AEC
RESEARCH
and
DEVELOPMENT
REPORT

BNWL-79

C 50

**QUARTERLY PROGRESS REPORT
METALLURGY RESEARCH OPERATION
JANUARY, FEBRUARY, MARCH, 1965**

APRIL 15, 1965

NAME	PROJECT	STATUS	DATE
Extra	33069	3702	
G. B. Ruscher	33069	3702

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For The

U. S. ATOMIC ENERGY COMMISSION Under Contract No. AT(45-1)-1830

PRINTED BY/FOR THE U. S. ATOMIC ENERGY COMMISSION

BNWL-79
UC-25, Metals, Ceramics,
and Materials

QUARTERLY PROGRESS REPORT
METALLURGY RESEARCH OPERATION
JANUARY, FEBRUARY, MARCH, 1965

FIRST UNRESTRICTED
DISTRIBUTION MADE JUN 22 '65

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April 15, 1965

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

Printed in USA. Price \$ 4.00. Available from the
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards
U. S. Department of Commerce
Springfield, Virginia

Previous reports in this series:

HW-76228	October, November, December, 1962
HW-77052	January, February, March, 1963
HW-77954	April, May, June, 1963
HW-78962	July, August, September, 1963
HW-79766	October, November, December, 1963
HW-81269	January, February, March, 1964
HW-82651	April, May, June, 1964
HW-84281	July, August, September, 1964
HW-84573	October, November, December, 1964

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SUMMARYPHYSICAL METALLURGYRADIATION EFFECTS ON METALS

Molybdenum irradiated at 60° C to 10^{20} nvt ($E > 1$ Mev) has been studied by transmission electron microscopy. After annealing at 750°C, a larger number of small spots and loops is present in the molybdenum containing <10 ppm C, but the average size of loops is greater in the molybdenum containing approximately 450 ppm C. After annealing at 1000°C, spot and loop defects disappear and stable configurations of dislocation nodes identified as $\frac{a}{2}$ <111> and a <100> form.

Very small defect clusters, 35Å in diameter, have been detected in rhenium, a cph metal, after an exposure of 7×10^{18} nvt ($E > 1$ Mev) at 60° C. Channeling of dislocations occurs during straining of irradiated rhenium.

Four methods have been considered for determining foil thickness for electron transmission microscopy applications. With present facilities, experimental difficulties appear too great for successful application.

Single crystal rods of molybdenum irradiated to 7×10^{18} nvt ($E > 1$ Mev) have been subjected to annealing treatments. After a 4 hr anneal at 180° C, approximately 10% recovery of macroscopic length occurs, and no recovery of lattice parameter occurs. After a 14 hr anneal at 200°C, 60% recovery of macroscopic length occurs, and only 0.003% recovery of lattice parameter occurs. Vacancies are the migrating defects responsible for the change.

Rhenium foils, irradiated to 7×10^{18} nvt ($E > 1$ Mev), show unit cell changes: c_0 decreases and a_0 increases. The net unit cell volume increase in rhenium is of the same magnitude as that in molybdenum for equal exposures. The lattice parameter change due to irradiation is recovered completely by annealing at 800°C.

Analysis of thermally-activated deformation in unirradiated and irradiated polycrystalline molybdenum between 223 and 350° K has proceeded.

It would appear that in unirradiated molybdenum the results are more consistent with motion of jogs in screw dislocations than with the Peierls mechanism.

SWELLING OF IRRADIATED FISSIONABLE MATERIALS

Specimens from the first pressure-temperature controlled swelling capsule irradiated to 0.13 at. % BU at 1000 psi and at 450° C have been examined in the Radiometallurgy Laboratory. Although the microstructure had assumed the "worked-swirled" structure typical of this irradiation temperature, microtearing was virtually eliminated in the high purity, the U + Fe-Al, and the U + Fe-Si specimens.

The second such irradiations, at a pressure of 1000 psi and a temperature of 575° C, have been successfully completed and postirradiation studies are in progress. Preliminary indications are that pressure at this temperature also significantly reduces swelling.

A capsule containing 2.88 at. % enriched, high purity uranium in several heat-treated conditions has been discharged after reaching goal exposure and will provide data on the effect of burnup rate on swelling mechanisms.

PLUTONIUM PHYSICAL METALLURGY

The total strain accompanying the $\beta \rightarrow \alpha$ transformation of plutonium has been investigated as a function of tensile stresses and transformation rates. Some of the observed strain is ascribed to excess creep strain, particularly during the early stage of the transformation.

The total strain accompanying the $\alpha \rightarrow \beta$ transformation has also been studied as a function of tensile stresses and transformation rates. Excess creep strain occurs primarily during the later stages of the transformation.

The temperature for the beginning of the $\beta \rightarrow \alpha$ transformation is increased by both compressive and tensile loading.

Specimens having a [010] rod axis texture undergo a 90° shift in fiber texture during an alpha-beta-alpha transformation cycle. Application of tension during the $\alpha \rightarrow \beta$ transformation and compression during the $\beta \rightarrow \alpha$

transformation enhances the development of a fiber texture in plutonium. Very little micro-cracking occurred in specimens transformed under these conditions of external stress and slow transformation rates.

Microstructural patterns and anisotropic geometry changes in textured specimens subject to $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations have been considered and interpreted as conforming with martensitic transformations.

DAMAGE MECHANISMS

The possibility that the initial yield point and pseudo yield points observed in iron after strain rate changes are introduced by transient machine effects has been eliminated; the effects are real. An equation currently under development is being applied to irradiated iron and should predict the magnitude of the effective stress under certain testing conditions. After an exposure of 5×10^{18} nvt, bulk iron specimens contain no defect clusters normally visible as spots or loops by transmission electron microscopy.

REACTOR METALS RESEARCH

ALLOY SELECTION

Under the alloy selection portion of the Irradiation Effects on Reactor Metals program several nickel base alloys are being irradiated and tested at ambient and elevated temperatures to determine the effects of irradiation and environment upon their mechanical properties. During the past quarter, specimens of Hastelloy N, Hastelloy X-280, Incoloy 800, Inconel 600, and Inconel 625 were tested at room temperature and 650° C.

Specimens of Hastelloy N irradiated at 280 C and tested at room temperature showed increases in strength and slight decreases in ductility properties. Those specimens tested at 650° C showed marked decreases in ductility. Similar results were found for specimens of Hastelloy X-280 irradiated under the same conditions. Additional specimens of Hastelloy X-280 irradiated at 740° C and tested at 650° C showed a severe decrease in ductility of the alloy.

Specimens of Incoloy 800 and Inconel 600 were irradiated in the mill annealed, 20% cold worked, and solution treated conditions and tested at

650° C. Substantial differences in the postirradiation properties were found with marked decreases in ductility common to all specimens. For both materials, specimens in the solution treated conditions maintained the best ductility characteristics but were substantially lower in strength. Additional specimens of Inconel 600 were irradiated at 280° C and tested at 650° C. Results from these tests showed a considerable increase in strength and ductility at an exposure of 5.68×10^{20} nvt.

To determine the effects of irradiation upon the mechanical properties of Inconel 625, specimens were irradiated at 50, 280, 580, and 740° C and tested at room temperature and 650° C. The room temperature test results for specimens irradiated at 50 and 280° C show some increase in strength with small decrease in ductility. Specimens irradiated at 50° C showed the greater effect. The effects of irradiation temperature upon the room temperature properties of irradiated material show a large decrease in ductility in materials irradiated above 580° C. Similarly, specimens irradiated at 280° C and tested at 650° C show large decreases in both strength and ductility at exposures of 1.46×10^{21} nvt.

A liquid metal irradiation capsule has been designed and a laboratory bench model constructed to determine heat transfer and other operational characteristics of the capsule.

IN-REACTOR MEASUREMENTS OF MECHANICAL PROPERTIES

The objective of the in-reactor measurement program is to define the effects of fast neutron irradiation on the mechanical properties of structural materials.

The temperature and stress dependency of the in-reactor creep of annealed 304 stainless steel are presently being investigated.

In-reactor creep tests at 650° C, 10,000 psi, and 525° C, 30,000 psi were started and a test at 550 C, 30,000 psi was concluded during the past quarter.

Reduced rupture life and ductility at failure were observed for the in-reactor 550° C, 30,000 psi creep test as compared to the out-of-reactor

test. The in-reactor creep strain at failure was 3 to 4% with a failure time of 760 hr. The out-of-reactor test, still in progress, has accumulated 3500 hr of test time with a creep strain of 4.8%. The minimum creep rate for the in-reactor test was 2.6×10^{-5} /hr. The out-of-reactor creep test exhibited a creep rate 9.3×10^{-6} /hr.

The 650° C, 10,000 psi in-reactor creep test yielded a creep rate of 4.4×10^{-6} /hr after 800 hr of testing while the out-of-reactor control test showed a rate of about 2.2×10^{-6} /hr. Significant creep rates have not yet been determined for the 525° C, 30,000 psi as it is still in the primary creep stage.

The metallographic investigation being conducted on the recovered 304 stainless steel in-reactor creep specimen is partially completed. Photomicrographs of transverse sections from one buttonhead and from near the fracture show no unusual features when compared to the unirradiated material. Longitudinal sections from the area near the fracture and the buttonheads will be made soon.

A second in-reactor test capsule has been discharged from the reactor and sent to the Radiometallurgy facility.

With the objective of expanding present testing capabilities, a new facility has been developed in the Hanford Plutonium Recycle Test Reactor.

During the past quarter, modification of an in-reactor creep capsule for use in the PRTR has been completed. The same capsule currently being used has been modified to include an outside cooling chamber since testing in the PRTR necessitates operating in a thimble tube inserted into a dry fuel channel. Facilities installation has also been completed at the PRTR and the capsule will be charged during the next outage.

The capsule contains an annealed Zircaloy-2 specimen. The test conditions will be 300° C and 10,000 psi stress.

IRRADIATION FACILITIES OPERATION

With the conclusion of ETR Cycles 69 and 70, over 180 test specimens have been discharged from the ETR. Materials irradiated at 50 and 290°C include Zircaloy-2, nickel-base alloys, and stainless steels. Over 150 control specimens were discharged from the out-of-reactor loop upon completion of Cycle 67.

Tensile tests have been conducted on the BONUS transition joint specimens irradiated in the G-7 loop to exposures between 2.0 and 2.5×10^{20} nvt (> 1 Mev). Specimens exposed in the out-of-reactor loop and in the as-received condition have also been tested. Tests were performed at 550°F and at room temperature on specimens in each condition.

Preliminary results indicate there is no change in properties due to exposure in the out-of-reactor loop. Tests on the irradiated samples also resulted in failures entirely through the Zircaloy-2 portion of the specimen. These failures were of an extremely "brittle" nature for both the room temperature and 550°F tests. Uniform elongations of less than 1% and total elongations of less than 2% were observed. In some tests it was impossible to measure any plastic deformation.

Metallographic studies are in progress to determine the nature and cause of this type of behavior. Two additional quadrants are scheduled to be tested after exposures of approximately 5.0×10^{20} nvt (> 1 Mev). It is concluded from the results of these tests that the integrity of both types of joints is acceptable. However, the joining processes and/or prior fabricating steps have resulted in very low ductility in the Zircaloy-2. This low ductility is further reduced by nominal neutron exposures and should be considered of prime importance in developing these joints.

IRRADIATION DAMAGE TO NICKEL-BASE ALLOYS

The nickel-base alloys hold much promise for high-temperature nuclear reactor application. However, they do suffer embrittlement when irradiated at high temperature. An effort to modify their microstructure by pre-irradiation treatments to reduce this effect is continuing.

Previous work on Hastelloy X-280 led to the belief that a preirradiation treatment which would give controlled precipitation of carbides within the matrix with a concomitant reduction of grain boundary deposits would be worthy of investigation. Initial attempts, using a combination of 26% cold work and aging treatments, showed successful dispersion of the carbide phase though with ductility loss. Tensile and stress-rupture specimens representing treatment using four lesser levels of cold work are now being irradiated at 540° F to goal exposures of approximately 2×10^{20} nvt. Others are being prepared for 120 F and 1300 F irradiations.

Irradiation of an alloy containing a precipitated phase may result in solution of the precipitates, formation of additional precipitates, or the disintegration of precipitates into small pieces by a combination of these two processes. Specimens of Inconel X-750 specimens representing five different heat treatments are undergoing irradiation at 540° F to goal exposures of approximately 2×10^{20} nvt, with others being prepared for 120 and 1300° F irradiations. Micrographs of the microstructures developed by these treatments show that the desired range of precipitate aging (from underaged to overaged) was obtained. The effects of irradiation on the tensile and stress-rupture properties of irradiated material in these several conditions will be investigated.

A program of pre- and postfabrication quality control tests for specimen manufacture was initiated. The specimens mentioned above underwent these tests. The program involves ultrasonic screening of the stock material and visual, optical comparator, zyglow dye, and ultrasonic testing of the fabricated specimen. During initial tests, variation in ultrasonic attenuation was found between batches of stock material of Inconel X-750. Metallographic examination showed this to be due to grain size differences and inhomogeneity. It is possible, however, to select fine grain, homogeneous material from the stock using this technique.

FRACTURE STUDIES

Eight annealed Zircaloy-2 double cantilever beam (DCB) specimens have been charged into the ETR G-7 hot water loop. These specimens will

be used to determine the effects of hot water irradiations on the fracture toughness and transition temperature for four exposure levels and two orientations. As has been pointed out in previous quarterly reports, the property of the DCB specimen is such that 30 to 40 measurements of the fracture toughness may be obtained from a single specimen.

As was mentioned in the previous quarterly report (HW-84573) one of the difficulties encountered in testing the DCB specimen at temperatures other than room temperature is that of measuring the temperature at the crack tip. This problem arises as the crack tip changes position down the length of the specimen as testing progresses. A cryostat which satisfies both the temperature control and measurement problem has recently been built. The temperature of the specimen is lowered by passing liquid nitrogen through channels in special chill plates held in contact against the sides of the specimen by springs. Counteracting this, a special conducting paint is applied to the other side of the plate. This paint acts as a 3000 W heater. The temperature at the crack tip is measured by one of seven thermocouples spaced 3/4 in. apart and mounted in one of the plates. Based on the compliance of the specimen, one can make a quick calculation as to the position of the crack and select the closest thermocouple for control. By using solid state control and power units, temperature control to $\pm 0.4^\circ\text{C}$ has been obtained throughout the temperature range -196 to 170°C .

Since interest in the DCB specimen has been developing for only a short time, there still exists a lack of knowledge concerning the proper design of the specimen and the effect that changes in design might have on the measured fracture toughness. For this reason experiments are now underway to ascertain the effect of varying the specimen thickness, depth, groove depth, and groove tip radius on the compliance and fracture toughness.

CHEMICAL METALLURGY

ATR GAS LOOP SUPPORT

The rate of evaporation of Haynes Alloy 25 can be effectively decreased by an overpressure of an inert or reducing gas. Static tests run

at temperatures above 1100°C using hydrogen to inhibit the evaporation of Haynes Alloy 25 have shown that an overpressure of ~80 torr hydrogen will essentially stop metal evaporation.

The oxidation of pure niobium at temperatures of 800, 900, and 1000°C in continuously replenished oxygen atmospheres of 1 to 8 μ has been studied. Two points were made from these tests. The first is the affinity of niobium for oxygen at low oxygen pressures. The second is the role of oxygen transport in controlling the initial reaction kinetics.

Samples of Hastelloy X, Haynes Alloy 25, and Cb-1 Zr have been exposed to high pressure, high velocity, Grade A helium at 2050°F. The oxidation rates found are comparable to those obtained in static systems, indicating that gas velocity to 525 fps has little if any effect on oxidation kinetics. The Cb-1 Zr alloy showed greater resistance initially to oxide spallation than the superalloys.

Initial evaluation of the rate of in-leakage of air into a pressurized helium system has been made under ATR gas loop operating conditions using the dynamic materials test apparatus as the test facility. During 5 days of operation O₂ increased by 1 ppm while 25% of the helium had escaped through mechanical seals, etc., in the system.

Intersite analyses of a standard helium-H₂, O₂, N₂, CH₄, CO gas mixture were in generally good agreement. Substantially greater peak heights were obtained with the BNW ionization detector chromatograph than with conventional thermal conductivity chromatographs.

N-REACTOR CORROSION AND HYDRIDING

Inhibition of gas phase hydriding of Zircaloy by water vapor at low temperatures is not effective when the metal surface has been mechanically sensitized. During a 4 month exposure to H₂ + 3% H₂O, hydride penetrations up to 9 mils (100°C), and 50 mils (200°C) were observed.

PRODUCTION REACTOR CORROSION AND COATING STUDIES

Mild steel chain 80% "black" to neutrons has been successfully produced by pack-boridizing the chain in Ferroboration for 9 hr at 1160°C or 12 hr at 1140°C.

MATERIALS ENGINEERING

GAS LOOP DEVELOPMENT

Conductivity tests were run on the foil insulation using the new cooling water jacket that was installed on the test section of the model gas loop. The new tests showed a decrease in the effect of flow, and indicated that the leakage flow around the insulation may be the primary cause of the flow dependency. The insulation was removed and repaired, and a new set of tests have been started. The loop has been thermal cycled 70 times between room temperature and various heater outlet temperatures up to 2125° F. The gas bearing compressor has been started 100 times, and has accumulated 400 hr of operation.

A new model gas loop heater has been designed to incorporate and investigate essential details of a compact two-stage heater presently being ordered for the ATR Gas Loop. Components for the model heater are now being fabricated on-site.

A bypass purification system utilizing pressure drop available from the gas bearing compressor was installed. Purity levels of less than 0.1 ppm for all species except water, which was 3.5 ppm, were achieved at a loop temperature of 916° C (1680° F). A larger, dual bypass purification system will be installed.

Testing of thermal conductivity has begun on Hastelloy-X felted wire being considered by the Architect Engineer for insulation on 1600 F ATR Gas Loop piping. Conventional methods are being used to measure thermal conductivity of 5 in. OD by 3/4 in. wall cylindrical test piece.

The results of bend and tensile tests of the weld metals and base metals aged 500 hr suggest that Hastelloy W has better aging characteristics than Hastelloy-X when used as filler wire for joining Hastelloy X-280 to AISI 316 stainless steel and Haynes 25. This suggestion is based on considerations of the weld metal yield stress being larger than the base metals yield stresses, the relative sluggishness of weld metal aging and the amount of useful ductility remaining in the weld metal and the base metals after an

aging treatment. The tensile testing of the weld and base metals aged 1000 hr and greater than 1000 hr is continuing.

PRTR PRESSURE TUBES

A transition from ductile to brittle crack propagation of unirradiated PRTR Zircaloy-2 pressure tube specimens occurs at a temperature between 70 and 200° F. This transition temperature for irradiated specimens appears to be above 200° F. At room temperature, the stress for propagating a 1 in. long crack has been indicated to be 11,500 psi greater for a fast neutron exposure of 1.5×10^{21} nvt than for a fast neutron exposure of 2×10^{20} to 5×10^{20} nvt or than for unirradiated specimens.

K-Reactor Welded Zircaloy-2 Process Tubing is described in report BNWC-51.

Irradiation Burst Test Facility is described in report BNWL-6 entitled "Irradiated Tube Testing Facility."

USA-AECL COOPERATIVE PROGRAM

About 75 ft of test material in the 30% CW and 60% CW tempers has been purchased. Burst tests at room temperature and 300° C have been made.

PROCESS TUBE DEVELOPMENT

A Zircaloy-2 C-overbore process tube was removed from C-Reactor after 2 yr use. Postirradiation examination indicated no radiation effects on structure or grain size. Ring tensile tests indicated an increase of 14% in ultimate strength.

N-REACTOR TUBE

Accelerometers and piezoelectric strain gages are under investigation as sensors in the acoustical detection of crack growth in pressure tubing subjected to cyclic pressures. The technique is expected to be useful in extracting the maximum amount of crack propagation information from a single test specimen. The frequency response of the strain gage has been shown to extend into the megacycle range assuring that signals emanating from a growing crack will be detected.

PHYSICAL METALLURGY

RADIATION EFFECTS ON METALS - T. K. Bierlein, J. L. Brimhall, H. E. Kissinger, J. J. Laidler, B. Mastel, and K. R. Merckx

This program is directed toward establishing the combined effect of impurities and neutron irradiation on the properties and structure of specific metals, and deducing from thermally-activated recovery processes how the damage state can be altered. The major effort of present studies has been concentrated on single and polycrystalline specimens of molybdenum and the minor effort has been directed toward nickel and rhenium.

Transmission Electron Microscopy

Transmission electron microscopy of molybdenum samples containing <10 ppm and 450 ppm carbon which have been irradiated to 10^{20} nvt ($E > 1$ Mev) has continued. Recovery processes of the irradiated material are currently being studied by postirradiation annealing. The microstructures of the low- and high-carbon doped molybdenum show both similarities and differences after annealing at 750°C for 2 hr. Loops and spots are present in both types of material. The density of the smaller loops and spots is greater in the low carbon samples. The average size of the loops is greater in the high carbon doped samples which also show the presence of dislocation nodes after annealing. Although no difference was discernable in the microstructure of the as-irradiated samples, the annealing behavior indicated that the loops formed during irradiation grow larger in the high-carbon molybdenum. This observation is consistent with those reported for samples annealed at similar temperatures but irradiated to lower exposures.

After annealing at 1000°C for 2 hr, the microstructure of the low- and high-carbon doped molybdenum is similar in all respects. Figure 3.1 shows the microstructure of molybdenum containing 450 ppm carbon after annealing at 1000°C for 2 hr. The same area is shown with three different g vectors giving contrast. Using standard techniques, the Burgers vectors of the dislocations comprising the nodes were determined. They were of the type $\frac{a}{2} \langle 111 \rangle$ and a $\langle 100 \rangle$ as expected. Dislocation nodes are stable

FIGURE 3.1

Dislocation Formed in Molybdenum Containing 450 ppm Carbon Irradiated to 10^{20} nvt and Annealed at 1000 C for 2 hr
(Shown is the same area with three different reflections, (a) (020), (b) (301), and (c) $(\bar{1}01)$, Giving Contrast: Burgers Vectors of Dislocations Comprising the Modes are Indicated

configurations and may be responsible for the retention of radiation damage after high temperature annealing.

A paper entitled "Combined Effect of Carbon and Neutron Radiation on Molybdenum" has been submitted to Acta Metallurgical for publication. The study of slip lines on irradiated molybdenum single crystals has been concluded and a paper entitled "Effect of Neutron Irradiation on the Slip Lines on Molybdenum" has been submitted for publication to Trans. AIME.

Radiation damage in rhenium, a CPH metal, is being studied in a manner analogous to that of molybdenum, a BCC metal. Rhenium foils have been irradiated at 60°C to an exposure of 7×10^{18} nvt ($E > 1$ Mev) and examined by transmission electron microscopy. Defect clusters in the form of dark spots having diameters of approximately 35 Å were observed. The density of spots is approximately 4×10^{16} /cc if a sample thickness of 750 Å is assumed. Figure 3.2 shows the defect clusters and a band which is relatively free of spots. Presumably, dislocations have moved through this region and absorbed the spot defects. The large loops in the channel probably result from dipole formations of the moving dislocation and are not the result of irradiation. The nature of the spots has not been established. Because of the low vacancy mobility in rhenium, it is presumed that the spots are interstitial clusters or irresolvable interstitial-type dislocation loops. A note entitled "Effect of Neutron Irradiation on Rhenium," which describes these observations has been submitted for publication in the Philosophical Magazine. Higher neutron exposures and postirradiation annealing should provide data to establish the type and character of the cluster defects.

Foil Thickness Determination by Electron Diffraction

The determination of foil thickness for electron microscopy specimens was investigated. Plots of transmitted and diffracted electron intensity as functions of foil thickness and deviation from the Bragg diffraction condition were made to correlate measured and theoretical variations in electron intensity. The measurements of deviations from the Bragg condition can be determined with the separation of the diffracted beam and the Kikuchi line in the image of the focal plane. A description of this method was written. ⁽¹⁾

(1) K. R. Merckx. Use of Kikuchi Lines in Determining Deviation from Bragg Diffraction, BNSA-144. March 10, 1965.

FIGURE 3. 2

150, 000 X

Defect Clusters in Rhenium
Irradiated to 7×10^{18} nvt ($E > 1$ Mev)

Neg. 3027A

Four methods of determining foil thickness were investigated:

(1) Kossel-Mollenstedt fringe separation for divergent incident beams, (2) thickness contour separations in wedge shaped crystals, (3) separation of fringes from bent foils with images of the plane containing the center of curvature, and (4) variation of transmitted intensity with foil rotation. The measurements of intensities and control of foil rotation could not be made accurately enough for the fourth method to be applied. The third method requires the determination of the location of the plane containing the center of curvature and a foil with uniform bending. The second method is limited to foils which are not so thick as to have inelastic scattering destroy the beat type extinction due to preferential absorption of the elastically scattered waves. Experimental difficulties are found in obtaining a sufficiently divergent incident beam necessary in the first method without overheating the foil. Thus, the application of each method to foil thickness determinations appears to be severely limited.

X-Ray Diffraction

Single crystal rods of molybdenum have been irradiated to 7×10^{18} nvt ($E > 1$ Mev) for length change and lattice parameter measurements. As described in a previous report (HW-84573), a length increase of about 0.024% and a lattice parameter increase of 0.016% were measured after irradiation. Low-temperature annealing experiments have now been completed on these samples. Prolonged annealing at 160°C did not result in a measurable length change. At 180°C, approximately 10% of the irradiation-induced length increase is recovered after 4 hr, with no further recovery on prolonged annealing. Upon annealing at 200°C, 60% of the increase was recovered after 14 hr, with no further recovery observed.

Lattice parameters were measured after completion of the annealing. No change was observed after 180°C anneal, and only a small change, 0.003%, after 200°C.

Calculations of point defect density from these data by the method of Baluffi and Simmons⁽¹⁾ discloses an excess of vacancy defects over interstitials

(1) R. W. Baluffi and R. O. Simmons. Journal of Applied Physics, vol. 31, p. 2284. 1960.

before annealing, and an excess of interstitial sites after annealing at 200°C. This result indicates that vacancies are the principal defect responsible for the low temperature annealing of irradiated molybdenum. Preliminary measurements on irradiated polycrystalline foils show that the recovery of lattice parameter is greater for molybdenum with the higher impurity content. These experiments will be continued to further define the role of impurity atoms in annealing of point defects.

X-ray diffraction examination of irradiated rhenium foils has been initiated. After an exposure of 7×10^{18} nvt ($E > 1$ Mev) the c_o axis of this hexagonal metal is smaller by about 0.05% while the a_o axis is greater by 0.07%. The net volume increase observed was about 0.09%, which is of the same magnitude as the volume increase in molybdenum at the same exposure. Apparently the type and rate of accumulation of defects are very similar in the two metals, but differences in the defect configuration can be expected as a result of the difference in crystal structure. Preliminary annealing experiments indicate little or no change in lattice parameter after 100 and 200°C anneals. All irradiation-induced lattice parameter change is recovered after an 800°C annealing treatment. This behavior is analogous to that of molybdenum, and supports the conclusion that the two metals are subject to neutron damage in a very similar manner.

Mechanical Properties

Experiments on thermally-activated deformation in unirradiated and irradiated polycrystalline molybdenum have nearly reached conclusion, although a great deal of interpretative work remains to be done. These experiments were conducted over the temperature range 223 to 350°K, the lower temperature being dictated by the inherent brittleness of this material, and the upper temperature by the minor effect of thermal activation above 350°K. While a discussion of the detailed conclusions is premature at this time, a few general observations can be made. At the present, there seems to be little evidence to justify selection of the Peierls mechanism for the controlling process in the unirradiated material at the temperatures investigated. Rather, the results are more consistent with the motion of jogs in screw dislocations. Neutron irradiation results in a significant change in the flow

properties at low strains, but very little change at higher strains (e. g., $> 15\%$), at which the deformed substructure is thought to consist of a stable cellular network of dislocations. This observation concurs with the hypothesis that small point defect clusters interact strongly with glissile dislocations and are swept up by these moving dislocations. Evidence also exists that the dislocation multiplication rate in irradiated molybdenum annealed at 600°C is at least an order of magnitude greater than in the as-irradiated condition. This has been interpreted as the bowing out of loop segments from clusters which have collapsed to form small dislocation loops. Further correlation and interpretation of the data are expected to confirm these tentative conclusions.

SWELLING OF IRRADIATED FISSIONABLE MATERIALS - R. D. Leggett,

H. A. Taylor, B. Mastel, and J. L. Brimhall

When fissionable metals are irradiated, the ordered metallic structure is perturbed in a number of ways. High energy neutrons and fission fragments collide with metal atoms, knocking them out of their structural positions. The resultant defects, vacancies, interstitials, dislocation loops, etc., can adversely affect metallic properties. Similarly, the fission products generated, some of which are noble gases, may be incompatible with the structure and precipitate or coalesce as second phase inclusions or gas pores. Furthermore, the localized high temperatures associated with the fission event can cause anisotropic fissionable metals such as uranium to change shape drastically in accordance with the crystallography of the specimens. These effects and their interactions are being studied with respect to the influence of such variables as burnup, burnup rate, irradiation temperature and pressure, metallurgical history, and specimen composition and geometry. The primary emphasis has been on determining the basic processes responsible for the behavior of uranium irradiated in the high alpha phase temperature range, where swelling is known to be important. Fundamental information developed by this program will provide a basis for the engineering exploitation of fissionable fuel materials in advanced reactor applications.

Irradiation Program

The second controlled pressure-temperature swelling capsule (P-3) was discharged from a reactor after reaching goal exposure and was disassembled in the Radiometallurgy Laboratory. The capsule operated at a pressure of 1000 psi and temperature of 575° C, regardless of the operating state of the reactor. This capsule contained a total of ten specimens representing several metallurgical states: six high purity uranium specimens, two U + 400 Fe-640 Al specimens, and two U + 140 Fe-95 Si specimens. Each specimen is half tubular in shape, being machined from a hollow cylinder 5/8 in. long by 1/2 in. OD by 0.030 in. wall thickness.

A third pressurized capsule (P-4) was charged into a reactor and is operating successfully at control temperature and pressure of 625° C and 1000 psi. It, too, contains high purity uranium U + Fe-Al, and U + Fe-Si specimens representing several metallurgical states with the half tubular geometry. Additionally, two controlled pressure-temperature capsules (P-5 and P-6) were assembled and bench-tested. They are scheduled to operate in a reactor at 500 psi and 450 C and 575 C, respectively.

A general swelling capsule containing eight specimens of 2.88 at. % enriched, high purity uranium in several heat-treated conditions was charged in a reactor and operated successfully at a controlled temperature of 375° C. It was discharged after reaching goal exposure and will be examined to obtain better insight into the role of burnup and burnup rate on swelling mechanisms.

Postirradiation Examination

Optical and electron metallography was completed on several specimens from the first controlled pressure (1000 psi) - temperature (450° C) capsule (P-2) which was irradiated to 0.13 at. % specimen burnup. As anticipated from the small density changes reported in HW-84573,⁽¹⁾ microtears were not present in the microstructure although the microstructure had assumed the "worked-swirled" structure typical of this irradiation temperature. Thus, it is observed that external pressure is very effective in

(1) Quarterly Progress Report - Metallurgy Research Operation, October, November, December 1964, HW-84573, General Electric Company, Richland, Washington. January 15, 1965.

overcoming the mechanical tearing that occurs in unrestrained uranium irradiated at a temperature of 450° C. This pronounced inhibiting effect of pressure is another bit of evidence that fission gas agglomeration plays only a minor role in the swelling of alpha uranium.

The second controlled pressure-temperature capsule (P-3), which operated at 1000 psi and 575° C to about 0.13 at. % BU was opened in the Radiometallurgy Laboratory and the specimens recovered. The capsule contained six high purity uranium specimens, two U + Fe-Al specimens and two U + Fe-Si specimens which had undergone several different heat treatments prior to irradiation. Preirradiation specimen data and postirradiation photomacrographs of the specimens are given in Table 3.1 and Figure 3.3, respectively. Preliminary density results on capsule P-3 specimens also show that pressure inhibits swelling significantly at an irradiation temperature of 575° C and a pressure of 1000 psi. The analysis of the microstructure of these specimens and its correlation with density change data is being investigated and will be available shortly.

TABLE 3.1
PREIRRADIATION DATA ON SPECIMENS IN CAPSULE P-3^(a)

Specimen No.	Material	Heat Treatment
P-3, A-1	High Purity Uranium ^(b)	As-Extruded
A-2	High Purity Uranium	As-Extruded
B-1	U + Fe-Al ^(c)	As-Extruded
B-2	U + Fe-Al	730° C for 15 min, O. Q.
C-1	High Purity Uranium	As-Extruded
C-2	High Purity Uranium	As-Extruded
D-1	High Purity Uranium	730° C for 15 min, O. Q.
D-2	High Purity Uranium	730° C for 15 min, O. Q.
E-1	U + Fe-Si ^(d)	1000° C for 24 hr, W. Q.
E-2	U + Fe-Si	1000° C for 24 hr, W. Q. +730° C for 15 min, O. Q.

(a) Irradiated at 1000 psi, 450° C to ~0.13 at. % BU

(b) High Purity Uranium - 65 ppm Fe, < 5 ppm Al, 24 ppm Si, 6 ppm C

(c) U + Fe-Al - 400 Fe, 640 Al, 85 Si, 500° C

(d) U + Fe-Si - 140 Fe, 25 Al, 95 Si, 400° C

W. Q. - Water Quenched

O. Q. - Oil Quenched

Side
View

End
View

3. 10

BNWL-79

FIGURE 3. 3

As-Irradiated Macro Appearance of Specimens Irradiated in Capsule P-3
(Table 3. 1) 2X

Neg. C-8558, 8561, 8564, 8567, 8570
C-8573, 8576, 8579, 8582, 8585

Neg. C-8660, 8664, 8666, 8569, 8572
C-8575, 8578, 8581, 8584, 8587

PLUTONIUM PHYSICAL METALLURGY - R. D. Nelson and F. E. Bowman

The objective of this program is to conduct basic studies on the properties of high-purity unalloyed and alloyed plutonium. Studies will include investigation of phase transformation, phase stabilization, deformation, fracture, texturing, internal stresses, microcracking, grain growth, recovery and recrystallization, and measurement of mechanical and physical properties. The unique structure of several of the plutonium allotropes and their narrow temperature span of stability leads to characteristics and anomalies which, if explained, will contribute to the general area of solid state science.

Research studies have been performed on the effect of concurrent applied uniaxial compressive and tensile stresses and prior plastic deformation on the low temperature allotropic transformations in plutonium. The primary objective of this activity was to gain information about the mechanisms of these transformations and to compare the transformation in plutonium to transformations in more common metals. The study of the total strain accompanying the transformations under applied compressive stress was reported previously.^(1, 2) This work has been extended to include, (1) the total strain accompanying the $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ transformation under applied tensile stresses, (2) the effect of applied compressive and tensile stresses on the temperature for the start of the $\beta \rightarrow \alpha$ transformation, and (3) the effect of plastic deformation of the stable parent phase on the $\alpha \rightarrow \beta$ and $\beta \rightarrow \gamma$ transformations.

Total Strain Accompanying the $\beta \rightarrow \alpha$ Transformation Under Applied Tensile Stresses

The total strain accompanying the various transformations in pure plutonium is most conveniently analyzed by considering three contributions

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- (1) J. J. Cadwell, ed. Quarterly Progress Report - Metallurgy Research Operation - July, August, September, 1963, HW-78968, General Electric Company, Richland, Washington. October 15, 1963.
 - (2) J. J. Cadwell, ed. Quarterly Progress Report - Metallurgy Research Operation - October, November, December, 1964, HW-84573, General Electric Company, Richland, Washington. January 15, 1965.

to the total strain:

1. Normal creep strain. This has been discussed previously. Normal creep can occur significantly during transformation under stress.
2. Excess creep strain. This is the creep at small applied stresses where normal creep is not expected. Excess creep has been postulated to be caused by an excess concentration of point defects.
3. Transformation strain. Transformation strain is defined as the macroscopic shape change resulting from the summation of the shape change accompanying the formation of each grain of the product phase. In a perfectly random transformation this strain would be isotropic and equal to one-third the fractional volume change accompanying transformation. If the transformation occurs anisotropically, the resulting transformation strain will be similarly anisotropic.

The total strain during the $\beta \rightarrow \alpha$ transformation in plutonium was investigated as a function of applied tensile stresses at different transformation rates. A tensile stress tends to oppose the contraction of the transformation in the direction of the applied stress. Representative total strains at stresses from 250 psi to 7000 psi at a cooling rate of 2° C/hr are shown in Figure 3. 4. These curves if compared to similar curves pertaining to applied compressive stresses show that the effect of applied tensile stresses was more pronounced and somewhat different from the effect of applied compressive stresses on the $\beta \rightarrow \alpha$ transformation. The total strain accompanying the transformation with very low tensile stresses was negative but the contraction in the specimens was less than the contraction with no applied stress.

At 1500 psi the specimens elongated during the first stages of transformation. The positive rate of strain decreased as the transformation proceeded, became zero, and then became negative because of the contraction associated with the volumetric change. The effect was similar at 2900 psi,

FIGURE 3. 4

Total Strain Accompanying the Plutonium $\beta \rightarrow \alpha$ Transformation
Under Applied Tensile Stresses at a Cooling Rate of $2^\circ\text{C}/\text{min}$

the elongation being greater and the contraction less than at 1500 psi. Some net elongation was observed in the direction of the applied tensile stress in specimens transformed at 7000 psi. It is reasonable to assume that the linear transformation strain is limited to values between zero and $\Delta V/V_0$, i. e., 0 to 0.09. Therefore, the elongation observed in specimens transformed in tension must be attributed to excess creep strain.

Total Strain Accompanying the $\alpha \rightarrow \beta$ Transformation Under Applied Tensile Stresses

Extra creep deformation during $\alpha \rightarrow \beta$ transformation was more evident under applied uniaxial tensile stresses than it was under applied uniaxial compressive stresses. An applied tensile stress favors expansion in the direction of the applied stress during $\alpha \rightarrow \beta$ transformation. If all of the transformation were to occur in the direction of the applied stress without normal creep and without excess strain accompanying the transformation, then $\Delta L/L_0$ would equal $\Delta V/V_0 \approx 0.09$; this is analogous to

the $\beta \rightarrow \alpha$ transformation conducted under an applied uniaxial compressive stress. The total strain accompanying the $\alpha \rightarrow \beta$ transformation increased with applied stress and increased with decreasing transformation rate. Experiments showed that $\Delta L/L_0$ during $\alpha \rightarrow \beta$ transformation under an applied tensile stress exceeded 0.09, evident in Figure 3. 5.

FIGURE 3. 5

Total Strain Accompanying the Plutonium $\alpha \rightarrow \beta$ Transformation Under Applied Tensile Stresses at a Heating Rate of 2 to 5°C/hr

Since the formation of disks of beta during the $\alpha \rightarrow \beta$ transformation, analogous to the formation of alpha disks during the $\beta \rightarrow \alpha$ transformation, has not been verified, it is impossible to place limits on the range of transformation strain.

Some excess transformation creep was observed but it was much less than the anisotropic transformation strain of the transformations under

applied stress. Compressive stresses applied during the $\beta \rightarrow \alpha$ transformation produced very little excess creep. More excess creep was observed during the $\alpha \rightarrow \beta$ transformation. The excess creep occurred during the latter stages of the $\alpha \rightarrow \beta$ transformation and during the first stages of the $\beta \rightarrow \alpha$ transformation. The significance of this observation is that excess creep occurs in the weaker beta phase during both the $\alpha \rightarrow \beta$ and the $\beta \rightarrow \alpha$ transformations. The anisotropic transformation strain during $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations is large compared to the sum of normal creep and extra transformation creep deformation.

Effect of Applied Stress on the $\beta \rightarrow \alpha$ Transformation Temperature

The temperature for the beginning of the $\beta \rightarrow \alpha$ transformation was determined as a function of applied uniaxial tensile and uniaxial compressive stresses. These temperatures were compared with those for the start of the transformation as a function of hydrostatic pressure, Figure 3.6.

FIGURE 3.6

Effect of Applied Uniaxial Tensile and Compressive Stresses on the Temperature for the Start of the Plutonium $\beta \rightarrow \alpha$ Transformation at Heating and Cooling Rates of 2°C/hr

The three methods of stressing the specimens produced virtually linear changes in the transformation temperature up to 7000 psi. The slopes of the curves were $+3^\circ\text{C}/1000\text{ psi}$ for tension and compression and $+0.4^\circ\text{C}/1000\text{ psi}$ for hydrostatic pressure. These data clearly show that there was

an appreciable difference between the three types of loading. Note that all three types of loading increased the transformation temperature. The very large temperature increase of the start of the transformation of specimens transformed in compression and transformed in tension is difficult to explain on the basis of a diffusional transformation, but it can be explained on the basis of a martensitic type transformation.

Crystallographic Relationships

The application of the elevated temperature specimen holder on the X-ray diffractometer to the study of the $\alpha \rightarrow \beta \rightarrow \alpha$ transformation processes has revealed several significant phenomena which have, more or less, determined the direction of the effort devoted to this area of study during the past quarter.

It became apparent early in this study that the interphase cycling of an initially textured plutonium specimen did not result in the restoration of the random orientation characteristic of an as-cast sample. The elevated temperature diffraction investigations indicated that a texture could be introduced in a randomly oriented samples by means of a slow $\alpha \rightarrow \beta \rightarrow \alpha$ cycle. This is undoubtedly the result of the heat flow or stress patterns, or a combination of both, and suggests an explanation for the apparent persistence of the texture which had existed initially in the specimen studied.

A logical extension of the investigation was the study of the effects of cycling on massive textured samples. It was found, in the case of several samples textured by conducting the $\beta \rightarrow \alpha$ transformation under a compressive load, that the $[010]$ direction initially parallel to the applied load direction was shifted 90° after the first $\alpha \rightarrow \beta \rightarrow \alpha$ cycle. The validity of this observation was substantiated by metallographic evidence in which the axis of the columnar grains was found to have undergone the 90° shift in direction relative to the applied stress direction.

This observation suggests that this $[010]$ direction is the direction of maximum change as the unit alpha cell forms from beta. It would then tend to line up relative to an applied stress in a direction which in conjunction with the stress would most readily effect the transformation. In other

words, the increase in density characteristics of the $\beta \rightarrow \alpha$ transformation would be favored by the application of a compressive stress in the $[010]$ direction, whereas the $\alpha \rightarrow \beta$ transformation with its decrease in density should be favored by a tensile stress applied perpendicular to the $[010]$ direction. An apparatus which permits this cyclic application of the two types of loads has been constructed and applied to the plutonium phase transformation studies. It was found that the degree of texturing was indeed enhanced by the use of compression during the $\beta \rightarrow \alpha$ and tension during the $\alpha \rightarrow \beta$ transformations.

With the exception of a thin shell of near random grains, the sample exhibited an almost perfect fiber texture. In the transverse section only the (020) reflection appears in the diffraction pattern if the sample surface is maintained essentially parallel with the diffractometer axis. Any lack of coincidence between the sample surface and the reflecting plane can be attributed to inherent errors in cutting the specimen. It has been found possible, by altering the pole figure goniometer angles, to bring several strongly diffracting planes, individually, into reflecting positions. In this respect, the specimen resembles a single crystal in its diffraction behavior. Although the diffraction pattern pertains to a surface area of the order of square centimeters, the volumes of these pseudo single crystals can only be inferred. An attempt is currently underway to plot the experimentally determined diffracting plane positions on a pole figure and compare the angular relationships thus obtained with those calculated from the reported crystal structure and unit cell dimensions.

It is interesting to note that there has been very little evidence of microcracking as indicated by metallographic examination and density determinations in these samples which have been cycled under stress at temperatures ranging only slightly above and below the equilibrium value. Ordinarily such cycling would result in a rapid deterioration of the specimen.

The X-ray diffraction studies of these textured samples has revealed several apparent anomalies. For example the second and fourth order (010) reflections, the (020) and (040), have been found to vary in relative intensity

in different specimen positions. Without altering any of the diffractometer parameters other than goniometer angles the relative intensity of the (040) reflection has been found to vary from 40% to less than 10% of the (020). The value calculated from the Argonne computer program data is about 60%. Since it is necessary, for the most part, to depend upon the (040) reflection as a means of positively identifying the (020) because the latter is almost coincidental with the (211), this variation in the (040) intensity results in uncertainties in the interpretation of the diffraction pattern. Some feature of the alpha structure, as yet unrecognized, must be responsible for this and other observed anomalies.

Metallographic Relationships

At high alpha temperature the transformation of $\beta \rightarrow \alpha$ initially proceeds by the formation of thin disk-shaped volumes, which consist of many individual columnar grains of the alpha phase. The long axes of the grains are all perpendicular to the plane of the disk. In a polished and etched cross-section the disks appear as bands, which are randomly oriented in the absence of stresses. When a compressive stress is applied to the metal, the disks form at right angles to the compressive stress, while the grains within the disks are parallel to the applied stress. The alignment of the alpha disks is particularly evident from the appearance of the bands in specimens only partially transformed with 15,000 psi uniaxial compression. During $\beta \rightarrow \alpha$ transformation under compressive stress the (010) alpha planes form perpendicular to the direction of the applied compressive stress.

When a tensile stress was applied to the metal, the disks formed parallel to the direction of the applied stress, while the grains within the bands were elongated in a direction perpendicular to the applied stress. The (010) planes become aligned parallel with the direction of the applied stress, revealed by the very weak (040) reflection from a section perpendicular to the applied stress.

When plutonium is transformed under compression the dimensions of the disks perpendicular to the plane of the disk is determined or limited by impingement with adjacent disks; the disks extended to the boundaries

of the specimen in the direction parallel to the plane of the disk. When plutonium is transformed under no applied stress or under tension all dimensions of the disk are restricted either by impingement or by the boundaries of the specimen.

Anisotropy of the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ Transformations

A highly textured cylindrical rod specimen, with the (010) perpendicular to the rod axis, was cycled once through the $\alpha \rightarrow \beta$ and back through the $\beta \rightarrow \alpha$ transformation without applied stress. The specimen was first heated to 115° C and the diameter and the length of the specimen in the alpha phase were measured. The dimensions of a rod in the alpha phase can be readily compared to the corresponding dimensions in the beta phase at 115° C since both beta and alpha can exist at this temperature for long times. The specimen was then heated to 160° C, held for 1 hr to transform the metal completely to beta, and then cooled to 115° C for measuring the length and diameter of the rod in the beta phase. Transformation of the beta phase to the alpha phase was accomplished by cooling to room temperature at a rate of 50° C/hr. The rod was then reheated to 115° C and the dimensions of the alpha phase rod were measured. The dimensions of the rod in the alpha phase before cycling, in the beta phase, and in the alpha phase after cycling could therefore be compared.

The length increased 7.9% and the diameter increased 0.8% during the $\alpha \rightarrow \beta$ transformation. This indicated that nearly all of the volume change during transformation occurred by expansion in a direction parallel to the length of the specimen. The length and the diameter decreased 1.2 and 4.3%, respectively, during the $\beta \rightarrow \alpha$ transformation. The sample after transformation cycling had increased in length by 6.5% and had decreased in diameter by 3.4%. From this it can be concluded that both the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations occurred anisotropically.

The anisotropic nature of the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations of the textured rod indicated that a texture should still be present in the alpha phase after an $\alpha \rightarrow \beta \rightarrow \alpha$ cycle with no applied stress. A texture after cycling was confirmed by X-ray diffraction and by metallography, but it was different

from the texture that had been observed before cycling. After cycling, the disks were aligned predominantly parallel to the direction of the original applied compressive stress, rather than 90° to the applied stress as observed before cycling. The alignment of the elongated alpha grains in the radial direction is in accordance with the observation that strain accompanying transformation occurred primarily radially.

The anisotropic $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformation strains and the 90° rotation of the crystallographic and microstructural textures imply that there is a specific crystallographic orientation relationship between the parent and the product phases in the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations. From the 90° rotation of the texture it can be concluded that there is more than one crystallographic variant of the orientation relationship in spite of the low symmetry of the two phases. These are characteristics to be expected of martensitic transformations.

Martensitic $\beta \rightarrow \alpha$ Transformation

Although the results of this investigation are not sufficient criteria for determining unequivocally whether the $\beta \rightarrow \alpha$ transformation is martensitic or diffusional, they do indicate that the transformation behaves in a manner consistent with known martensitic transformations. This conclusion is based on the following observations:

1. Both uniaxial tensile stresses and uniaxial compressive stresses raise the temperature for the beginning of the $\beta \rightarrow \alpha$ transformation at a fixed cooling rate.
2. Stresses applied during either the $\alpha \rightarrow \beta$ or the $\beta \rightarrow \alpha$ transformations produce pronounced crystallographic texturing and microstructural shape texturing.
3. There is an apparent crystallographic relationship between the alpha and the beta phases.

The alpha grains of plutonium are aligned during transformation under applied stress. The alpha grains of plutonium formed by transformation from beta under stress are in the form of needles or rods. The grains transformed

under compressive stresses are longer and thinner than those formed by transformation under no stress; the higher the stress the thinner are the alpha grains. More significantly, the elongated grains are aligned in disks which are random in the absence of applied stresses, perpendicular to applied compressive stresses, and parallel to applied tensile stresses.

Both applied uniaxial tensile and compressive stresses raise the temperature for the beginning of the $\beta \rightarrow \alpha$ transformation for a given cooling rate. That is, all applied stresses favor the alpha nucleation process. This is completely in keeping with the hypothesis that the $\beta \rightarrow \alpha$ transformation is a martensitic transformation. The shear and normal components of a compressive stress favor the plutonium $\beta \rightarrow \alpha$ transformation; the shear component of tensile stresses also favors the $\beta \rightarrow \alpha$ transformation in plutonium; but the normal component of tensile stresses opposes the $\beta \rightarrow \alpha$ transformation. There are no shear components in hydrostatic compression.

DAMAGE MECHANISMS - F. A. Smidt

The objective of this program is to determine how the mechanical properties of irradiated iron are modified by interaction of moving dislocations with irradiation induced defects.

The method being utilized is the application of thermally activated flow theory to the deformation process; the method yields information on the rate controlling barriers to dislocation motion. Correlations are being sought between changes in these and other physical properties with variations in interstitial impurity content and neutron exposure. This method of analysis, therefore, will help identify the defects introduced by irradiation and provide insight into the mechanism by which mechanical properties change as a result of irradiation.

During this period, additional refinements have been made in an equation to predict the effective stress at various conditions of temperature and strain rate or at a given activation energy. The equation is presently being applied to the experimental data obtained from irradiated iron specimens.

The initial yield point and pseudo yield points after strain rate changes have been investigated. Analysis of the data indicates that the changes observed are real and not produced by transient machine effects.

Thinning of bulk samples of iron irradiated to 5×10^{18} nvt has been successfully accomplished. Inspection of the thinned samples by electron transmission microscopy did not show any of the spots or loops observed in molybdenum irradiated to this exposure.

REACTOR METALS RESEARCH

ALLOY SELECTION - T. T. Claudson

Several nickel base alloys are being irradiated and tested to determine the effects of irradiation and environment upon their mechanical properties. These materials are being irradiated at temperatures between 50 and 740° C and tested at room temperature, 300 and 650° C.

Several room temperature and elevated temperature tests have been completed on tensile specimens of Hastelloy N. Figure 4. 1 shows

FIGURE 4. 1

Effect of Neutron Irradiation on the Room Temperature Mechanical
Properties of Hastelloy N Irradiated at 280 C
in the Solution Treated Condition

the data for specimens tested at room temperature which were irradiated at 280° C in a water environment to exposures between 3.7×10^{19} to 8.27×10^{20} nvt ($E > 1$ Mev). Compared to the control specimens, the irradiated specimens showed an increase in yield strength of about 50% with a slight decrease in percent uniform elongation. Good room temperature ductility was observed over all exposures, indicating no serious deterioration of the room temperature properties.

Additional specimens of Hastelloy N irradiated at 280° C were tested at 650° C. Two specimens were tested after reaching an exposure of 1.03×10^{20} and 1.46×10^{21} nvt ($E > 1$ Mev), respectively. The results of these tests are shown in Figure 4. 2.

FIGURE 4. 2

Effects of Neutron Irradiation at 280° C on the Mechanical Properties of Hastelloy N Tested at 650° C in the Solution Treated Condition

The specimen irradiated to an exposure of 1.03×10^{20} nvt showed a higher yield and ultimate strength than the specimen irradiated to the higher exposure. More important is the evidence of extreme brittleness in the specimen irradiated to an exposure of 1.46×10^{21} nvt. Almost a

complete loss in uniform and total elongation was found with only a small (7. 19%) reduction in area. This loss in ductility together with a large loss in strength indicates severe damage to the high temperature properties of the material.

Similar specimens of Hastelloy X-280 were irradiated at 280° C to exposures of 1.03×10^{20} , 1.57×10^{20} , and 3.2×10^{20} nvt ($E > 1$ Mev) and tested at 650 C. Results of these data are shown in Figure 4. 3. These data indicate an increase in yield and ultimate tensile strength with increasing exposure. The ductility of the material was reduced from room temperature ductility previously reported but is still in the 10-15% range for both uniform and total elongation. Necking of the specimens was also observed and indicated by an ultimately large percent reduction in area.

FIGURE 4. 3

Effects of Neutron Irradiation at 280° C on the Mechanical Properties of Hastelloy X-280 Test at 650° C in the Solution Treated Condition

An addition specimen of Hastelloy X-280 irradiated to an exposure of 1.91×10^{20} nvt ($E > 1$ Mev) at an average temperature of 740° C was

tested at 650° C. The results of this test are shown in Figure 4. 4, and compared to those of a specimen irradiated at 280° C to a slightly lower but comparable exposure. Comparing these results there is found a higher yield and ultimate strength in the specimen irradiated at 740° C. More important, the higher irradiation temperature has caused a substantial loss in ductility. Control tests for these irradiations are presently being performed and analyzed.

FIGURE 4. 4

Effect of Neutron Irradiation Temperature on the Mechanical Properties of Hastelloy X-280 Tested at 650 C

Incoloy 800 is a nickel base solution hardenable alloy whose mechanical properties and corrosion resistance indicate that it may be applicable for nuclear superheat fuel cladding. Specimens of this alloy were irradiated at 740° C in the mill annealed, 20% cold-worked and solution treated conditions and tested at 650° C. The results of these tests are plotted in Figure 4. 5. A considerable difference in high temperature strength and

ductility can be seen in comparing the three conditions with the solution treated specimen retaining the most ductility. The ductility of these alloys is considerably lower than the room temperature ductility previously reported.

FIGURE 4. 5

Effects of Preirradiation Treatment on the Mechanical Properties of Incoloy 800 Irradiated at 740° C and Tested at 650° C Exposure 1.91×10^{20} nvt ($E > 1$ Mev)

The effect of irradiation temperature upon the 650°C tensile properties of Incoloy 800 is shown in Figure 4. 6. It is evident from these data that while little change in yield strength is evident a considerable difference in elongation is present with the higher temperature irradiation specimen retaining only 9.59% uniform elongation as compared to 23 to 37% for the specimen irradiated at 280° C.

FIGURE 4.6

Effects of Irradiation Temperature on the Mechanical Properties of Incoloy 800
Tested at 650°C in the Solution Treated Condition

Specimens of Inconel 600 also irradiated in the mill annealed, 20% cold worked, and solution treated condition were tested at 650° C after being irradiated at 280 and 740°C. The results of these tests are given in Figures 4.7 and 4.8, respectively. At both irradiation temperatures, the ductility of the specimens was low, ranging from 2.08 to 7.62% in uniform elongation. In both cases the solution annealed specimens exhibited the lowest strength. Comparing the strength of both sets of specimens it is evident that a substantial lowering in the elevated temperature strength was evident in the 740 C irradiation.

FIGURE 4.7

Effects of Preirradiation Treatment on the Mechanical Properties of Inconel 600 Irradiated at 280° C and Tested at 650° C
Exposure 5.68×10^{20} nvt ($E > 1$ Mev)

FIGURE 4.8

Effect of Preirradiation Treatment on the Mechanical Properties of Inconel 600
 Irradiated at 740°C and Tested at 650°C
 Exposure 1.91×10^{20} nvt ($E > 1$ Mev)

Specimens of Inconel 600 irradiated at 280° C to exposures of 1.57×10^{20} nvt and 5.68×10^{20} nvt ($E > 1$ Mev) were tested at 650° C. As shown in Figure 4.9, the specimen irradiated at the higher exposure showed a higher yield and ultimate strength and a correspondingly lower ductility. An increase in yield strength from 26,420 psi for the specimen irradiated to an exposure of 1.57×10^{20} nvt to 39,140 psi for the specimen irradiated to 5.68×10^{20} nvt was found. This was accompanied by a decrease in percent uniform elongation from 10.35 to 3.18% for the same specimens.

FIGURE 4.9

Effects of Neutron Irradiation at 280° C on the Mechanical Properties
of Inconel 600 Tested at 650° C
in the Solution Treated Condition

The results of room temperature tests performed upon irradiated specimens of Inconel 625 are found in Figure 4. 10 through 4. 12. Specimens to date have been irradiated to exposures ranging between 5.3×10^{19} to 8.7×10^{20} nvt ($E > 1$ Mev). Figures 4. 10 and 4. 11 show the data for

FIGURE 4. 10

Effects of Neutron Irradiation on the Room Temperature Properties of Inconel 625 Irradiated at 50° C in the Solution Treated Condition

specimens irradiated at 50° C and 280° C, respectively. Results of these tests indicate an increase in strength of the alloy with only small changes in the ductility of the alloy. Specimens irradiated at 50° C were affected to a greater extent than those irradiated at 280° C. Specimens irradiated to an exposure of 9.8×10^{19} nvt showed a 33% increase in yield strength and a 24% decrease in uniform elongation. After an exposure

FIGURE 4. 11

Effects of Neutron Irradiation on the Room Temperature Properties of Inconel 625 Irradiated at 280°C in the Solution Treated Condition

of 7.5×10^{20} nvt, specimens irradiated at 50°C had an average yield strength of 160,600 psi, for an increase of 72%. The uniform elongation was found to be only 8.43% as compared to 36.44% for the control specimens. Changes in the specimens irradiated at 280°C were much smaller and good ductility was noted over all irradiation levels.

Figure 4.12 illustrates the effects of irradiation temperature on the room temperature properties of Inconel 625 irradiated to an exposure of about 1×10^{20} nvt ($E > 1 \text{ Mev}$). The range of exposures actually used in plotting this data was necessary since specimens were irradiated at different times in various facilities and the goal exposure of 1×10^{20} nvt

FIGURE 4. 12

Effects of Irradiation Temperature on the Room Temperature Properties of Inconel 625 Irradiated to an Exposure of About 1×10^{20} nvt ($E > 1$ Mev)

was not exactly achieved. As can be seen in Figure 4.12, the primary effect produced by the irradiation at different temperatures was to reduce the ductility of the material with increased temperature. The 50 and 280°C irradiation temperatures maintained good uniform elongation and reduction in area ductility, while this value decreased at the 580 and 740°C irradiation temperatures. Comparing the irradiated data with the control data, it is evident that much of this decrease is due to only the thermal environment. There was, however, a considerable effect at the 580°C irradiation temperature upon the uniform elongation of the material. After an exposure of 9.2×10^{20} nvt the uniform elongation was 17.6% as compared to 27.3% for the control specimens.

More interesting are the results of the 650° C tensile tests performed on specimens of Inconel 625 irradiated at 280° C. These results are shown in Figure 4. 13. Control specimens for these tests were exposed in the

FIGURE 4. 13

Effects of Irradiation at 280° C on the Mechanical Properties of Inconel 625 Tested at 650° C in the Solution Treated Condition

out-of-reactor hot water loop facility at a temperature of 280°C and tested in air at 650° C. The irradiated specimens were tested in a similar manner on identical equipment. The results of these tests indicate little change in yield strength up to an exposure of 5.68×10^{20} nvt. A decrease in ultimate strength is prevalent at this exposure, decreasing to 108,600 psi from the control level of 133,800 psi. Some increase in uniform elongation is seen although this may be well within the experimental

error. There was essentially no change in the reduction in area of the specimens. At an exposure of 1.46×10^{21} nvt, a marked deterioration in strength and uniform elongation was found. The specimens were found to have very little elongation and necked in a very localized region as shown by the increase in the reduction in area values. Additional tests are being performed to verify these effects although similar effects are being noticed in other nickel base alloys.

Present and future reactor concepts utilize a liquid metal for the reactor primary cooling system. Past experience has shown the environmental effects upon the mechanical properties of reactor materials to be substantial. This may be especially true in the case of liquid metal environments where in addition to corrosion the mass transfer of metallic elements presents a distinct problem.

To evaluate the effects of environment and neutron irradiation on the mechanical properties of reactor structural materials, a liquid metal temperature controlled capsule has been designed. A bench model of this capsule has been fabricated using an electrical heater to simulate the gamma heat generation. This model capsule will be used to test the convection principle used in the capsule design and to determine certain heat transfer characteristics.

A schematic of the capsule is shown in Figure 4. 14. The specimen container portion of the capsule is surrounded by a vacuum insulator region which inhibits radial heat flow from the capsule. As the coolant increases in temperature its density decreases and thus rises to the cooler regions of the capsule, between the inner container and specimen container walls, thus setting up a convection current. Temperature control is maintained by flowing a helium-argon gas mixture between the inner and outer container walls. A substantial amount of control can be accomplished by varying the ratio of helium to argon in the control gas mixture. Figure 4. 15 shows the as-fabricated model liquid metal capsule. A simple manually controlled safety circuit and temperature control system is used. Schematic diagrams of both these systems are shown in Figures 4. 16 and 4. 17, respectively.

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FIGURE 4. 13

Effects of Irradiation at 280° C on the Mechanical Properties of Inconel 625 Tested at 650° C in the Solution Treated Condition

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FIGURE 4. 14

Liquid Metal Environ-
mental Capsule

Neg. 0650847-1; 0650869-2

FIGURE 4. 15

As-Fabricated
Bench Model
of the Liquid
Metal Irradia-
tion Capsule

FIGURE 4. 17

Gas Flow Diagram for the Liquid Metal Environmental Capsule

IN-REACTOR MEASUREMENTS OF MECHANICAL PROPERTIES -

J. A. Williams, D. H. Nyman, and J. W. Carter

The objective of the in-reactor measurement program is to define the effects of fast neutron irradiation on the mechanical properties of structural materials.

The temperature and stress dependency of the in-reactor creep of annealed AISI 304 stainless steel are presently being investigated.

In-Reactor Measurements

In-reactor creep tests at 650° C, 10,000 psi and 525° C, 30,000 psi were started and a test at 550° C, 30,000 psi was concluded during the past quarter.

Reduced rupture life and ductility at failure were observed for the in-reactor 550° C, 30,000 psi creep test as compared to the out-of-reactor test. The in-reactor creep strain at failure was 3 to 4% with a failure time of 760 hr. The out-of-reactor test, still in progress, has accumulated 3500 hr of test time with a creep strain of 4.8%. The minimum creep rate for the in-reactor test was 2.6×10^{-5} /hr. The out-of-reactor creep test exhibited a creep rate 9.3×10^{-6} /hr.

The 650° C, 10,000 psi in-reactor creep test yielded a creep rate of 4.4×10^{-6} /hr after 800 hr of testing while the out-of-reactor control test showed a rate of about 2.2×10^{-6} /hr. Significant creep rates have not yet been determined for the 525° C, 30,000 psi as it is still in the primary creep stage.

The metallographic investigation being conducted on the recovered AISI 304 stainless steel in-reactor creep specimen is partially completed. Photomicrographs of transverse sections from one buttonhead and from near the fracture show no unusual features when compared to the unirradiated material. Longitudinal sections from the area near the fracture and the buttonheads will be made soon.

A second in-reactor test capsule has been discharged from the reactor and sent to the Radiometallurgy facility.

Capsule Development

With the objective of expanding present testing capabilities, a new facility has been developed in the Hanford Plutonium Recycle Test Reactor.

During the past quarter, modification of an in-reactor creep capsule for use in the PRTR has been completed. The same capsule currently being used has been modified to include an outside cooling chamber since testing in the PRTR necessitates operating in a thimble tube inserted into a dry fuel channel. Facilities installation has also been completed at the PRTR and the capsule will be charged during the next outage.

The capsule contains an annealed Zircaloy-2 specimen. The test conditions will be 300° C and 10,000 psi stress.

IRRADIATION FACILITIES OPERATION - J. E. Irvin

The purpose of this phase of the program is to provide for the timely accomplishment of irradiating, testing, and subsequent processing of the data obtained from a number of different structural materials. Irradiations will be performed in aqueous, gaseous, and molten metal environments. Elevated temperature irradiations and tests will be emphasized. Special tests applicable to both irradiated and unirradiated specimens will be developed.

Facilities Operation

The experiments discharged from the various ETR core positions at the conclusion of Cycles 69 and 70 are given in Table 4. 1 along with

TABLE 4. 1

SUMMARY OF EXPERIMENTS DISCHARGED FROM THE ETR FOLLOWING CYCLES 69 AND 70

<u>Experiment Designation</u>	<u>Number and Type of Specimen</u>	<u>Estimated Flux, nv^(a)</u>	<u>Estimated Exposure, nvt^(a)</u>
<u>Quadrant Discharges - ETR Cycle 69</u>			
120	18 NPR Zircaloy-2 Tensiles	1.0×10^{14}	2.0×10^{21}
175	6 Zr-Inconel Tensiles	1.0×10^{14}	5.7×10^{20}
181	18 AISI 406 Tensiles	6.0×10^{13}	1.0×10^{20}
<u>Quadrant Discharges - ETR Cycle 70</u>			
129	18 Nickel-Base Tensiles	6.0×10^{13}	1.2×10^{21}
147	18 Zircaloy-2 Tensiles	6.8×10^{13}	3.6×10^{20}
148	18 Zircaloy-2 Tensiles	6.5×10^{13}	3.6×10^{20}
178	6 Zr-Inconel Tensiles	(b)	5.0×10^{20}
182	18 AISI 406 Tensiles	3.7×10^{13}	1.3×10^{20}
183	6 Hastelloy X-280 Tensiles	6.0×10^{13}	1.1×10^{20}
184	6 Hastelloy X-280 Tensiles	5.4×10^{13}	1.0×10^{20}
185	4 Hastelloy X-280 Tensiles	8.0×10^{13}	1.5×10^{20}
185	2 Hastelloy X-280 Stress-Rupture	8.0×10^{13}	1.5×10^{20}
186	6 Hastelloy X-280 Stress-Rupture	1.0×10^{14}	1.8×10^{20}
<u>Capsule Discharges^(c) - ETR Cycle 70</u>			
653	8 AISI 304 Tensiles	1.0×10^{14}	8.5×10^{20}
655	8 AISI 304 Tensiles	1.0×10^{14}	8.1×10^{20}
657	8 AISI 304 Tensiles	8.0×10^{13}	5.8×10^{20}
658	8 AISI 348 Tensiles	7.6×10^{13}	5.4×10^{20}
660	8 AISI 348 Tensiles	7.6×10^{13}	5.4×10^{20}

(a) Neutrons with energies greater than 1 Mev.

(b) This quadrant is twice the normal quadrant length and was irradiated in two different positions during successive cycles. Therefore, the average flux was not determined for this experiment.

(c) No ambient temperature capsules were discharged from the ETR at the end of Cycles 68 or 69.

Total number of specimens discharged, Cycles 69 and 70 = 184.

Total number of specimens discharged from G-7 Hot Water Loop,
Cycles 69 and 70 = 144.

Total number of specimens discharged from G-7 Loop to date = 2366.

the estimated neutron fluxes and exposures (> 1 Mev). The operating histories of the ETR and the G-7 Hot Water Loop are given in Table 4. 2.

TABLE 4. 2
OPERATING HISTORY OF ETR AND G-7 LOOP FACILITY

Operating History	Cycle Number	
	68	69
1. Reactor History		
Startup ^(a)	12-1-64	1-20-65
End of Cycle	1-3-65	2-14-65
Megawatt Days	3899	3277
Effective Days at 175 MW	22.3	18.7
Number of Scrums ^(b)	8	5
Number of Shutdowns	4	2
2. G-7 Loop History		
Maximum Temperature	535° F	540° F
Effective Days Above 200° F	23.9	19.4
Effective Days at Operating Temperature	19.6	18.3
Operating Efficiency ^(c)	107.2%	103.7%
Temperature Efficiency ^(d)	82.0%	94.3%

(a) Includes flux run.

(b) Power drops below half-full power with immediate recovery.

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

(d) Effective days of operation between 500 and 550° F (260 and 288° C) with respect to days of operation above 200° F (93° C).

Control tests discharged from the out-of-reactor loop at the end of Cycle 67 are listed in Table 4. 3. No tests were discharged upon completion of Cycle 68. An exposure duplicating the history of ETR Cycle 69 is currently in progress.

TABLE 4. 3
HISTORY AND DISCHARGE SCHEDULE FOR OUT-OF-REACTOR LOOP

Loop History	Cycle Number
	67
1. Time at temperature (in hours)	
200°C (392° F)	33.1
218°C (424° F)	14.3
246°C (474° F)	21.2
282°C (540° F)	657.7
Total time (in hours)	726.3
2. Specimen discharge (material, type, and number)	
Nickel-Base (tensiles)	126
AISI 406 (tensiles)	18
Inconel-Zircaloy-2 (tensiles)	6
Total number of specimens discharged*	150
Total number of specimens discharged to date*	1138

* Excludes corrosion coupons

Three of the four special quadrants containing BONUS transition joint tensile specimens have been irradiated and discharged from the G-7 Hot Water Loop. Quadrants 176 and 177 which received exposures of between 2.0 and 2.5×10^{20} nvt (> 1 Mev) have been tested. Specimens from both the coextruded and the explosive welded tubes were tested at room temperature and at 550° F. Six specimens from Quadrant 177X exposed for 725 hr in the out-of-reactor loop were also tested at the same temperatures. Preliminary results indicate there is no change in properties due to exposure in the out-of-reactor loop. Tests on the irradiated samples also resulted in failures entirely through the Zircaloy-2 portion of the specimen. These failures were of an extremely "brittle" nature for both the room temperature and 550 F tests. Uniform elongations of less than 1% and total elongations of less than 2% were observed. In some tests it was impossible to measure any plastic deformation.

Metallographic studies are in progress to determine the nature and cause of this type of behavior. Two additional quadrants are scheduled to be tested after exposures of approximately 5.0×10^{20} nvt (> 1 Mev). It is concluded from the results of these tests that the integrity of both types of joints is acceptable. However, the joining processes and/or prior fabricating steps have resulted in very low ductility in the Zircaloy-2. This low ductility is further reduced by nominal neutron exposures and should be considered of prime importance in developing these joints.

IRRADIATION DAMAGE TO NICKEL-BASE ALLOYS - I. S. Levy and
K. R. Wheeler

The combination of oxidation resistance and high temperature strength places the nickel-base alloys among the most promising materials for nuclear reactor application. These alloys, however, generally suffer embrittlement when irradiated at high temperature. The effort to modify microstructure of nickel-base alloys to reduce their adverse reaction to subsequent irradiation is continuing. These structural modifications are made by preirradiation thermal and/or mechanical treatments and are screened by tensile and stress-rupture tests and metallographic examination on both control and irradiated material.

During the past period it was decided that to achieve the optimum in specimen reliability, a program of pre- and postfabrication quality control tests should be initiated. The procedure now being used consists of the following steps:

- (1) The stock material is tested ultrasonically to screen out areas containing voids or other regions of inhomogeneity.
- (2) The specimens are fabricated from approved material.
- (3) When returned from the fabricator, they are visually inspected for general workmanship.
- (4) They are then placed on an optical comparator to determine that design specifications are met.
- (5) Surface condition is determined using the zyglow dye technique.
- (6) All nonrejected specimens are then ultrasonically tested for internal integrity (the additional ultrasonic testing is considered worthwhile since this test on the small diameter specimens would be more sensitive than it was on the larger stock material).

During the initial testing and setting up of necessary equipment, two of eight rods of stock material of Inconel X-750 were found to cause alteration of the ultrasonic radiation to a greater extent than the others. Optical metallographic examination of a specimen from each batch revealed that the rods exhibiting ultrasonic alteration had generally larger grains than the others with areas of inhomogeneity of grain size (Figure 4. 18). Such grain size

(a) Normal Material
ASTM Grain Size < 8

100 X (b) Large Grained Material 100 X
ASTM Grain Size 5-6

FIGURE 4. 18

Grain Size Variation in Stock Material of Inconel X-750

Neg. 4L2098B, 4L2097B

variations and inhomogeneity could have important effects on the results of tensile and stress-rupture tests. It is possible, however, to select fine grain homogeneous material from the stock using the ultrasonic test.

Solid-Solution Hardened Alloys

Results previously reported⁽¹⁾ on the room temperature tensile properties of control and irradiated specimens of Hastelloy X-280 led to the hypothesis that high temperature irradiation of solution-treated material causes carbides to precipitate at irradiation damage sites within the matrix rather than massively at grain boundaries, leading to increased strength and improved ductility. An initial attempt at controlled precipitation of

(1) J. J. Cadwell, ed. Quarterly Progress Report - Metallurgy Research Operation, January, February, March, 1964, HW-81269 (General Electric Company, Richland, Washington). April 15, 1964.

carbides by using a combination of preirradiation cold work (26%) and aging treatments was shown⁽¹⁾ to have successfully dispersed the carbide phase, but with ductility loss.

Tensile and stress-rupture specimens representing treatments using four lesser levels of cold work (0, 10, 15, and 20% with 24 hr age at 1000 or 1500° F) are now being irradiated at 540° F to a goal exposure of approximately 2×10^{20} nvt in the ETR. Other specimens are being readied for 120 and 1300° F irradiation. Metallography of these treatments are being processed.

Precipitation-Hardened Alloys

Specimens of Incoloy X-750 representing five different experimental heat treatments are currently being irradiated at 540° F to a goal exposure of approximately 2×10^{20} nvt in the ETR. Others are being prepared for 120 and 1300° F irradiations.

Irradiation of an alloy containing a precipitated phase may result in solution of the precipitates, formation of additional precipitates, or the disintegration of precipitates into small pieces by a combination of these two processes. The experimental treatments were chosen to give a range of particle size, from underaged to overaged conditions. Figure 4.19 shows electron micrographs of the microstructures resulting from these heat treatments. The matrix precipitate is $\text{Ni}_3(\text{Ti}, \text{Al})$ or γ' and the blocky grain boundary precipitate is M_{23}C_6 carbides. The effect of irradiation on the tensile and stress-rupture properties of irradiated material in these several conditions will be investigated.

(1) J. J. Cadwell, ed. Quarterly Progress Report - Metallurgy Research Operation, April, May, June, 1964, HW-82651 (General Electric Company, Richland, Washington). July 15, 1964.

(a) 2150° F/2 hr, air cool
1550° F/24 hr, air cool
1300° F/20 hr, air cool

(b) 2150° F/2 hr, air cool
1000° F/50 hr, air cool

10,000 X

FIGURE 4. 19
Inconel X-750 Heat Treatments

(c) 2150° F/2 hr, air cool
1550° F/24 hr, air cool
1000° F/20 hr, air cool

(d) 2150° F/2 hr, air cool
1550° F/24 hr, air cool
1650° F/20 hr, air cool

FIGURE 4. 19 (contd.)
Inconel X-750 Heat Treatments

10,000 X

(e) 2150° F/2 hr, air cool
1550° F/24 hr, air cool
1650° F/100 hr, air cool

FIGURE 4. 19 (contd.)
Inconel X-750 Heat Treatments

10,000 X

FRACTURE STUDIES - R. G. Hoagland

Eight annealed Zircaloy-2 double cantilever beam (DCB) specimens have been charged into the ETR G-7 hot water loop. A photograph of the quadrant assembly and a sample specimen is shown in Figure 4. 20. These specimens will be used to determine the effects of hot water irradiations on the fracture toughness and transition temperature for four exposure levels and two orientations. As has been pointed out in previous quarterly reports, the property of the DCB specimen is such that 30 to 40 measurements of the fracture toughness may be obtained from a single specimen.

FIGURE 4. 20

DCB Specimen Quadrant Assembly
for ETR G-7 Irradiations

Neg. 465622

As was mentioned in the previous quarterly report (HW-84573) one of the difficulties encountered in testing the DCB specimen at temperatures other than room temperature is that of measuring the temperature at the crack tip. This problem arises as the crack tip changes position down the length of the specimen as testing progresses. A cryostat which satisfies both the temperature control and measurement problem has recently been built. The cryostat is shown in Figure 4. 21. The temperature of the specimen is lowered by passing liquid nitrogen through channels in special

FIGURE 4. 21

Cryostat for Providing Precise
Temperature Control During Testing
of DCB Specimens
at Low Temperatures

Neg. 0650835-2

chill plates held in contact against the sides of the specimen by springs. Counter-acting this, a special conducting paint is applied to the other side of the plate. This paint acts as a 3000 W heater. The temperature at the crack tip is measured by one of seven thermocouples spaced $3/4$ in. apart and mounted in one of the plates. Based on the compliance of the specimen, one can make a quick calculation as to the position of the crack and select the closest thermocouple for control. By using solid state control and power units, temperature control to $\pm 0.4^{\circ}\text{C}$ has been obtained throughout the temperature range, 196 to 170°C .

Since interest in the DCB specimen has been developing for only a short time, there still exists a lack of knowledge concerning the proper design of the specimen and the effect that changes in design might have on the measured fracture toughness. For this reason experiments are now underway to ascertain the effect of varying the specimen thickness, depth, groove depth, and groove tip radius on the compliance and fracture toughness.

CHEMICAL METALLURGY

ATR GAS LOOP SUPPORT

Corrosion Studies - L. Charlot and R. E. Westerman

Previous tests have shown the rapid metal evaporation from Haynes Alloy 25 in 10^{-4} Torr vacuum at temperatures above 1000°C .⁽¹⁾ In an effort to evaluate the effect of an inert or reducing gas atmosphere on the rate of evaporation, static tests at 1120°C have been performed in which hydrogen was used to inhibit metal evaporation. Hydrogen addition, 10 Torr, decreased Haynes Alloy 25 rate by a factor of two. Additions of hydrogen to higher system pressures (above 80 Torr) essentially stopped the evaporation. Haynes Alloy 25 with air-formed film in 10 Torr hydrogen at 1120°C showed a small weight loss with time. The rate was less than that observed when a sample was pre-evaporated, then subjected to a system pressure of 10 Torr hydrogen.

Refractory metals have poor high temperature oxidation resistance. To establish the oxidation kinetics of niobium, as-received pure metal coupons have been exposed to continuously replenished oxygen atmospheres at 800, 900, and 1000°C . Table 5.1 summarizes the results.

TABLE 5.1

NIOBIUM OXIDATION RATES

<u>Temperature $^{\circ}\text{C}$</u>	<u>Initial Oxidation Rate $\text{mg}/\text{cm}^2 \text{ sec}$</u>	<u>Secondary Rate $\text{mg}/\text{cm}^2 \text{ sec}$</u>
1000		1.96×10^{-4} (50 to 1200 min)
900	2.44×10^{-4}	1.68×10^{-4} (175 to 1100 min)
900	2.48×10^{-4}	1.80×10^{-4} (130 to 1400 min)
800	2.38×10^{-4}	7.73×10^{-5} (50 to 800 min)

For each test, an oxygen pressure of 10 to 12μ was established. When the test was initiated, the pressure immediately decreased to approximately 1μ near the sample, increasing slowly as oxide scale was formed to an essentially constant pressure of 7 to 8μ near the sample. The initial

(1) J. J. Cadwell, ed. Quarterly Progress Report - Metallurgy Research Operation, April, May, June, 1964, HW-82651 (General Electric Company, Richland, Washington). July, 1964.

oxidation rate is controlled by the rate of arrival of oxygen to the sample. The secondary rate, however, compares favorably with rates appearing in the literature⁽¹⁾ at similar temperatures and pressures.

It has been reported⁽¹⁾ that the oxidation of niobium involves three stages: 1) an initial linear oxidation, 2) a subsequent parabolic stage, and 3) a final linear oxidation. Oxygen dissolution in the metal and thin film formation characterize the first stage of oxidation. To arrive at a relative picture of oxygen dissolution, a method proposed by Westerman⁽²⁾ was applied to the data at 900°C. The equation used to obtain maximum oxide solution during oxidation is

$$M_t = 2C_s \left(\frac{Dt}{\pi} \right)^{\frac{1}{2}}$$

where M_t = total amount of oxygen dissolved/unit area in time, t
 C_s = terminal solubility of oxygen in the metal at test temperature⁽³⁾
 D = diffusivity of oxygen in the metal at temperature. ⁽³⁾

The following table compares this maximum dissolvable amount to the experimental weight gain.

TABLE 5. 2
 COMPARISON OF TOTAL WEIGHT GAIN
 WITH OXYGEN DISSOLVED IN NIOBIUM

Time, min	M_t , mg/cm ²	Weight Gain, mg/cm ²	M_t /Weight Gain
1	0.086	----	----
5	0.192	0.058	3.31
10	0.271	0.134	2.02
50	0.606	0.751	0.81
100	0.757	1.493	0.51
500	1.916	5.793	0.33

The data obtained makes two pertinent points concerning niobium oxidation. The first is the obvious affinity niobium has toward oxygen. The second is the role of mass transport in partially controlling the kinetics of reaction.

(1) Per Kofstad and Svein Espavik. "Low Pressure Oxidation of Niobium," Electrochem. Soc. Journal, vol. 112, pp. 153-160. 1965.

(2) R. E. Westerman. High Temperature Oxidation of Zirconium and Zircaloy-2 in Oxygen and Water Vapor, HW-73511 (General Electric Company, Richland, Washington). April, 1962.

(3) Calculated from data of: R. A. Pasternak. High Temperature Oxidation and Nitridation of Niobium in Ultrahigh Vacuum, TID 19489. February, 1964.

Dynamic Materials Testing Apparatus - Oxidation Tests - R. A. Thiede,
R. E. Westerman

Samples of Hastelloy X, Haynes Alloy 25, and Cb-1 Zr have been exposed to high-temperature, high-velocity untreated Grade A helium. All tests have been done at 2050° F, at a helium pressure of 350 psi, and a flow rate of about 525 fps. The primary purpose of the tests has been to determine the effect of gas velocity on the oxidation rate of candidate ATR structural materials. No attempt has been made to date to purify the gas to the extent necessary to keep the sample clean, for the study of erosion and evaporation; this will be the subject of future tests.

A comparison of superalloy oxidation rates in the DMTA with those obtained in a static system by means of a recording microbalance are shown in Figures 5. 1 and 5. 2. The oxidant partial pressure in the DMTA over the sample surface does not stay constant during a run, but varies ordinarily from 2 to 3 ppm on loop startup (partial pressure 30 to 50 μ) to an undetectable level after the loop has attained temperature (this could be a partial pressure of oxygen as high as 10 μ). Because of this range of oxidant pressure in the DMTA, several curves from static-atmosphere investigations are included in Figures 5. 1 and 5. 2 for comparison to

DMTA data points. It can be seen that the effect of high velocity on the sample surface has little if any effect on the oxidation rate of Hastelloy X and Haynes Alloy 25 in the gas purity and temperature range investigated.

FIGURE 5. 1

Comparison of Oxidation Rates of Haynes Alloy 25 in the DMTA and Static Systems

FIGURE 5. 2

Comparison of Oxidation Rates of Hastelloy X
in the DMTA and Static Systems

A sample of Cb-1 Zr was exposed to 2050°F gas at 350 psi and 525 fps for 285 hr. It was found that the sample formed a tenacious oxide film, with no evidence of the spallation found during oxidation of cobalt- or nickel-base superalloys. The sample showed a weight gain of 2 mg/cm², and an average increase in the diameter (nominally 0.25 in.) of 0.004 in. Upon re-exposure in the loop under similar test conditions, the sample showed extensive oxide spallation, primarily associated with the outer layer of a duplex scale.

Contaminant In-Leakage - R. A. Thiede, R. E. Westerman

Air in-leakage by molecular flow through very small pores into a pressurized system has been the subject of much discussion, as the mechanical integrity of the ATR high-temperature gas loop components and the refractory metal samples exposed in the reactor may well depend on the magnitude of the in-leakage phenomenon.

In order to evaluate the in-leakage effect in the case of the DMTA, the system was filled with helium to 340 psi. The helium was purified to 0.3 ppm O₂ by a zirconium getter bed, then continuously circulated through the system for 5 days. All loop components were maintained at room temperature. At the end of this time, the oxygen level of the gas had increased only 1 ppm, although the leaks present in the system had allowed 25% of the helium to escape. This first experiment tends to make in-leakage appear to be unimportant as a possible source of contamination.

Intersite Comparison of BNW Gas Chromatograph - D. W. Shannon

The gas chromatograph developed by BNW for the ATR gas loop has been used to analyze a helium gas standard shipped to BNW for inter-comparison of analytical accuracy and sensitivity. The BNW analysis of this gas is shown in Table 5. 3 along with analyses obtained at three other sites. These results show the BNW chromatograph results to be close to the average. However, the significant comparisons are the BNW peak heights versus a thermal conductivity chromatograph peak heights. The BNW chromatograph is more sensitive, especially for H₂ and CO.

TABLE 5. 3

COMPARISON OF ANALYSES FOR A STANDARD GAS MIXTURE

	Analysis of Gas Standard, ppm					BNW* Ionization Detector Peak Height	Thermal Conductivity Peak Height	Peak Height Ratio $\frac{h_{\text{BNW}}}{h_{\text{T. C.}}}$
	Site	Site	Site	BNW	Four-Site Average			
H ₂	74	42	24-30	59	51	1104	26	42
O ₂	12.4	4	11-22	11	12	912	109	8.4
N ₂	21	24	22	17	21	436	281	1.6
CH ₄	13.8	10	15	16	14	1232	143	8.6
CO	13.6	20	16	12	15	480	not detected	∞

*Chart units for 5 cc samples - 100 = full scale on recorder.

N-REACTOR CORROSION AND HYDRIDING

Exposure of Zircaloy-2 to Low Temperature Moist Hydrogen -

D. W. Shannon

It is generally believed that H_2O inhibits the hydriding of Zircaloy-2 by forming an impermeable oxide film. An exploratory experiment was undertaken to test the possibility that, at low temperature, defects in the oxide film might not effectively "heal" because of the slow reaction of zirconium with H_2O at 100 or 200°C. Zircaloy-2 coupons with various pretreatments were exposed at temperatures of 100 and 200°C to 1 atm H_2 with 25 mm Hg H_2O added. A flow of a few ml/min was maintained. The pretreatments and hydrogen pickup results are given in Table 5. 4.

TABLE 5. 4

HYDROGEN PICKUP OF ZIRCALOY-2 IN 1 atm MOIST HYDROGEN
(97% H_2 - 3% H_2O Vapor)

Surface	Time, Days	100° C			200° C		
		Weight Gain, mg/dm ²	ΔH, ppm	Maximum Hydride Penetration, mils	Weight Gain, mg/dm ²	ΔH, ppm	Maximum Hydride Penetration, mils
Defected Rolled Sheet	26	---*	0	3	2. 1	2	18
	60	---	0	7	4. 2	3	24
	126	0. 7	15	9	6. 0	3	28
Vapor Blasted	26	---	7	3	0. 3	63	28
	60	---	1	5	4. 7	29	47
	126	0. 9	25	13	19. 6	32	50
Rounded and Etched	26	---	0	ND**	---	0	ND
	60	---	0	ND	---	4	ND
	126	---	0	ND	---	2	ND
Etched	26	---	0	ND	0. 3	2	ND
	60	---	0	ND	1. 1	4	ND
	126	0. 2	0	ND	1. 3	2	ND
Preauto- claved 20 mg/dm ²	26	---	4	ND	---	2	ND
	60	---	1	ND	---	7	ND
	126	0	8	ND	---	6	ND
Preauto- claved 50 mg/dm ²	26	---	2	ND	0. 6	20	ND
	60	---	8	ND	0. 8	30	ND
	126	---	6	ND	1. 5	49	ND
Preauto- claved 200 mg/dm ²	26	---	3	ND	---	31	ND
	60	---	7	ND			
	126	---	2	ND			

*--- Weight Gain - less than 0. 1 mg/dm²

** ND -Not detected

These data show clearly that hydriding occurred in the samples with mechanically disturbed surface layers at both 100 or 200°C. No hydriding was detected in samples which were etched or etched and preautoclaved 72 hr. Samples with thick, post-transition films also picked up a little hydrogen, but no detectable hydride case formed.

A typical hydride case formed in 126 days at 100°C is shown in Figure 5.3 and in 126 days at 200°C, Figure 5.4. These hydrided areas occur at holes punched in the samples. Hydrided spots formed on vapor blasted surfaces after 126 days at 100°C are shown in Figures 5.5 and 5.6. Measurements of the depth of hydriding at 26 and 60 days (Table 5.4) indicate the hydriding began immediately after the start of the test and continued through 126 days.

Where prior existing oxide films have been mechanically damaged (by hole punching or vapor blasting) and temperatures are too low for protective film formation, hydriding can occur at temperatures as low as 100°C in the presence of water vapor.

FIGURE 5. 3

Hydride Case in Punched Hole of As-Rolled Zircaloy-2 Sheet
(1 atm, 97% H_2 , 3% H_2O , 100° C, 126 days) 75X

FIGURE 5. 4

Hydride Case in Punched Hole of As-Rolled Zircaloy-2 Sheet
(1 atm, 97% H_2 , 3% H_2O , 200° C, 126 days) 75X

Neg. 475639, 465726

FIGURE 5. 5

Hydride Case in Punched Hole of Vapor Blasted Zircaloy-2 Sheet
(1 atm, 97% H_2 , 3% H_2O , 100° C, 126 days) 75 X

FIGURE 5. 6

Hydride Spots on Surface of Vapor Blasted Specimen
(1 atm, 97% H_2 , 3% H_2O , 100° C, 126 days) 75 X

Neg. 465640A, 465640B

PRODUCTION REACTOR CORROSION AND COATING STUDIESBoridizing Steel Chain - R. E. Westerman

It has been recently suggested (J. P. Cooke, IPD, GE) that mild steel chain charged with boron be dropped into reactors for reactivity control during down-time. In order to charge the chain to the required boron level, several pack-boridizing compounds were tried: B_4C , amorphous boron, and Ferroboron (18% boron, remainder iron). The best results by far were obtained by packing the chain sections in Ferroboron, and holding at $1160^{\circ}C$ for 9 hr, or $1140^{\circ}C$ for 12 hr. The resulting chain is 80% "black" to neutrons by volume comparison with boron-steel balls now in use, a satisfactory degree of blackness.

A 40 ft length of chain has been boridized for pilot experiments. The microstructure of the chain is shown in Figure 5.7. The boron appears to be primarily associated with the ring of intermetallic particles under the sample surface. The resulting microstructure is advantageous for reactor applications, as the high-boron particles (probably boron carbides) are separated from the sample surface by a band of alpha-iron of low boron content, and so are protected from abrasion and loss in the reactor.

FIGURE 5. 7

Microstructure of Boridized Chain - 100 X

Neg. 465838A

MATERIALS ENGINEERING

GAS LOOP DEVELOPMENT

Model Loop Studies - L. J. Defferding

The installation of the cooling water jacket on the test section of the model loop was completed. The jacket is instrumented to provide data for the calculation of the heat flow from the test section. The jacket provides a 3/8 in. water annulus, and is open to the atmosphere at the top. The water is in contact with the outer wall of the test section, and the flow is parallel to the helium flow. Using the new jacket, another series of conductivity tests were run on the foil insulation. The results indicated a conductivity which was dependent on helium flow, similar to the previous reported run with the ceramic heat meter, but to a much smaller degree. The decreased effect of flow as a result of the lower skin temperature on the test section indicated that the helium leakage was not in the foil insulation but may have been through the annulus between the outer wall of insulation and the inner wall of the test section. The foil insulation was removed for inspection and repair of one joint with a deliberate gap of at least 1/16 in. Visual inspection indicated a definite flow pattern on the exterior of the foil insulation. The major portion of the leakage flow originated at the joint with a 1/16 in. gap, but some flow did come from the prototypical ATR joint. Further inspection indicated the flow dam installed by the vendor to prevent flow longitudinally through the foil was in place and intact. The joint with the gap was repaired by machining the foil to give the overlapping joint, and a flow dam was installed on the top section of the joint. The insulation has been installed, and another set of conductivity tests has been started.

The present loop is constructed with double contained pipe from the regenerative heat exchanger through the heater to the test section, without bellows for differential expansion. The loop has been thermal cycled 70 times between room temperature and various heater outlet temperatures up to 2125° F. The shroud piping in the double contained section has operated up to 1150° F. No leakage has been detected between these two independent systems.

During a recent test run a maximum heater outlet temperature of 2125° F was held for several hours. Under these test conditions a momentary power failure caused a complete loss of power to the loop. The coast-down characteristics of the gas bearing compressor are such that flow was maintained for a period long enough to cool the heater outlet to a safe temperature before the flow stopped. The heater components never exceeded the temperature of operation before power failure.

The gas bearing compressor has accumulated a total of 400 hr of operation at various speeds up to 24, 500 rpm. During this operation the compressor has been started 100 times.

The next tests proposed for the model loop are, conductivity of a static helium layer, and a test to determine the steady state helium purity that can be obtained in the loop at high temperatures. The high purity run will require a gas make-up with less impurities than grade A helium; therefore, it will be run after the gas cleanup system is operating satisfactorily.

Columbium Sleeve Leak Test. Difficulty was encountered during the assembly of the test section for this test. The Invar seal galled severely at the threaded area where it screws into the stainless steel outer tube. The two pieces were taken apart and remachined to provide more clearance, and have been reassembled. Fabrication is continuing on the heaters for the test apparatus.

Model Gas Loop Heater - R. J. Evans

A new model gas loop heater has been designed to incorporate and investigate essential details of a compact two-stage heater presently being ordered for the ATR Gas Loop. The model loop heater has an outer ring of 12 Nichrome tubes and employs a flow reversal to an inner ring of six molybdenum tubes to produce an outlet gas temperature of 2100°F. Alumina surrounds all heating tubes to act as an electrical insulator and flow block to force the majority of gas through the inside of the heated tubes. Metallic foil insulation around the Alumina and an outer water jacket will reduce the temperature of the pressure-bearing shell to less than 500°F.

Material and/or components for all parts of the heater, with the exception of the Alumina, have been ordered. The original Alumina bid request, cancelled due to excessive costs, has been revised and additional bids have been requested. Savings of 25 to 50% over original bids are anticipated. Additional significant savings on heater components have been realized by relaxing the wall thickness tolerance on the molybdenum tubes. The cost of these tubes was reduced by 70% of the original low bid. Some material has arrived and fabrication of heater components has begun. Detail drawings of piping changes associated with the heater change are now being made. Figures 6.1 and 6.2 show the new heater and the appearance of the model loop after the heater has been installed.

FIGURE 6. 1

High Temperature Gas Loop Heater

FIGURE 6. 2

High Temperature Gas Loop

Helium Purification System - J. H. Hoage

A bypass helium purification system was installed on the gas loop utilizing pressure drop available from the gas bearing compressor. It consists of:

1. A heated catalytic copper oxide bed for removal of hydrogen.
2. A water cooled heat exchanger.
3. Activated charcoal and 13X molecular sieve at the temperature of liquid nitrogen for the removal of N_2 , O_2 , CO , CO_2 , CH_4 , and H_2O .

Purity levels of less than 0. 1 ppm of H_2 , N_2 , O_2 , CO , CO_2 and CH_4 and 3. 5 ppm H_2O have been achieved with a loop temperature of $916^\circ C$ ($1680^\circ F$).

It is planned to install dual 13X molecular sieves operating at room temperature between the copper oxide bed and the low temperature beds to remove water and CO_2 .

The low temperature beds of activated charcoal and 13X molecular sieve will be enlarged and dual beds installed. This will permit one set of beds to be reactivated while the other set remains in the purification system.

Exterior Insulation for the ATR Gas Loop Hot Piping - J. P. Hickerson

Hastelloy X wire, felted and sinter bonded, is considered a candidate material for insulating the 1600° F piping of the ATR Gas Loop. The Architect Engineer is interested in this material because of its mechanical strength and freedom from impurities that could cause stress corrosion cracking.

Preparations for making the thermal conductivity tests are nearly complete. The specimen consists of two half cylinders 11 in. long by 5 in. OD and 3/4 in. wall thickness. Each half section has a 0.010 in. thick Inconel foil sheath sinter bonded to the outside surface. The density of the felted wire is 15% of the density of Hastelloy X. The method employed to measure the conductivity is conventional. A heater is placed inside and coaxial with the insulation cylinder with adequate precautions against end losses. Conductivity is then calculated from measured temperature drop across the wall thickness of the foil knowing the power input across a known length of the coaxial heater. An independent measure of radial heat flux is obtained by measuring the temperature drop across an insulator (K-23 firebrick) surrounding the felted metal insulation and having a known thermal conductivity.

Dissimilar Metal Welds - D. R. Ireland

The construction of the ATR gas loop will require the welding of thick wall sections of Hastelloy X-280 to Haynes 25 and AISI 316 stainless steel. The results were previously reported⁽¹⁾ on the welding procedure and properties of these weld combinations utilizing Hastelloy W filler metal. Hastelloy W has a long successful history in joining dissimilar high temperature alloys but the results of short time elevated temperature tensile tests of wrought Hastelloy W indicate a sharp loss in ductility near 1200°F. The results of tensile tests on wrought Hastelloy X indicate a similar but less severe loss in ductility at 1200°F and Hastelloy X filler wire has also been successfully used in joining dissimilar metals. The effect of aging on the ductility of Hastelloy X and Hastelloy W weld metals aged at temperatures in the low ductility temperature range of 1200°F to 1600°F has not been previously investigated. A testing program to evaluate the effects of prolonged aging on typical ATR gas loop thick wall dissimilar metal welds utilizing Hastelloy X and Hastelloy W filler wires is currently in progress. The aging is being done in inert gas at 1000°F and 1200°F for the AISI 316 stainless steel joints and at 1400°F and 1600°F for the Haynes 25 joints.

The relative ductilities of the welded joints are being determined from bend tests of 0.625 in. thick longitudinal weld specimens. The ductility is measured as the maximum elongation in the outer fibers of the material where cracking or failure was initiated. The bend testing of unaged, 500 hr aged and 1000 hr aged specimens is complete. The tensile properties of aged Haynes 25 and Hastelloy X-280 base metals, and Hastelloy X and Hastelloy W weld metals are also being determined. The tensile testing of the 500 hr aged material is complete and specimens of 1000 hr aged material are being machined. Weld specimens for aging periods greater than 1000 hr are currently being aged.

(1) R. L. Knecht. Dissimilar Metal Welds of Certain Superalloys and Stainless Steels, HW-78574 (General Electric Company, Richland, Washington). August, 1963.

All of the unaged, 500 hr aged and 1000 hr aged bend specimens initially failed in the weld metal. The variation of the bend ductility of both weld metals with a 500 hr age followed the corresponding variation of room temperature tensile test ductility depicted in Figure 6.3. As expected with aging a loss in ductility and increase in yield strength was observed. The effect of aging on the tensile properties of the weld metals at the aging temperatures is depicted in Figure 6.4. The curves indicate that the aging is considerably more sluggish in Hastelloy W than in Hastelloy X. Although both weld metals with a 500 hr age reach similar minimum room temperature ductilities, the tensile tests at the 1000 and 1200° F aging temperatures indicate that Hastelloy W has a more useful amount of ductility remaining than Hastelloy X. A 500 hr age at 1400 and 1600° F results in Hastelloy X being considerably more ductile than Hastelloy W when tested at these aging temperatures, but the low 10% elongation of Hastelloy W at 1400° F is still a useful amount of ductility. A comparison of the yield strengths of the unaged and 500 hr aged weld metals at the aging temperatures indicate that Hastelloy W has a consistently higher yield strength than Hastelloy X. The room temperature yield strengths also indicate a preference for Hastelloy W, with the exception of the 500 hr age at 1200° F where the yield strengths are 84,600 psi for Hastelloy X and 78,600 psi for Hastelloy W.

The effects of the 500 hr age on the yield stress and ductility of Hastelloy X-280 and Haynes 25 base metals at room temperature and elevated temperature are depicted in Figures 6.5 and 6.6. The effect of a 500 hr age at 1000° F on the yield stress and ductility of Hastelloy X-280 is quite small and in good agreement with the results of a 100 hr age at 1000° F reported by Levy and Wheeler.⁽¹⁾ The effects of a 500 hr age at temperatures greater than 1000° F on yield stress and ductility of Hastelloy X-280 are relatively small when tested at the aging temperature and quite pronounced when tested at room temperature. The room temperature yield stress of these aged Hastelloy X-280 specimens is not greatly affected but the room

(1) J. J. Cadwell, ed. Quarterly Progress Report - Metallurgy Research Operation - January, February, March, 1964, HW-86269 (General Electric Company, Richland, Washington). April 15, 1964.

FIGURE 6.3

Hastelloy X and Hastelloy W Weld Metals Room Temperature Ductility and Yield Stress Variation with 500 hr Age

FIGURE 6. 4

Hastelloy X and Hastelloy W Weld Metals Aging Temperature Ductility and Yield Stress Variation with and without 500 hr Age

FIGURE 6. 5

Hastelloy X-280 and Haynes 25 Base Metals Room Temperature Ductility and Yield Stress Variation with 500 hr Age

FIGURE 6. 6

Hastelloy X-280 and Haynes 25 Base Metals Aging Temperature Ductility and Yield Stress Variation with and without 500 hr Age

temperature ductility is reduced to a low of 12% which is still a useful amount of ductility. The effect of the 500 hr age on the ductility of Haynes 25 at the aging temperatures is an increase from 20% or less to greater than 40% at 1400 and 1600° F. The room temperature ductility of the aged Haynes 25 is affected much the same as is Hastelloy X-280 and a low ductility of 7.3% is reached at 1600° F.

The above results of the tests on the weld metals and base metals aged 500 hr suggest that Hastelloy W has better aging characteristics than Hastelloy X when used as filler wire for joining Hastelloy X-280 to AISI 316 stainless steel and Haynes 25. This suggestion is based on considerations of the weld metal yield stress being larger than the base metals yield stresses, the relative sluggishness of weld metal aging and the amount of useful ductility remaining in the weld metal and the base metals after an aging treatment. The tensile testing of the weld and base metals aged 1000 hr and greater than 1000 hr is continuing.

PRTR PRESSURE TUBES

Properties of Irradiated PRTR Pressure Tube - M. C. Fraser

Crack propagation of unirradiated and irradiated tube specimens has been indicated to be describable by the equation:

$$\text{STRESS (Crack Length)}^{-0.62} \approx 44,000 \quad (1)$$

The irradiated specimens had average exposures of 2×10^{20} nvt to 5×10^{20} nvt ($E > 1$ Mev) and all tests were done at room temperature.

Several additional tests of specimens having comparable neutron exposure have been performed at temperatures greater than 70° F. A room temperature test of a specimen which had significantly greater neutron exposure has also been performed.

At temperatures of 200 and 300° F, propagation of 0.750 in. long cracks in unirradiated tube specimens was preceded by plastic deformation. The amount of plastic deformation was greater at 300° F than at 200° F and it was manifest by local bulging of the tube wall. About 85% of the fracture was inclined to the tube surface approximately 45°, indicating failure mode during propagation was shear. The propagation for the 200° F test increased the length of the 0.750 in. long crack to 4 3/4 in., about 30% of the specimen test length, and for the 300° F test increased the length of the 0.750 in. long crack to 5 1/16 in., about 32% of the specimen test length. Comparison of these data with previously performed room temperature tests where propagation was 100% of the specimen test length indicates that a transition in the nature and extent of crack propagation occurs between room temperature and 200° F for unirradiated material.

An irradiated specimen was tested at 200° F. Propagation was preceded by only a slight amount of plastic deformation; the fracture appeared to be normal to the surface and only one end of the propagating crack arrested before reaching the limit of the specimen. The average neutron exposure of this specimen was 1.5×10^{20} nvt ($E > 1$ Mev). This test information suggests that the temperature for transition from brittle to ductile propagation for irradiated material is higher than for unirradiated material.

An irradiated specimen which had an average exposure of 1.5×10^{21} nvt ($E > 1$ Mev) was tested at room temperature with a 1.000 in. long crack. The stress at propagation was 55,500 psi which is about 11,500 psi greater than equation (1) would yield. Whereas previous tests on specimens irradiated to exposures up to 5×10^{20} nvt showed agreement with equation (1), at the exposure of 1.5×10^{21} the stress required to cause crack propagation has increased.

K-Reactor Welded Zircaloy-2 Process Tubing - P. M. Jackson

A series of burst tests at room temperatures and elevated temperatures were made on sections of welded Zircaloy-2 tubing for the Hanford K-Reactors. In general the strength of the tubing compares favorably with extruded and drawn tubing. At room temperature, failure is in the weld zone and at 300°C the greatest strain is in the weld zone and heat affected areas, but failure is opposite the weld zone. Report BNWC-51 describes the tests and results; it has been issued.

Irradiation Burst Test Facility. This test facility for evaluating the effects of reactor irradiation upon the pressure tubing has been operating since early 1964. A report, BNWL-6, entitled "Irradiated Tube Testing Facility, " describing this facility is awaiting distribution.

USA-AECL COOPERATIVE PROGRAM

Zr-Nb Pressure Tubing - P. J. Pankaskie

About 75 ft of Zr-2 1/2 wt% Nb alloy tubing has been received. This material was supplied in two cold-worked tempers - 30% and 60% CW. The amount of 30% cold-worked tubing produced was less than expected because some of the sections contained cracks on the inside surface. This initial lot of tubing was produced as follows:

1. Extrude at 790°C with an extrusion ratio of about 19 to 1, to produce an extrusion blank 3.00 in. OD x 0.300 in. wall thickness.
2. Tube reduce cold all extrusion blanks from 3.00 in. OD x 0.300 in. wall to 2.625 OD x 0.250 in. wall resulting in a cold reduction of about 30%.
3. Anneal one-half of the tubing at about 600°C.
4. Tube reduce from 2.625 OD x 0.250 in. wall to 2.375 OD x 0.180 in. nominal wall thickness resulting in a cold reduction of about 30%.

Four specimens of this tubing were burst tested. One room temperature and one 300° C temperature test was made with both the 30 and 60% cold-worked tubing. The results of these tests are summarized as follows:

<u>Zr-2 1/2 wt% Nb Burst Tests</u>			
<u>Material</u>	<u>Test Temperature</u>	<u>Ultimate Hoop Strength</u>	<u>Hoop Elongation</u>
30% CW	Room	124,000 psi	1%
	280°C	91,000 psi	1%
60% CW	Room	134,000 psi	1%
	295°C	1,000,000 psi	1%

In these tests the ratio of hoop to axial stress was two to one. In each test the specimens failed in a shear mode. In no case did the specimen fail end to end as is frequently the case with heavily cold-worked Zircaloy-2 when tested at room temperature.

Work is underway in an attempt to conduct pressure tube creep test while the specimen is exposed in a flowing 550° F water loop environment. For the first test, an assembly was made in which the pressure tube specimen is held concentric with two venturi rings. One venturi located around a thick walled portion of the specimen measures rate of water flow around the specimen; the other venturi, located around the thin walled gage section of the tube, provides a continuous indirect measure of strain. Modification of the 314 Building high pressure loop to contain the assembly has begun.

PROCESS TUBE DEVELOPMENTPostirradiation Examination of a C-Overbore Tube - R. J. Evans

A Zircaloy-2 C-overbore process tube was removed from C-Reactor after 2 yr use. General Electric Co., IPD requested that this tube be examined for irradiation effects. Two small pieces from the downstream end of the tube and one 3 ft piece from the center of the tube were sent to Radiometallurgy for examination. Borescope examinations indicated only superficial scratches on the interior of the tube. No effects of irradiation on structure or grain size were observed. One small area of localized hydriding was observed on a sample from the downstream end of the tube. Ring tensile tests indicate an increase of approximately 14% in ultimate strength due to irradiation. A BNWC report was issued to report results of the postirradiation examination.

N-REACTOR TUBE

Brittle Fracture of Pressure Tubing - R. C. Aungst and J. P. Hickerson

A need has developed in the study of brittle fracture of Zircaloy-2 pressure tubes for a method to control the fatigue growth of a crack in a piece of tubing. As a section of tubing is cycled to some selected elevated pressure, a successful method of measuring the crack extension per cycle will allow the operator to stop the test just before the crack reaches the critical length at which it would propagate. The test piece can then be cycled at a lower pressure and in this manner the entire curve for propagation stress versus crack length can be constructed using a single specimen. Such a technique will be extremely valuable when working with irradiated tubing specimens which are usually in short supply.

Acoustical methods for detecting crack growth are under investigation. Detection of small increments of crack growth has been demonstrated using accelerometers and piezoelectric strain gages as sensors. A wide frequency response is desirable in the detection equipment so that the optimum frequency in the system under study will be within its range. The piezoelectric strain gage has been shown to give a strong response at frequencies extending into the megacycle range.

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