to the surrounding cavities. An inhomogeneous distribution of cavities adjacent to grain boundaries is noted after annealing. Figure 12.24 shows the denuded zone to be much wider on one side of the boundary. This could be explained by grain boundary migration sweeping up the cavities during annealing. Annealing at a higher temperature

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FIGURE 12.23. Change in the Void Size Distribution as a Function of Annealing Time at an Annealing Temperature of 600 °C

of 700 °C for 2 hr produces a very inhomogeneous distribution of the cavities within the foils. After annealing at 800 °C for 2 hr, the cavities completely disappear. The inhomogeneous distribution formed during high temperature annealing may or may not be a result of the boundary migration. This phenomenon is being studied further.

Annealing *in situ* in the electron microscope has shown that cavities disappear in the temperature range 700 to 800 °C. It was noted that the cavities disappear very rapidly when they are very small. No evi-

dence of transformation to a dislocation loop was observed.

Nickel has also been irradiated at ambient temperatures (~ 50 °C) to $4 \times 10^{19} \text{ n/cm}^2$ and annealed at elevated temperatures. A high density of dislocation loops is observed in the as-irradiated material, Figure 12.25a. Annealing at 500 °C produces large dislocation loops identified as interstitial type and small black spots tentatively assumed to be vacancy type defects, Figure 12.25b. Measurements show that the total area of the black spots is approximately equal to the

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FIGURE 12.24. Nickel Irradiated at 400 °C to 5×10^{19} n/cm² and Annealed 2 hr at 600 °C

area of the large loops. This is consistent if the black spots are vacancy type defects and the large loops are interstitial. Annealing at 600 °C reduces the damage and only a few dislocation loops remain, Figure 12.25c. No cavities were

ever observed in this material after the postirradiation annealing. These observations will be the subject of further investigations. Particular emphasis will be placed on the role of boundaries during postirradiation annealing.

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(a) As-Irradiated

Neg 5832 E

(b) 6 hr at 500 °C

Neg 6004A

(c) 2 hr at 600 °C

FIGURE 12.25. Nickel Irradiated at ~ 50 °C to 4×10^{19} n/cm² and Annealed at Temperatures and Times Indicated

SOME OBSERVATIONS ON THE ANNEALING
OF NEUTRON-IRRADIATED SINGLE-CRYSTAL
AND POLYCRYSTALLINE MOLYBDENUM

Introduction

After irradiation with fast neutrons near room temperature, molybdenum shows an increase in lattice parameter with a decrease in density,⁽²⁹⁾ a higher resistivity,⁽³⁰⁾ and an increased hardness.⁽³¹⁾ Electron microscopy reveals defect clusters in the form of black spots or small loops which interact at sufficiently high fluences to form extended dislocation networks.⁽³²⁾

Annealing of the neutron-irradiated molybdenum in the temperature range 150 to 250 °C, about 0.15 of the absolute melting point, results in the recovery of an appreciable fraction of the irradiation-induced changes and the evolution of stored energy. This annealing stage is commonly known as "Stage III" from its analogy to a similar annealing stage in fcc metals.⁽³³⁾ The defects responsible for this annealing stage and the mechanisms whereby they interact are not completely understood. While several models for Stage III annealing have been proposed,^(30,33,34) none satisfactorily accounts for all the phenomena observed.

The work described in this paper was undertaken to reexamine the Stage III annealing behavior of molybdenum by simultaneous lattice parameter and length change measurements, together with stored energy release measurements, during annealing of high purity samples.

Single-crystals produced by multiple zone-refining of selected high-purity stock was the source of the very pure sample material used in the studies and allowed very precise measurements of lattice parameter. Parallel experiments which utilized samples of the high purity stock without the zone refining treatments provided data on the effects of zone refining on the accumulation of irradiation-induced defects and their subsequent annealing.

Experimental Procedure

The single crystals used in this work were prepared from 3.2 mm diameter sintered rod obtained from the Lamp Metals and Components Division, General Electric Company. The techniques employed in zone-refining have been described earlier.⁽³⁵⁾ Analyses of the samples before and after zone-refining are given in Table 12.7. Most crystals were seeded to a $[14\bar{4}]$ axis orientation during the final pass. After three-pass zone-refining, the crystals were cut into 38 mm lengths and the blunt ends were finished into rounded points by grinding, followed by heavy electropolishing. A (011) metallographically prepared face parallel to the rod axis facilitated subsequent lattice parameter measurements. Residual cold work was again removed by heavy electropolishing followed by annealing at 1300 °C. This treatment has been found to produce single crystals that have a high degree of crystalline perfection.⁽³⁵⁾ Polycrystalline rod samples were processed

TABLE 12.7. Analysis of Zone-Refined Single Crystals of Molybdenum

Element	Vendor's Analysis	Before Zone Refining	After Zone Refining	Gas Fusion
		BNW (Mass Spectograph)	BNW (Mass Spectograph)	
W	100	60	60	
Fe	10	13	2	
Ni	<10	2	<2	
K	-	6	40	
Si	10	10	12	
C	<10	9	3	12.8
H ₂	1-2	-	-	
O ₂	6-11	-	-	3-3.5
N ₂	4-8	-	-	1.2

Note: Impurities in parts per million by weight

in the same manner, except that a flat face was not utilized.

Single-crystal lattice parameters were measured by a symmetrical back-reflection technique. Lattice parameters for polycrystalline rods were measured by conventional high-angle diffractometry with extrapolation to $2\theta = 180^\circ$. Lengths were measured by the gage-block comparator method. These procedures are described in detail elsewhere.⁽³⁵⁾ The estimated precision of all length measurements and single-crystal lattice parameters was 0.001%; for polycrystalline lattice parameters, it was 0.003%.

Stored-energy measurements were made in a calorimeter designed specifically for small single-crystal specimens. Three samples, each 3.2 mm in diameter and 38 mm long, were attached to three junctions of a chromel-alumel thermophile. The electromotive force (emf) generated by this thermopile was opposed by

that from a similar thermopile to which were attached three reference samples, identical except for the irradiation treatment. The differential emf was amplified and recorded. A bank of quartz infrared lamps controlled by an electronic programmer comprised the heat source. The heating rate employed in these experiments was 2 °C/min. The melting of measured amounts of lead in hollow molybdenum samples provided a means for calibrating the heat evolved as a function of the amplified differential emf. The combined weight of the three samples was about 10 g.

Electron microscopy was performed on sections cut from selected single-crystal and polycrystalline rods. These sections were thinned by standard electropolishing techniques.⁽³²⁾

For each irradiation, six specimens were housed in cylindrical

cavities in a helium-filled aluminum capsule. All capsules were irradiated at 40 °C in a Snout Facility in a Hanford reactor.

Results and Discussion

Both the single-crystal rods and the polycrystalline rods showed changes in length and lattice parameter as a result of irradiation, as shown in Figure 12.26. The length change $\Delta L/L$ approached a saturation value with increasing fluence, while the lattice parameter disclosed an initial increase and then a decrease with increasing fluence. The two sample types differed, however, in that the relative lattice parameter increase of the polycrystalline rods was greater than the relative length increase for exposures up to $4 \times 10^{19} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$), while the relative lattice parameter increase for the single crystals was less than the relative length increase for all the fluences studied.

Electron microscopy of the irradiated samples also showed differences between single-crystal and polycrystalline molybdenum after irradiation. Micrographs of the two sample types are compared in Figure 12.27. In both cases, the irradiation-induced defects appear as black spots. These defect clusters, or small dislocation loops, believed to be interstitial in nature,⁽³⁶⁾ are smaller (50 to 60 Å) in the polycrystalline samples as compared

to 90 to 100 Å in the single crystals after a neutron fluence of $1 \times 10^{19} \text{ n/cm}^2$. The defect configuration is presumed to consist of molybdenum interstitials which exist as clusters and as isolated defects in traps of varying strengths, as well as vacancy defects which exist as isolated defects or very small clusters.

The greater lattice parameter change and the smaller clustered defects observed in the polycrystalline samples, as compared to similarly irradiated single-crystal samples, leads to a tentative

FIGURE 12.26. Changes in Length $\Delta L/L$ and Lattice Parameter $\Delta a/a$ for Neutron-Irradiated Single-Crystal and Polycrystalline Molybdenum

FIGURE 12.27. Transmission Electron Micrographs of (a) Single-Crystal and (b) Polycrystalline Molybdenum Showing Differences in Defect Cluster Sizes After Exposure to a Fast Neutron Fluence of 1×10^{19} n/cm²

conclusion that a much higher concentration of isolated interstitials exists in the polycrystalline samples. At the irradiation temperature, 40 °C, interstitial defects are mobile. The high isolated interstitial concentration must therefore be a result of some trapping process.

The results of annealing studies and stored energy release measurements further confirmed the existence of dissimilar defect configurations in the two sample types.

In Figure 12.28, typical isochronal annealing data to 900 °C are shown for a single crystal rod and a polycrystalline rod. The as-irradiated length changes $\Delta L/L$ were identical for the two samples and length-change recovery with annealing was very similar. Recovery of lattice parameter changes were, however, quite different up to 400 °C. A partial recovery of the lattice expansion took place between 100 and 200 °C in the irradiated polycrystalline rod, but

FIGURE 12.28. Recovery of Length and Lattice Parameter During Isochronal Annealing of Neutron-Irradiated Single-Crystal and Polycrystalline Molybdenum. Annealing Time Was 2 hr at Each Temperature.

no such recovery was observed in the single-crystal rod. Stored-energy-release measurements showed a comparatively large release of stored energy at 150 to 200 °C upon heating the irradiated polycrystalline rods, while the single-crystals showed but a small release. Typical stored energy release curves are shown in Figure 12.29. The lattice parameter recovery and the stored energy release in the polycrystalline samples are apparently directly related.

From the equation given by Simmons and Balluffi,⁽³⁷⁾

$$C_V - C_I = 3(\Delta L/L - \Delta a/a)$$

where C_V and C_I are the concentrations of single vacancies and interstitials, respectively, it is apparent that the interstitial concentration exceeds the vacancy concentration in the irradiated polycrystalline samples, while the opposite is true in the single crystals. The difference $\Delta L/L - \Delta a/a$ does not remain constant upon annealing, so the annealing is not entirely a result of mutual annihilation of isolated defects. From the similarity of the length change annealing data and the completely dissimilar nature of the lattice parameter annealing below 400 °C, it follows that the lattice parameters and lengths anneal independently of one another. From the annealing curve of the polycrystalline sample irradiated to 1×10^{19} n/cm² of Figure 12.29, and from the Simmons and Balluffi relationship, it can be shown that the concentration of excess interstitials is reduced from 3.3×10^{-4} to almost zero with the evolution of 0.48 cal/g, which is equivalent to the evolution of 1.25×10^{19} eV/g during the loss of 2.1×10^{18} defects/g. This evolution of 6 eV per defect is probably too low for mutual annihilation of isolated vacancies and interstitials, but is compatible with the expected energy evolved by an interstitial upon absorption at some alternative sink such as an interstitial cluster. Because some length change recovery occurs also, some mutual annihilation may be indicated.

FIGURE 12.29. Release of Stored-Energy from Irradiated Single-Crystal and Polycrystal Molybdenum

The same analysis applied to the single-crystal annealing data shows that about 0.1 cal/g, or 2.5×10^{18} eV/g, is evolved by the loss of 1.8×10^{18} excess vacancies/g, or roughly 1.4 eV per vacancy. This is almost certainly the result of vacancies moving to vacancy clusters or to sinks other than interstitials. In the single-crystal annealing data shown in Figure 12.28, an increase in unit cell volume of 1.05×10^{-4} is observed to occur simultaneously with the length change recovery. This is equivalent to 0.37 atomic volume per vacancy if the interstitial concentration is assumed to be un-

changed. Prior estimates⁽³⁸⁾ have placed ΔV_v , the relaxation about a vacancy in cubic metals, in this range. The length change is not completely recovered at 400 °C, so that only a fraction of the total vacancy defects are involved. The fraction annealed out represents one-fourth to one-third of the total, a fraction which corresponds fairly closely to the relative concentration of divacancies in irradiated iron calculated by Beeler.⁽³⁹⁾ If these calculations apply to other bcc metals as well, the low temperature length change annealing could result from the annealing out of the comparatively mobile divacancy

complex, leaving the less mobile vacancy configurations in the crystal lattice.

The present analyses have not been extended to samples exposed to neutron fluences higher than 1×10^{19} n/cm². After 1×10^{19} n/cm² ($E > 1$ MeV), it appears that post-irradiation annealing of single crystals and polycrystalline samples involves appreciably different recovery processes. Some characteristic of polycrystalline samples must inhibit the clustering of interstitial defects. Whether the interstitials are trapped at impurities or are perturbed by differences in crystal perfection is a question not readily answered at present. The difference in purity after zone-refining is slight (Table 12.7) and it is difficult to ascribe the observed annealing behavior to this cause. The distribution of impurities, however, could well be altered by the zone-refining treatment. The impurities which normally segregate at grain boundaries or dislocations would of necessity remain dispersed or form a unique complex in the single crystals.

Experiments intended to extend and clarify the above results are in progress. For example, the effect of carbon and iron on the normal behavior of irradiated molybdenum single crystals is being investigated. In another series of experiments, irradiated polycrystalline rods, prepared from zone-refined crystals by torsional deformation alternated with annealing for very

short periods, will be compared to single-crystal samples. The results of these experiments, which are currently in progress, should do much to explain the inconsistent annealing behavior of single-crystal and polycrystalline molybdenum after neutron irradiation.

Summary

Single-crystal rods and polycrystalline rods of molybdenum, exposed to high-energy neutron irradiation at 40 °C, exhibit very dissimilar annealing behavior at temperatures below 400 °C. The polycrystalline samples exhibit simultaneously a large lattice parameter recovery and stored energy release at 150 to 200 °C, while length change recovery is relatively slight. Single crystals, on the other hand, show almost no lattice parameter recovery and very little evolution of stored energy in the same temperature range. The interstitial defects and vacancies appear to anneal independently without appreciable mutual annihilation. Accordingly, the so-called Stage III annealing in irradiated molybdenum may consist of both reactions proceeding simultaneously.

From the behavior of the interstitial defects in the polycrystalline samples, a higher concentration of trapping sites is postulated. Whether these are a result of differences in impurity content or a difference in the distribution of impurities is not clear. Additional

irradiation experiments, on carbon-doped and iron-doped single-crystals and on polycrystalline rods made from zone-refined single crystals, are underway. Results of these experiments should make possible a more definite assignment of recovery mechanisms in neutron irradiated molybdenum.

ATR GAS LOOP OPERATION AND MAINTENANCE

J. E. Minor

ATR GAS LOOP SUPPORT

Several GCL equipment and manufacturing problems were referred to PNL for evaluation and recommendation. Some of the major problems are as follows:

- Fabrication of an enclosure was recommended to resolve a leakage problem in the Transfer Facility cooling coil.
- A process for joining 303 SS to 304 SS was developed, and the components were shipped to PNL for brazing.
- Recommendations for welding stainless steel pipe adjacent to metallic foil insulation have been successfully bench-tested, and the process will be used on the In-Reactor tube by the vendor.

Vendor data received to date have been reviewed, and appropriate comments have been transmitted to cognizant agencies.

PNL, in continuation of responsibility as inspection agency for the analytical instrument panel, has followed development of the processes

for welding 1/8 in. diam tubing. Acceptable welds have been produced, but further development is required to establish the settings for the weld parameters which will consistently yield acceptable welds. The 303 SS valves modified by PNL were received by Montek, and work on the larger diameter tubing is progressing. The diffusion cell system is now ready for helium leak testing.

Efforts to determine affects of forging on Hastelloy-X are continuing. Research completed to date indicates that a severe directionality problem exists. We have reviewed several of the forgings to be used on the regenerative heat exchanger for the ATR Gas Loop, and preparations are being made to inform the vendor as to manufacturing processes so that the maximum strength and elongation in the maximum stress intensity direction of the material are guaranteed.

Circulator lead-unit tests in the Gas Cooled Loop were successfully completed the week of June 17, with disassembly and inspection now in progress. Portions of the test program were witnessed by PNL personnel. All requested tests were completed, and performance under all conditions tested is extremely good. The circulator system is expected to meet or exceed all functional requirements of the Gas Cooled Loop.

ATR WATER LOOP SUPPORT

The Advanced Test Reactor startup is being delayed until a suitable evacuation plan is approved by the

AEC. Consequently, no progress is being made to further water chemistry testing in the North pressurized water loop. One more test is planned in which oxygen concentration will be controlled at about 1 ppm when the reactor reaches 100 MW. So far, test results show that oxygen can be controlled in the loop in the absence of neutrons.

Flux and gamma heat data were generated during initial ATR Core I testing. These data are being distributed to personnel at Battelle Northwest who need them for designing planned experiments in the North pressurized water loop. Planned experiments for the ATR Water Loop for Cores II and III are now fabricated and ready for insertion. Subsequent experiments are planned; they are being designed and fabricated for Cycle I (Core IV) and beyond.

Spare parts for the pressurized water loop were ordered and are being received. An inventory of the pressurized water loop equipment has been turned over to Battelle Northwest. The list of items has been checked and all items have been tagged with an AEC property tag and number. However, several items were not included in the inventory, and checks are being made with property accounting people to determine appropriate action.

A program proposal was submitted to the AEC by Battelle for the purpose of identifying various irradiation tests for the pressurized water loop. The proposal was to irradiate structural materials, fuel cladding,

and fuels under the program designated as BNW-3. Each test will be identified as BNW-3-1, 2, 3, etc. The proposal is in the final stages of being approved by INC, AEC, and BNW.

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FRACTURE MECHANICS EVALUATION OF REACTOR VESSEL STEELS

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An experimental program directed towards radiation effects on pressure vessel material is being performed. Experimental testing of a group of reactor vessel steels is being performed to investigate the application of the fracture mechanics technique to these materials in both the unirradiated and irradiated conditions.

SPECIMEN FABRICATION

The specimens required for the preirradiation portion of the program were fabricated from the 12 in. thick plate of A533 Grade B Class I (Mn-Mo with Ni) steel. Specimens were machined from the top surface, 1/4 thickness, 1/2 thickness, 3/4 thickness and bottom surface of the 12 in. plate. Tensiles, Charpy-V notch, drop weight, 1X WOL, 1/2T, 1T and 2T Compact K_{IC} specimens were machined from each of the five locations through the thickness of the plate. The required 1X WOL's, 1/2T, 1T and 2T Compact K_{IC} specimens were fatigue cracked prior to the initiation of the test program.

MECHANICAL TESTING

The preirradiation testing of the tensile, Charpy-V notch and drop weight specimens fabricated from the 12 in. thick plate of A533 Grade B Class 1 steel was completed this quarter. The tensile properties

were evaluated from -250 to +600 °F. The results are summarized in Figures 13.1 through 13.5 and Table 13.1. As expected, the two surfaces (top and bottom) exhibit much higher yield strength than the interior of the plate.

Charpy-V notch and drop weight test specimens from the 12 in. thick plate were tested at various temperatures. Test data are shown in Figure 13.6 through 13.10 (energy versus temperature), Figures 13.11 through 13.15 (per cent brittle fracture and lateral expansion versus temperature) and Tables 13.2 through 13.6. The drop weight data are summarized in Table 13.7. Again, the NDT determined both by Charpy-V notch and the drop weight specimens is correlated with the yield strength, low at the top and bottom near zero through the remainder of the plate. The 30 ft-lb fix Charpy energy is in good agreement with the drop weight data.

Fracture toughness testing was initiated utilizing 1X WOL, 1T and 2T Compact K_{IC} specimens fabricated from the 12 in. thick plate. The stress intensity factors (fracture toughness) for the 12 in. thick plate are summarized in Tables 13.8 through 13.12. As discussed in previous quarterly reports pertaining to fracture mechanics, the requirements of the type of fracture

FIGURE 13.1. Preirradiation Tensile Properties of 12 in. Thick Plate of A533 Grade B Class I Steel - Top Surface

FIGURE 13.2. Preirradiation Tensile Properties of 12 in. Thick Plate of A533 Grade B Class I Steel - Quarter Thickness

FIGURE 13.3. Preirradiation Tensile Properties of 12 in. Thick Plate of A533 Grade B Class I Steel - Half Thickness

FIGURE 13.4. Preirradiation Tensile Properties of 12 in. Thick Plate of A533 Grade B Class I Steel - Three Quarter Thickness

FIGURE 13.5. Preirradiation Tensile Properties of 12 in. Thick Plate of A533 Grade B Class I Steel - Bottom Surface

toughness testing employed are unusual in that it is necessary to compare post-test data to specimen pre-test dimensions to determine if the fracture toughness measured (K_Q) is the critical value (K_{IC}) for the material. In other words, validity of the test can be determined only after testing. Two criteria for validity have been proposed.

1. Reference 1 proposes use of the secant off-set method to determine that the majority of strain is due to slow-crack growth.
2. Reference 2 proposes that both the crack length and specimen thickness exceed $2.5 \times (\text{Fracture Toughness}/\text{Strength})^2$. Fracture toughness measured for specimen

meeting these criteria is considered to be the critical value (K_{IC}) for the material. If the criteria are not met, the measured fracture toughness (K_Q) is considered to have been biased by specimen size.

Fracture toughness of both the top and bottom surfaces of the 12 in. thick plate appears to turn upward rather sharply as the temperature approaches NDT. For the center and quarter thicknesses of the plate, the approach to NDT appears to involve a more gradual slope. The phenomenon of a sharp transition of the fracture toughness for a given material as the temperature approaches NDT is being investigated utilizing heavy section specimens

TABLE 13.1. Preirradiation Tensile Properties of 12 in. A533
Grade B Class I Steel Heat No. 1195-1, Plate 2

Specimen Number	Test Temp, °F	Yield Point, psi	Tensile Strength, psi	Elongation, %	Reduction of Area, %
Top Surface					
T1	-250	132,050	134,900	23.1	58.7
T2	-250	127,700	137,700	20.0	58.0
T3	-150	103,250	119,150	21.1	61.7
T4	-150	105,950	118,300	22.1	64.3
T5	75	80,900	97,550	20.7	66.5
T6	75	82,450	97,450	20.5	71.9
T7	300	74,750	88,050	16.7	71.8
T8	300	75,350	87,150	17.6	70.5
T9	600	68,050	91,600	16.1	66.5
T10	600	67,850	92,800	17.3	69.1
Top Quarter Thickness					
1Q1	-250	114,300	119,650	5.9	7.6
1Q2	-250	111,400	124,850	19.7	50.9
1Q3	-150	91,250	112,000	21.8	62.1
1Q4	-150	91,400	112,450	21.1	60.3
1Q5	75	71,850	91,250	18.4	66.1
1Q6	75	69,100	91,150	16.6	62.8
1Q7	300	63,350	83,150	15.6	66.9
1Q8	300	63,550	83,350	17.0	69.2
1Q9	600	59,100*	85,900	16.4	66.3
1Q10	600	58,500*	86,350	16.6	61.9
Center Thickness					
C1	-250	116,900	128,300	6.5	7.9
C2	-250	113,750	128,550	18.9	53.4
C3	-150	90,500	110,750	18.9	56.3
C4	-150	95,150	110,150	18.9	52.0
C5	75	67,650	92,050	19.1	63.4
C6	75	68,400	91,600	17.4	61.8
C7	300	64,150	83,150	15.2	65.7
C8	300	62,950	83,350	16.0	64.3
C9	600	59,950*	86,150*	16.1	59.4
C10	600	59,750*	86,050*	14.1	49.5
Bottom Quarter Thickness					
3Q1	-250	113,600	125,550	21.9	59.0
3Q2	-250	116,100	128,100	21.4	60.5
3Q3	-150	88,100	110,000	18.6	60.8
3Q4	-150	93,850	111,300	19.6	59.8
3Q5	75	72,600	91,150	17.9	65.1
3Q6	75	72,150	91,950	19.1	68.2
3Q7	300	63,550	82,950	15.7	70.2
3Q8	300	63,850	81,300	16.4	70.2
3Q9	600	59,550*	86,250	16.5	64.7
3Q10	600	59,450*	84,200	16.1	66.1
Bottom Surface					
B1	-250	127,200	137,550	19.6	57.9
B2	-250	128,250	134,000	21.2	59.0
B3	-150	104,100	118,750	21.3	63.1
B4	-150	99,800	118,900	21.8	64.7
B5	75	88,000	97,550	20.0	71.5
B6	75	85,300	97,950	18.3	71.0
B7	300	74,650	88,700	17.3	71.0
B8	300	75,050	88,300	17.1	71.0
B9	600	68,250*	87,150	17.5	67.2
B10	600	69,050	92,350	17.5	60.6

* Indicates 0.2% Offset Yield Strength

FIGURE 13.6. Preirradiation Charpy-V Notch Energy as a Function of Temperature - Top Surface

FIGURE 13.7. Preirradiation Charpy-V Notch Energy as a Function of Temperature - Quarter Thickness

FIGURE 13.8. Preirradiation Charpy-V Notch Energy as a Function of Temperature - Half Thickness

FIGURE 13.9. Preirradiation Charpy-V Notch Energy as a Function of Temperature - Three Quarter Thickness

FIGURE 13.10. Preirradiation Charpy-V Notch Energy as a Function of Temperature - Bottom Surface

FIGURE 13.11. Preirradiation Charpy-V Notch Lateral Expansion and Brittle Fracture as a Function of Temperature - Top Surface

FIGURE 13.12. Preirradiation Charpy-V Notch Lateral Expansion and Brittle Fracture as a Function of Temperature - Quarter Thickness

FIGURE 13.13. Preirradiation Charpy-V Notch Lateral Expansion and Brittle Fracture as a Function of Temperature - Half Thickness

FIGURE 13.14. Preirradiation Charpy-V Notch Lateral Expansion and Brittle Fracture as a Function of Temperature - Three Quarter Thickness

FIGURE 13.15. Preirradiation Charpy-V Notch Lateral Expansion and Brittle Fracture as a Function of Temperature - Bottom Surface

TABLE 13.2. Preirradiation Charpy-V Notch Impact Data for 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

TOP SURFACE				
Specimen Number	Test Temp, °F	Impact Strength, ft-lbs	Fracture Appearance, (% brittle fracture)	Lateral Expansion, mils
T43	-140	14	95	1
T44		10	95	2
T45		10	95	4
T46	-120	34	85	22
T47		27	80	20
T48		37	80	27
T40	-100	35	80	25
T41		40	80	30
T42		43	80	31
T37	- 60	82	40	56
T38		88	30	60
T39		55	60	42
T5	- 10	75	50	50
T7		101	15	67
T9		117	0	76
T10	+ 10	115	0	74
T12		111	0	71
T15		108	0	70
T17	+ 30	111	0	74
T18		114	0	74
T19		113	0	72
T20	+ 60	116	0	72
T21		112	0	73
T22		112	0	74
T23	+110	114	0	74
T25		116	0	80
T27		117	0	75
T31	+160	126	0	80
T33		125	0	79
T35		120	0	77

TABLE 13.3. Preirradiation Charpy-V Notch Impact Data for 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

TOP QUARTER THICKNESS				
Specimen Number	Test Temp, °F	Impact Strength, ft-lbs	Fracture Appearance, (% brittle fracture)	Lateral Expansion, mils
IQ1	- 35	9	95	6
IQ2		11	95	8
IQ6		11	95	8
IQ8	- 10	33	95	25
IQ9		20	95	15
IQ10		15	95	11
IQ34		45	85	32
IQ36		39	85	28
IQ11	+ 10	35	90	25
IQ12		35	85	25
IQ14		27	85	21
IQ16	+ 30	36	80	26
IQ19		46	75	34
IQ20		46	80	33
IQ21	+ 60	58	65	42
IQ22		58	70	41
IQ24		53	70	39
IQ25	+110	90	30	64
IQ27		95	30	69
IQ28		110	25	78
IQ35		108	25	75
IQ30	+160	124	0	80
IQ31		118	10	84
IQ32		127	0	84

TABLE 13.4. Preirradiation Charpy-V Notch Impact Data
for 12 in. A533 Grade B Class I Steel Heat No. 1195-1
Plate 2

Specimen Number	Test Temp, °F	CENTER THICKNESS		
		Impact Strength, ft-lbs	Fracture Appearance, % brittle fracture	Lateral Expansion, mils
C1	- 35	50	65	35
C2		41	75	29
C4		19	85	15
C36		15	90	9
C6	- 10	30	80	23
C9		35	80	25
C12		20	85	16
C37		22	85	17
C13	+ 10	38	70	38
C14		37	80	27
C15		36	80	27
C16	+ 30	40	80	30
C18		35	80	26
C20		45	80	33
C21	+ 60	61	55	44
C24		51	70	38
C25		54	65	41
C26	+110	83	30	62
C27		100	25	71
C28		83	35	61
C38		90	30	65
C32	+160	126	2	82
C34		120	1	84
C35		119	10	80

TABLE 13.5. Preirradiation Charpy-V Notch Impact Data
for 12 in. A533 Grade B Class I Steel Heat No. 1195-1
Plate 2

Specimen Number	Test Temp, °F	THREE QUARTER THICKNESS		
		Impact Strength, ft-lbs	Fracture Appearance, % brittle fracture	Lateral Expansion, mils
3Q1	- 35	9	95	6
3Q2		10	95	6
3Q3		14	95	9
3Q4	- 10	14	90	10
3Q5		17	95	13
3Q6		18	95	14
3Q7	+ 10	30	85	23
3Q8		30	85	23
3Q9		33	90	24
3Q10	+ 30	46	75	34
3Q11		46	80	33
3Q12		37	80	29
3Q13	+ 60	55	70	38
3Q14		38	70	28
3Q15		43	75	33
3Q22		50	70	34
3Q16	+110	88	40	62
3Q17		85	40	60
3Q18		77	35	56
3Q19	+160	119	0	76
3Q20		119	0	79
3Q21		127	0	75

TABLE 13.6. Preirradiation Charpy-V Notch Impact Data
for 12 in. A533 Grade B Class I Steel Heat No. 1195-1
Plate 2

BOTTOM SURFACE

Specimen Number	Test Temp, °F	Impact Strength, ft-lbs	Fracture Appearance, % brittle fracture	Lateral Expansion, mils
B28	-140	23	90	14
B29		23	90	11
B30		22	90	14
B31	-120	38	85	25
B32		31	85	23
B33		24	85	17
B25	-100	51	65	33
B26		40	75	25
B27		34	70	23
B22	- 60	89	20	58
B23		76	30	52
B24		87	15	59
B1	- 35	101	0	66
B2		107	0	71
B3		107	0	74
B4	- 10	104	0	67
B5		112	0	69
B6		109	0	70
B7	+ 10	99	0	63
B8		111	0	72
B9		108	0	80
B10	+ 30	110	0	71
B11		112	0	73
B12		110	0	70
B13	+ 60	112	0	74
B14		105	0	71
B15		113	0	68
B16	+110	111	0	73
B17		119	0	81
B18		115	0	77
B19	+160	117	0	75
B20		125	0	77
B21		121	0	76

TABLE 13.7. Drop Weight Test Data for 12 in.
Thick Plate A533 Grade B Class I Steel -
Heat No. 1195-1 Plate 2

Specimen Length	5 in.
Specimen Width	2 in.
Specimen Thickness	5/8 in.
Support Span	4 in.
Stop Height	0.075 in.
Weight of Head	60 lb
Drop Weight	5 ft

TOP SURFACE

<u>No.</u>	<u>Temp, °F</u>	<u>Result</u>	<u>N.D.T.</u>
D2-T-1	0	Not Broken	
D2-T-2	- 40	Not Broken	
D2-T-3	- 80	Not Broken	
D2-T-4	-120	Broken	
D2-T-5	-100	Not Broken	
D2-T-6	-110	Broken	
D2-T-7	-100	Broken	
D2-T-8	- 90	Broken	-90 °F*

TOP QUARTER THICKNESS

D1-1Q-1	0	Not Broken	
D1-1Q-2	- 20	Broken	
D1-1Q-3	- 10	Broken	
D1-1Q-4	0	Not Broken	-10 °F

CENTER THICKNESS

D2-C-1	0	Not Broken	
D2-C-2	- 40	Broken	
D2-C-3	- 20	Broken	
D2-C-4	- 10	Broken	
D2-C-5	0	Broken	
D2-C-6	+ 10	Not Broken	
D2-C-7	+ 10	Not Broken	0 °F

THREE QUARTER THICKNESS

D1-3Q-1	0	Broken	
D1-3Q-2	+ 20	Not Broken	
D1-3Q-3	+ 10	Not Broken	
D1-3Q-4	+ 10	Not Broken	0 °F
D2-B-1	0	Not Broken	
D2-B-2	- 40	Not Broken	
D2-B-3	- 80	Not Broken	
D2-B-4	-120	Broken	
D2-B-5	-100	Broken	
D2-B-6	- 90	Broken	
D2-B-7	- 80	Broken	
D2-B-8	- 70	Broken	-60 °F*

* Tentative N.D.T. - Additional tests in progress

TABLE 13.8. Results of Fracture Toughness Tests of 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

TOP SURFACE								
Specimen Number	Size	Temp, °F	Y.S., ksi	a, in.	a/W	Plastic Zone Size r, in.	Stress Intensity Factor K_{IC} , K_Q (ksi√in.)	Validity*
W-1T10	1X	-250	131.0	0.533	0.474	0.005	39.2	1
W-1T7	1X	-250	131.0	0.508	0.452	0.008	52.0	1
K3-1T10	1T	-250	131.0	1.023	0.511	0.002	26.5	1
W-1T11	1X	-200	115.0	0.548	0.487	0.009	48.3	1
W-1T9	1X	-200	115.0	0.533	0.474	0.020	70.1	4
W-1T12	1X	-200	115.0	0.545	0.484	0.010	50.9	1
K3-1T5	1T	-200	115.0	1.022	0.511	0.010	50.1	1
K3-1T8	1T	-200	115.0	0.999	0.499	0.006	40.1	1
W-1T1	1X	-175	107.0	0.558	0.496	0.014	54.3	2
K3-1T9	1T	-175	107.0	1.011	0.506	0.010	47.2	1
W-1T13	1X	-150	104.0	0.539	0.479	0.018	60.1	4
K3-1T11	1T	-150	104.0	1.054	0.527	0.019	62.5	1
K3-1T6	1T	-150	104.0	1.018	0.518	0.015	54.9	1
K3-1T12	1T	-150	104.0	1.009	0.509	0.015	55.5	1
K2-2T5	2T	-150	104.0	2.041	0.510	0.011	51.0	1
K3-1T7	1T	-125	100.0	1.021	0.511	0.031	76.2	2
K1-2T3	2T	-100	97.0	2.029	0.507	0.028	70.0	1
K1-2T1	2T	-100	97.0	2.050	0.513	0.036	79.4	1
K2-2T8	2T	-100	97.0	2.200	0.550	0.078	117.3	4
K2-2T6	2T	-50	90.0	2.031	0.508	0.065	99.5	2

- * Notes: 1. Meets both criteria: a and B greater than $2.5 (K_{IC}/Y.S.)^2$ and secant offset
 2. Fails a and B greater than $2.5 (K_{IC}/Y.S.)^2$ meets secant offset
 3. Meets a and B greater than $2.5 (K_{IC}/Y.S.)^2$ fails secant offset
 4. Fails both criteria

TABLE 13.9. Results of Fracture Toughness Tests of 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

TOP QUARTER THICKNESS								
Specimen Number	Size	Temp, °F	Y.S., ksi	a, in.	a/W	Plastic Zone Size r, in.	Stress Intensity Factor K_{IC} , K_Q (ksi√in.)	Validity*
W-1Q12	1X	-200	99.0	0.523	0.465	0.008	37.5	1
W-1Q7	1X	-200	99.0	0.514	0.457	0.005	30.5	1
K6-1Q38	1T	-200	99.0	1.011	0.506	0.005	31.2	1
K6-1Q43	1T	-175	95.0	1.081	0.541	0.011	43.5	1
W-1Q2	1X	-150	90.0	0.530	0.471	0.007	31.5	1
W-1Q10	1X	-150	90.0	0.536	0.476	0.018	52.0	2
W-1Q1	1X	-150	90.0	0.521	0.463	0.011	41.2	1
W-1Q13	1X	-125	87.0	0.506	0.450	0.011	39.1	1
K6-1Q42	1T	-125	87.0	1.025	0.513	0.013	43.4	1
K6-1Q44	1T	-125	87.0	1.008	0.504	0.010	38.1	1
K6-1Q46	1T	-125	87.0	1.034	0.517	0.016	48.3	1
W-1Q11	1X	-100	85.0	0.593	0.527	0.028	61.8	2
K6-1Q45	1T	-100	85.0	1.008	0.504	0.025	58.1	2
K6-1Q41	1T	-100	85.0	1.061	0.530	0.031	65.4	2
K5-1Q16	2T	-100	85.0	2.034	0.509	0.026	59.0	1
K5-1Q17	2T	-100	85.0	2.029	0.507	0.023	56.2	1
K6-1Q47	1T	-75	83.0	1.027	0.514	0.022	53.6	1
K4-1Q12	2T	-50	81.0	2.035	0.509	0.037	67.5	1
K6-1Q20	2T	-50	81.0	2.010	0.503	0.034	65.0	1
K5-1Q18	2T	-10	78.0	2.091	0.523	0.041	68.9	1
K4-1Q13	2T	-10	78.0	2.169	0.542	0.068	88.3	4

- * Notes: 1. Meets both criteria: a and B greater than $2.5 (K_{IC}/Y.S.)^2$ and secant offset
 2. Fails a and B greater than $2.5 (K_{IC}/Y.S.)^2$ meets secant offset
 3. Meets a and B greater than $2.5 (K_{IC}/Y.S.)^2$ fails secant offset
 4. Fails both criteria

TABLE 13.10. Results of Fracture Toughness Tests of 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

Specimen Number	Size	Temp, °F	Y.S., in.	a, in.	a/W	Plastic Zone Size r, in.	Stress Intensity Factor K_{IC} , K_Q (ksi $\sqrt{\text{in.}}$)	Validity*
W1-C7	1X	-250	113.0	0.520	0.462	0.006	37.3	1
W1-C10	1X	-200	93.0	0.555	0.493	0.012	44.0	1
W1-C11	1X	-200	93.0	0.564	0.501	0.007	34.6	1
W1-C1	1X	-200	93.0	0.646	0.574	0.010	39.9	1
K3-1C16	1T	-200	93.0	1.030	0.515	0.008	35.1	1
W-1C13	1X	-150	84.0	0.541	0.481	0.016	46.8	2
W-1C9	1X	-150	84.0	0.543	0.483	0.015	44.1	2
K3-1C22	1T	-150	84.0	1.025	0.513	0.011	37.7	1
K3-1C20	1T	-150	84.0	1.028	0.514	0.013	42.1	1
K3-1C24	1T	-100	79.0	1.010	0.505	0.020	48.5	1
K3-1C18	1T	-100	79.0	1.010	0.505	0.020	48.5	1
K3-1C23	1T	-100	79.0	1.018	0.509	0.015	42.2	1
K1-2C4	2T	-100	79.0	2.091	0.523	0.021	50.2	1
K3-1C19	1T	-75	77.0	1.023	0.512	0.023	50.3	1
K3-1C21	1T	-75	77.0	1.028	0.514	0.037	64.3	2
K2-2C6	2T	-50	74.0	2.036	0.509	0.041	65.0	1
K1-2C1	2T	-50	74.0	2.041	0.510	0.061	79.2	2
K1-2C2	2T	-50	74.0	2.023	0.506	0.041	65.1	1
K2-2C8	2T	-25	74.0	2.041	0.510	0.076	88.4	2
K1-2C3	2T	-25	74.0	2.043	0.511	0.074	87.3	2
K2-2C5	2T	0	73.0	2.024	0.506	0.065	81.0	2

- * Notes: 1. Meets both criteria: a and B greater than $2.5 (K_{IC}/Y.S.)^2$ and secant offset
 2. Fails a and B greater than $2.5 (K_{IC}/Y.S.)^2$ meets secant offset
 3. Meets a and B greater than $2.5 (K_{IC}/Y.S.)^2$ fails secant offset
 4. Fails both criteria

TABLE 13.11. Results of Fracture Toughness Tests of 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

BOTTOM QUARTER								
Specimen Number	Size	Temp, °F	Y.S., in.	a, in.	a/W	Plastic Zone Size r, in.	Stress Intensity Factor K_{IC} , K_Q (ksi $\sqrt{\text{in.}}$)	Validity*
W-3Q4	1X	-200	98.0	0.626	0.556	0.009	41.0	1
W-3Q8	1X	-200	98.0	0.548	0.487	0.008	38.5	1
K6-3Q51	1T	-200	98.0	1.016	0.508	0.005	30.8	1
W-3Q5	1X	-150	92.0	0.529	0.470	0.006	30.5	1
W-3Q3	1X	-150	92.0	0.516	0.459	0.006	29.7	1
W-3Q6	1X	-150	92.0	0.566	0.503	0.015	48.4	2
K6-3Q50	1T	-150	92.0	1.018	0.509	0.010	40.7	1
W-3Q2	1X	-100	87.0	0.608	0.540	0.023	57.3	2
K6-3Q52	1T	-100	87.0	1.049	0.525	0.021	54.8	1
K6-3Q55	1T	-100	87.0	1.030	0.515	0.021	54.4	1
K4-3Q12	2T	-100	87.0	2.081	0.508	0.018	51.3	1
K5-3Q19	2T	-100	87.0	2.065	0.516	0.019	51.4	1
K6-3Q54	1T	-75	85.0	1.003	0.502	0.016	46.6	1
K6-3Q56	1T	-75	85.0	1.038	0.519	0.023	56.3	1
K6-3Q53	1T	-50	83.0	1.044	0.522	0.032	64.5	4
K6-3Q49	1T	-50	83.0	1.016	0.508	0.029	61.6	4
K4-3Q13	2T	-50	83.0	2.074	0.519	0.025	56.7	1
K4-3Q14	2T	0	79.0	2.026	0.507	0.037	66.4	1
K5-3Q17	2T	0	79.0	2.046	0.512	0.075	93.7	4
K5-3Q18	2T	0	79.0	2.040	0.510	0.059	83.4	4

- * Notes: 1. Meets both criteria: a and B greater than $2.5 (K_{IC}/Y.S.)^2$ and secant offset
 2. Fails a and B greater than $2.5 (K_{IC}/Y.S.)^2$ meets secant offset
 3. Meets a and B greater than $2.5 (K_{IC}/Y.S.)^2$ meets secant offset
 4. Fails both criteria

TABLE 13.12. Results of Fracture Toughness Test of 12 in. A533 Grade B Class I Steel Heat No. 1195-1 Plate 2

Specimen Number	Size	Temp, °F	Y.S., in.	a, in.	a/W	BOTTOM SURFACE		Validity*
						Plastic Zone Size r, in.	Stress Intensity Factor K_{IC} , K_Q (ksi $\sqrt{\text{in.}}$)	
W-1B2	1X	-250	128.0	0.534	0.475	0.006	44.3	1
W-1B3	1X	-250	128.0	0.570	0.507	0.006	41.4	1
W-1B9	1X	-200	109.0	0.534	0.475	0.012	51.0	1
W-1B4	1X	-200	109.0	0.561	0.499	0.018	63.9	2
W-1B8	1X	-200	109.0	0.546	0.485	0.007	38.3	1
K3-1B28	1T	-200	109.0	1.023	0.512	0.016	59.5	1
K3-1B31	1T	-200	109.0	1.060	0.530	0.011	50.4	1
K3-1B29	1T	-200	109.0	1.015	0.508	0.014	56.0	1
W-1B6	1X	-150	102.0	0.583	0.518	0.026	71.3	4
W-1B5	1X	-150	102.0	0.541	0.481	0.023	67.3	2
K3-1B32	1T	-150	102.0	1.020	0.510	0.014	51.5	1
K3-1B25	1T	-150	102.0	1.024	0.512	0.027	73.0	2
K3-1B33	1T	-150	102.0	1.015	0.508	0.023	66.5	1
K3-2B11	2T	-150	102.0	2.031	0.508	0.018	60.2	1
K3-1B30	1T	-125	101.0	1.015	0.508	0.048	96.0	4
K3-1B27	1T	-100	98.0	1.009	0.505	0.046	90.8	4
K2-2B7	2T	-100	98.0	2.023	0.506	0.017	55.2	1
K1-2B2	2T	-100	98.0	2.005	0.501	0.021	61.5	1
K1-2B3	2T	-100	98.0	2.028	0.507	0.024	66.3	1
K1-2B4	2T	-100	98.0	2.024	0.506	0.036	80.5	1
K2-2B5	2T	-50	94.0	2.045	0.511	0.058	98.1	4
K2-2B8	2T	-50	94.0	2.530	0.633	0.082	116.9	4

- * Notes: 1. Meets both criteria: a and B greater than $2.5 (K_{IC}/Y.S.)^2$ and secant offset
 2. Fails a and B greater than $2.5 (K_{IC}/Y.S.)^2$ meets secant offset
 3. Meets a and B greater than $2.5 (K_{IC}/Y.S.)^2$ fails secant offset
 4. Fails both criteria

(4 to 12 in. thick). This information is being reported as part of the HSST program.

IRRADIATION EXPERIMENT

The irradiation of the second set of three capsules in the Babcock and Wilcox Test Reactor (BAWTR) is continuing. The total exposure accumulated by the capsules was 82 EFPD or about 60% of the target exposure. During BAWTR outing in July, the capsules will be rotated 180 degrees to homogenize the fast neutron exposure through the specimen thickness.

The testing of the irradiated Charpy-V notch specimens from the

European forging grade steel, the 8 in. thick plate of A533 Grade B Class I steel, and the weldment (both weld and HAZ) was initiated at Westinghouse's Post Irradiation Laboratory at Waltz Mill during this report period. Since the test data are incomplete, the results will not be presented graphically.

The incomplete set of data are summarized in Table 13.13. The material had a total exposure of 132 EFPD or an estimated fluence of 1.3×10^{19} . The dosimeter used for monitoring the exposure of the first three capsules is presently undergoing analytical evaluation at the Postirradiation Laboratory.

TABLE 13.13. Postirradiation Charpy-V Notch Impact Data

Specimen Number	Test Temp, °F	Impact Strength, ft-lbs	Capsule Number
A533 Grade B Class I - 8 In. Thick Plate			
L16	80	9.0	2
L23	150	14.0	3
L14	150	16.0	2
L10	200	20.5	2
L26	200	25.9	3
L15	225	25.0	2
L19	250	28.0	3
L28	250	31.0	3
L12	250	39.0	2
L18	350	40.0	3
L5	350	48.0	2
European Forging Grade Steel			
N5	80	12.5	2
N9	80	17.9	2
N14	80	38.0	2
N25	125	41.0	3
N8	150	17.0	2
N31	150	59.0	3
N10	175	37.0	2
N12	200	59.0	2
N24	350	67.0	3
N7	350	88.0	2
Weld, (A533 Grade B Class I)			
PW4	150	23.0	1
PW5	200	30.0	1
PW6	200	36.0	1
PW2	275	40.0	1
HAZ, (A533 Grade B Class I)			
PH6	-40	18.0	1
PH7	-25	23.0	1
PH2	0	50.0	1
PH4	32	44.0	1
PH3	80	56.0	1
PH5	150	110.0	1
A302 B Reference Steel			
R2	150	8.0	1
R5	150	8.0	3
R3	150	25.0	2
R6	275	26.0	3
R4	275	37.0	2
R1	275	61.0	1

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1. J. E. Srawley. Unpublished Data. January 2, 1967, (Notes Pertaining to Section 7.3.2 of Draft Recommended Practice, "Interpretation of Test Record and Calculation of K_{IC} ").
2. Unpublished Data. ASTM, E24 Sub I Task Group, December 14, 1966, ("Recommended Practice for Plane-Strain Fracture Toughness Testing of High Strength Metallic Materials Using a Fatigue-Cracked Bend Specimen," draft of proposed practice).

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