DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

**Cover Sheet for a Hanford
Historical Document
Released for Public Availability**

Released 1995

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**

MASTER

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

RECEIVED

HW-37979

Metallurgy and Ceramics
Special Distribution

This document approved by
J. J. Cadwell, Manager

300 AREA
CLASSIFIED FILES

This document consists
of 133 pages. Copy No. 48
of 200 copies. Series

QUARTERLY PROGRESS REPORT
METALLURGY RESEARCH SUB-SECTION

DECLASSIFIED

APRIL, MAY, JUNE, 1955

Written by the Members of the Metallurgy Research Sub-Section

S. H. Bush - Physical Metallurgy
M. J. Sanderson - Pile Metallurgy
L. D. Turner - Radiometallurgy
W. R. Smith - Corrosion and Welding
L. A. Hartcorn - Metallography Laboratory
O. J. Wick - Plutonium Metallurgy

CLASSIFICATION CANCELLED AND CHANGED TO
TECHNICAL

August 15, 1955

BY AUTHORITY OF CG-PR-2
DS Lewis 9-3-92

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

BY JE Savely 10-6-92
VERIFIED BY R. Schmitt 10-13-92

NOT UCNI

Operated for the Atomic Energy Commission by the
General Electric Company under Contract #W-31-109-Eng-52.

DECLASSIFIED DATA

This document contains restricted data as classified
in the Atomic Energy Act of 1954. It is submitted
for disclosure in its content in any manner to
an unauthorized person is prohibited.

WARNING - PRELIMINARY REPORT

This report contains information of a preliminary nature and
is prepared primarily for the use of Hanford Works technical
personnel. This report is subject to revision or further
checking or collection of additional data.

DECLASSIFIED

Route To:	Payroll No.	Location	Signature	Date
ETC				
H. D. Thorne	15672	326	[Signature]	9-15
H. D. Thorne	15672	326	[Signature]	
H. D. Thorne	15672	326	[Signature]	

MASTER

7-16-86

Metallurgy - Ceramics
Special Distribution

<u>COPY NUMBER</u>	<u>INTERNAL DISTRIBUTION</u>
1	F. W. Albaugh - E. A. Eschbach
2	C. R. Anderson
3	J. M. Atwood
4	J. A. Ayres
5	R. H. Beaton - J. L. Boyd
6	R. S. Bell - R. E. McGrath
7	J. A. Berberet - L. H. McEwen
8	C. W. Botsford - R. B. Hamilton
9	L. P. Bupp
10	S. H. Bush
11	J. J. Cadwell
12	M. W. Carbon
13	H. A. Carlberg - J. M. Fox, Jr.
14	V. R. Cooper
15	R. L. Dickeman - M. V. Davis
16	B. R. Elder - R. D. Switters
17	R. M. Fryar
18	O. H. Greager
19	A. B. Greninger
20	C. T. Groswith - E. Hilgeman
21	L. A. Hartcorn
22	W. M. Harty - O. H. Pilkey
23	O. F. Hill - K. M. Harmon
24	H. H. Hopkins, Jr.
25	R. T. Jessen - O. F. Beaulieu
26	M. Lewis
27	W. K. MacCready - J. H. Warren
28	W. M. Mathis - W. W. Windsheimer
29	J. S. McMahon - W. W. McIntosh
30	W. N. Mobley - E. G. Pierick
31	J. F. Music
32	E. W. O'Rorke
33	W. J. Ozeroff - J. L. Carter
34	T. Prudich - L. W. Finch
35	W. H. Reas
36	R. B. Richards - O. V. Smiset
37	M. J. Sanderson
38	H. P. Shaw - A. B. Carson
39	W. R. Smith
40	J. T. Stringer
41	G. W. Stuart - J. E. Faulkner
42	L. D. Turner
43	W. P. Wallace
44	O. J. Wick
45	E. C. Wood
46	F. W. Woodfield
47	W. K. Woods - P. F. Gast
48-52	Extra
53	300 File
54	Yellow Copy

DECLASSIFIED

Metallurgy - Ceramics
Special Distribution

COPY NUMBEREXTERNAL DISTRIBUTION**DECLASSIFIED**

55	AF Plant Representative, Wood-Ridge
56	Alco Products, Inc.
57	ANP Project Office, Fort Worth
58-63	Argonne National Laboratory
64	Armed Forces Special Weapons Project, Sandia
65	Armed Forces Special Weapons Project, Washington
66	Army Chemical Center
67-70	Atomic Energy Commission, Washington
71	Battelle Memorial Institute
72-75	Bettis Plant (WAPD)
76-78	Brookhaven National Laboratory
79	Bureau of Mines, Albany
80	Bureau of Ships
81-88	Carbide and Carbon Chemicals Company (ORNL)
89	Carbide and Carbon Chemicals Company (K-25 Plant)
90	Chicago Patent Group
91	Directorate of Research (WADC)
92-95	duPont Company, Augusta
96	duPont Company, Wilmington
97	Duquesne Light Company
98	Engineer Research and Development Laboratories
99-101	General Electric Company (ANPD)
102	Hanford Operations Office
103-104	Headquarters, Air Force Special Weapons Center
105	Iowa State College
106-109	Knolls Atomic Power Laboratory
110-111	Los Alamos Scientific Laboratory
112	Mallinckrodt Chemical Works
113	Materials Laboratory (WADC)
114	Mound Laboratory
115	National Advisory Committee for Aeronautics, Cleveland
116	National Lead Company, Ohio
117	Naval Research Laboratory
118	New York Operations Office
119-121	North American Aviation, Inc.
122	Nuclear Development Associates, Inc.
123	Nuclear Metals, Inc.
124	Patent Branch, Washington
125-128	Phillips Petroleum Company (NRTS)
129	Powerplant Laboratory (WADC)
130	Pratt & Whitney Aircraft Division (Fox Project)
131	Princeton University
132	Sylvania Electric Products, Inc.
133	USAF Project RAND.
134	U. S. Naval Postgraduate School
135	U. S. Naval Radiological Defense Laboratory
136-137	University of California Radiation Laboratory, Berkeley
138-139	University of California Radiation Laboratory, Livermore
140	Vitro Engineering Division
141	Walter Kidde Nuclear Laboratory, Inc.
142-206	Technical Information Service, Oak Ridge

DECLASSIFIED

QUARTERLY PROGRESS REPORT - METALLURGY RESEARCH

By

J. J. Cadwell

Reports previously issued are as follows:

1. HW-30114 - QUARTERLY REPROGRESS REPORT - METALLURGY UNIT - JULY, AUGUST, SEPTEMBER, 1953 - January 25, 1954.
2. HW-30896 - QUARTERLY PROGRESS REPORT - METALLURGY UNIT - OCTOBER, NOVEMBER, DECEMBER, 1953 - March 5, 1954.
3. HW-31799 - QUARTERLY PROGRESS REPORT - METALLURGY UNIT - JANUARY, FEBRUARY, MARCH, 1954 - May 10, 1954.
4. HW-32236 - QUARTERLY PROGRESS REPORT - METALLURGY UNIT - APRIL, MAY, JUNE, 1954 - July 16, 1954.
5. HW-33602 - QUARTERLY PROGRESS REPORT - METALLURGY UNIT - JULY, AUGUST, SEPTEMBER, 1954 - October 29, 1954.
6. HW-34455 - QUARTERLY PROGRESS REPORT - METALLURGY RESEARCH SUB-SECTION, OCTOBER, NOVEMBER, DECEMBER, 1954 - February 1, 1955.
7. HW-36054 - QUARTERLY PROGRESS REPORT - METALLURGY RESEARCH SUB-SECTION, JANUARY, FEBRUARY, MARCH, 1955 - May 16, 1955.

TABLE OF CONTENTS**DECLASSIFIED**

SUMMARY - Pages 2.1 through 2.8

PHYSICAL METALLURGY - S. H. Bush - Pages 3.1 through 3.49

Irradiation Effects

HAPO Irradiations - Mechanical and Physical Properties of Uranium - R. E. Hueschen, R. S. Kemper, L. J. Chockie, and W. S. Kelly	3.1
HAPO Irradiation PT-105-3N-Supp. A - R. E. Hueschen, R. S. Kemper, and L. J. Chockie	3.6
Structural Stability of Uranium Alloys - L. F. Kendall and R. G. Wheeler. .	3.6
Properties of Depleted Uranium - R. E. Hueschen	3.6
Thermal Conductivity of Uranium - R. L. Hales and R. E. Hueschen	3.6
Physical Properties of Uranium Subjected to Electron Bombardment - R. L. Hales and J. M. Tobin	3.7
Dimensional Stability of Uranium - W. V. Cummings, O. K. Shupe, and E. C. Watts	3.14
Capsule Irradiations at the Materials Testing Reactor - Mechanical and Physical Properties of Uranium - R. S. Kemper	3.19
The Effects of Low Temperature Irradiation on the Properties of Uranium - R. S. Kemper and R. E. Hueschen	3.19
X-Ray Diffraction Studies of Radiation Damage to Uranium - W. V. Cummings .	3.19
The Effect of Irradiation on the Properties of Thorium - L. J. Chockie . .	3.20

Zirconium Metallurgy

Effect of Irradiation on the Mechanical Properties of Zirconium - R. S. Kemper and W. S. Kelly	3.20
Use of Notch Bend Test to Detect Embrittlement - R. G. Wheeler	3.20
Impact Strength of Zirconium Exposed to Circulating Pressurized Water - R. G. Wheeler and W. S. Kelly	3.23
The Effect of Present Pile Atmosphere on Uncooled Process Tube Sections - R. G. Wheeler	3.23
In-Pile Reaction of Zirconium Alloys with Selected Gases - R. G. Wheeler. .	3.26
Zirconium Gas Reactions - L. F. Kendall	3.26
Creep Strengths of Cold-Worked Zircaloy-2 - L. J. Chockie and S. H. Bush. .	3.26

Structure Studies

Electron and Optical Microscopy - T. K. Bierlein and R. S. Kemper	3.26
Diffusion Studies - D. R. Green and T. K. Bierlein	3.36
Effects of Irradiation on the Crystal Structures of Zirconium and Molybdenum - W. V. Cummings	3.39
Influence of Specimen Geometry on Line Broadening and Peak Intensity in Uranium - W. V. Cummings	3.39

Facilities

Elevated Temperature Tensile Testing Unit for Irradiated Materials - R. E. Hueschen and D. C. Kaulitz	3.40
References	3.49

PILE METALLURGY - M. J. Sanderson - Pages 4.1 through 4.27

Fuel Element Fabrication

Insulated Fuel Elements - J. E. Minor, O. K. Shupe, and P. W. Dickson . . .	4.1
Notched, Cored, Unbonded Fuel Elements - P. W. Dickson, D. C. Kaulitz, and T. W. Evans	4.1
Combined Fuel-Target Element for U-233 Production - J. W. Goffard	4.2
Hanford-Size Uranium-Magnesium Fuel Elements - J. W. Goffard and P. W. Dickson	4.2

TABLE OF CONTENTS (CONT)

DECLASSIFIED

Fuel Element Evaluation

Thermal Stresses in Fuel Elements - K. R. Merckx	4.2
In-Pile Testing of Mechanically Bonded Fuel Elements - C. L. Boyd and G. A. Last	4.3
In-Pile Testing of Unbonded Fuel Elements - J. E. Minor and D. L. Zimmerman	4.3
In-Pile Testing of Insulated Fuel Elements - J. E. Minor and O. K. Shupe	4.7
MTR Fuel Element Testing Facility - T. W. Evans and D. C. Kaulitz	4.8
High Temperature Irradiation of Uranium Test Specimens - C. L. Boyd and J. E. Minor	4.9

Fuel Materials

In-Pile Testing of the Uranium-Magnesium Matrix Fuel Material - M. D. Freshley and W. S. Kelly	4.11
Thorium-Uranium and Thorium-Uranium Zirconium Alloys - J. W. Goffard and R. H. Todd	4.13
Uranium Oxide Fuel Material - W. E. Roake	4.16
Fission Product Studies - T. W. Evans	4.17
Reaction of Fuel Materials with High Temperature Water - J. E. Minor and D. C. Kaulitz	4.17
Reaction of High Temperature Uranium with Water - W. E. Roake	4.18
Grain Growth in Uranium and Uranium-Rich Alloys - J. E. Minor and W. E. Roake	4.18

Uranium Reduction Studies

Reduction of UO_2 with Calcium - W. B. Tolley	4.21
Study of the Coalescence of Uranium Metal in the Reduction Process - W. R. DeHollander	4.25

References	4.27
----------------------	------

RADIOMETALLURGY - L. D. Turner - Pages 5.1 through 5.26

Low Exposure Failures - D. P. O'Keefe	5.1
Examination of Cored Uranium Slugs - D. P. O'Keefe	5.1
Examination of Failed Unbonded and Mechanically Bonded Slugs with Point Closures - D. L. Zimmerman	5.3
The Effects of Irradiation on the Properties of Deformed Non- Ruptured Slugs - C. L. Boyd	5.6
MTR Capsule Irradiation - Examination of Capsules GEH-3-5-C and GEH-3-5-D - R. S. Kemper, W. S. Kelly, and E. C. Watts	5.11
Impact Strength of Zircaloy-2 Irradiated in Circulating, Pressurized Water - R. G. Wheeler and W. S. Kelly	5.11
Examination of Zirconium and Zircaloy-2 Tube Sections Irradiated While Exposed to Pile Gases - PT-105-516-SI - W. S. Kelly	5.14
Metallographic Cell - J. R. Morgan	5.14
Vacuum Cathodic Etching Procedures - G. R. Mallett	5.15
Macroetching - G. R. Mallett	5.15
Replication of Irradiated Specimens for Optical and Electron Microscopy - T. K. Bierlein, J. R. Morgan, and G. R. Mallett	5.17
327 Basin Corrosion Study - G. R. Mallett	5.23
Parting Lathe - E. C. Watts	5.25
NaK Opening Equipment - E. C. Watts	5.25

TABLE OF CONTENTS (CONT)

DECLASSIFIED

CORROSION & WELDING - W. R. Smith - Pages 6.1 through 6.16

Redox Phase II Corrosion Problems - N. D. Groves, M. C. Fraser, and W. L. Walker	6.1
Field Corrosion Tests in Underground Waste Storage Tanks - N. D. Groves, M. C. Fraser, and W. L. Walker	6.9
Heat-Transfer-Corrosion Tests to Determine the Corrosivity of Synthetic 2WW Purex Waste Acid Concentrate - M. C. Fraser	6.9
Factorially Designed Static Corrosion Tests - W. L. Walker	6.11
Bearing Studies - N. D. Groves	6.12
The Weldability of an Alloy and Low Alloy Steels - G. W. Riedeman and L. C. Lemon	6.13
Weld Joint Design for Butt Welding Pipe - G. W. Riedeman and L. C. Lemon .	6.13
Joint Design for Joining Tubes to Tube Sheets for Corrosive Radioactive Chemical Service - W. R. Smith	6.16

PLUTONIUM METALLURGY - O. J. Wick

Due to the restricted distribution list for plutonium metallurgy, this
portion of the Quarterly Report is issued as a separate document, HW-38376.

SECRET

SUMMARY

PHYSICAL METALLURGY

DECLASSIFIED

Irradiation Effects

One uranium tensile specimen irradiated to 620 MWD/T (1.75×10^{20} nvt) was tested at 285 C (545 F). The values obtained were: ultimate tensile strength 71,000 psi, 0.1% offset yield strength 70,000 psi, percent elongation (one inch gage length) 0.7, and modulus of elasticity 12×10^6 . These values are comparable to the as-irradiated values of specimens tested at room temperature which were: ultimate tensile strength 76,000 psi, 0.1% offset yield strength 71,500 psi, and percent elongation (one inch gage length) 0.36. The elongation of the specimen tested at 285 C was less than that of a specimen annealed at 700 C (1290 F) after irradiation, then tested at room temperature.

Experiments have been initiated to determine the structural stability of irradiated uranium-silicon alloy when this alloy is in the stable epsilon phase. Present work is limited to the evaluation of thermal expansion as a criterion for degree of epsilonization. This technique has not proven satisfactory.

The damage resulting from the bombardment of uranium with electrons in the 1-2 MEV range is to be evaluated using electrical resistivity and x-ray diffraction to determine the extent of the damage. Calculations indicate a threshold energy of about 1.25 MEV electrons is necessary to generate vacancy-interstitial pairs.

The final assemblies from PT-105-514-SI have been opened. This test has as its purpose the evaluation of dimensional stability on irradiation of uranium containing various types and degrees of orientation. Present results confirm those obtained previously where a high (020) orientation results in growth as does a (020)(110) duplex orientation with the (020) predominating. A random and a (020)(110) orientation with the (110) predominating are both stable. In one instance the growth was sufficiently great to cause the container to split. It was observed that there was a tendency for the density to decrease as a function of integrated flux.

Zirconium Metallurgy

The investigation of the mechanical properties and microstructure of zirconium prepared with several degrees of cold work, then irradiated to 190, 490, and 780 MWD/AT, is completed. Post-irradiation annealing of specimens exposed to 780 MWD/AT revealed that 100 hours at 300 C (570 F) did not completely remove radiation damage in the material with 0 percent cold work. Annealing of specimens initially containing 10, 30, and 40 percent cold work resulted in varying degrees of recovery of both radiation damage and cold work.

Specimens of annealed and cold-worked Zircaloy-2 were exposed to atmospheres of helium-air and helium-CO₂ at 500 C (930 F) at BMI. The gases were ionized and nonionized to determine the effect of ionization on reaction rate. Some of these specimens have been notch bend tested to determine if the conditions of exposure influences certain mechanical properties. It was found that fracture angle and fracture moment is a function of weight gain rather than of type of atmosphere or ionization state. Cold-worked specimens were tougher after exposure than were the annealed specimens. Apparently, ionization affects the reaction rate with CO₂ but has no effect when the atmosphere consists of air.

SECRET

An examination of the weight gain of Zircaloy-2 and zirconium after exposure at 410 C (770 F) to 897 MWD/AT in pile atmosphere (CO_2 -He) indicates that the reaction rates in dry air and carbon dioxide are comparable, and these rates appear to be independent of pile irradiation, based on a comparison with ex-pile results.

Ex-pile tests to determine the reaction rate of zirconium and Zircaloy-2 have been extended to 300 and 400 C (570 and 750 F). Experimental results after 2000 hours compare favorably with those obtained by extrapolation of data at higher temperatures, using activation energy as a basis for this extrapolation.

Structure Studies

Immersion of vacuum cathodic etched Zircaloy-2 and uranium in NaK followed by heating for 18 days at 600, 700, and 800 C has indicated that NaK does not obliterate the etched microstructure. It was observed that the NaK actually acted as an etchant, and a precipitate was observed in the Zircaloy-2 at 800 C. Such a precipitate has not been reported previously, and it has been assumed that Zircaloy-2 is a solid solution. In the case of uranium the phase transformations occurring caused grain growth and some cracking. In this instance the NaK again acted as an etchant, and the beta grain size so delineated was about 0.3 micron. This is comparable to the structure observed with vacuum cathodic etching rather than the size generally reported of 0.1 - 0.15 mm. The smaller size tends to agree with the size predicted on the basis of x-ray diffraction peak widths.

Capsules containing diffusion couples of U-Al and U-AlSi have been inserted in the MTR to aid in the evaluation of the diffusion rates of these materials. Laboratory experiments on diffusion couples where the contact surfaces were cleaned by various methods has confirmed the value of cathodic vacuum etching. Initial results with U-AlSi at 200 and 250 C indicate that there is little or no diffusion after times in excess of 100 hours. A comparison of the diffusion constant K as a function of temperature obtained here with those obtained at BNL and ORNL for the U-Al system indicates a comparable but lower slope in the case of ORNL which could be due to cleanliness of surface and a marked difference in the case of BNL. This difference cannot be accounted for at this time.

The use of a curved rather than a flat uranium specimen for the evaluation of line broadening, intensity change, or peak shift in x-ray diffraction patterns has been investigated. When the curvature fits the focal circle, it has been found that the resolution of immediately adjacent peaks is markedly improved together with an increase in intensity. This would appear to be particularly valuable in the evaluation of irradiation damage where the effects are essentially undetectable with the conventional flat specimen.

Facilities

A complete description is given of the various parts of the high temperature tensile testing unit used for testing irradiated specimens. The optical unit, vacuum furnace, grips, and method used for gage marking is covered.

DECLASSIFIEDPILE METALLURGYFuel Element Fabrication

A group of cored, unbonded, Hanford-size fuel elements have been fabricated for irradiation in the MTR Fuel Element Testing Facility. These slugs have prefabricated stress-risers in the form of internal and external longitudinal notches extending the entire length of the uranium slug. The pieces were canned in aluminum by conventional cold-canning point-closure techniques. Three of these specimens will be irradiated to failure in the MTR to determine the type of failure that may be induced by these simulated metal quality defects.

Twenty-five uranium-magnesium matrix slugs have been prepared for testing at Hanford. Initial results of several high temperature (240 C) flow laboratory tests of this material have been obtained. Defective fuel elements containing this material swell rapidly when placed in 220 C water. Flow stoppage in a simulated process tube was found to occur in about three minutes at 240 C. Additional tests at lower temperatures are in progress.

Fuel Element Evaluation

Study of the complex relationship between thermal stresses, irradiation effects, and fuel element failures is continuing. Analysis of creep data recently obtained at BMI indicates that a previously proposed model of material behavior is not applicable to uranium over the temperature range in which Hanford slugs operate (100-500 C). The proposed model is not general enough to handle the transition from one creep law to another that is observed to occur between 250 and 400 C.

In-pile tests of unbonded, point-closed Hanford geometry fuel elements have been carried out to determine the rupture resistance of unbonded slugs as well as to determine whether or not an aluminum-uranium bond is essential for the heat transfer requirements of Hanford fuel elements. Eight solid slugs have now been irradiated at specific powers up to 47 kw/ft and exposures up to 1015 MWD/T. These slugs showed no evidence of preferential corrosion at the point-closure nor of non-uniform heat transfer through the can wall. A tube charge of unbonded cored slugs was discharged last quarter after 280 MWD/T tube exposure because of failure of one of the specimens. Examination of the failure in the Radiometallurgy facility has been completed. It has been concluded that the slug failed because a faulty weld at the end plug of this slug permitted pickle solution (HNO_3) to get to the inside of the slug. The buildup of corrosion product during autoclaving and operation then led to failure of the slug.

The Fuel Element Testing Facility in the MTR resumed operation during the quarter after modification of the hot waste venting system of the MTR was completed. The new rechargeable B-block was put into use. Despite several minor difficulties, which are being remedied, it was found to permit the temporary removal of specimens for examination with a minimum of damage to the specimens.

In-pile testing of insulated slugs is being carried out in order to study the behavior of Hanford geometry fuel elements in which the uranium is maintained at elevated temperature during irradiation. Two insulated cored fuel elements have been undergoing irradiation in the MTR Fuel Element Testing Facility at 60-65 kw/ft since May 20. The calculated uranium surface and maximum temperatures are 380 C and 790 C, respectively. The specimens were removed from the reactor at 90 and

SECRET

DECLASSIFIED

200 MWD/T for measurement. The diameter of both specimens was found to have increased by 0.015 to 0.018 inch (1.1-1.3%) by the time of the first measurement with no further change being detected on the second measurement. It seems likely that this change resulted from inelastic thermal expansion rather than from radiation-induced growth. The exposure at the end of the quarter was 300 MWD/T. The irradiation is scheduled to continue to 800 MWD/T.

Small uranium rods have been irradiated for 24 hours in special assemblies under conditions such that the calculated temperature was 50 C at one end of the rod and 900 C at the other. The hardness was found to vary uniformly from 99 (Rockwell G) at the cool end to 88 near the hot end. The pre-irradiation hardness was 82.

Fuel Materials

Uranium-magnesium matrix fuel material is being studied for high burnup applications. In an experimental irradiation in the MTR, four 0.40-inch diameter specimens canned in Zircaloy-2 have been irradiated to 1000 MWD per ton of uranium, four to 5000 MWD/T, two to 10,000 MWD/T, and two are currently being irradiated to 20,000 MWD/T. The last two specimens are presently at about 16,000 MWD/T. One of the 10,000 MWD/T specimens was found upon decanning to have fractured transversely. The bend test curve for the other was identical, within the experimental error, with that of the 1000 and 5000 MWD/T specimens. Irradiation of two 1.12-inch O.D. matrix fuel elements containing 64 volume percent uranium shot and canned in Zircaloy-2 has continued uneventfully. These slugs are generating 43 kw/ft² and are scheduled for irradiation to 5000 MWD/T (to be discharged March 1956). The exposure at the end of the quarter is estimated at 1300 MWD/T.

Thorium-uranium alloys are being studied as candidate high burnup fuel materials. Study of the solubility of uranium in thorium by a metallography-autoradiography approach is continuing. Addition of small amounts of zirconium to the alloy did not appear to reduce the solubility of uranium. A ternary alloy was found, however, to have a high aqueous corrosion resistance.

Uranium oxide is being evaluated as a fuel material. Sixteen oxide pins, canned in Zircaloy-2, have been irradiated at the MTR and are being returned to Hanford for examination. The dependence of the behavior of the oxide on flux, exposure, packing density, and 235/238 ratio is being studied. Maximum temperatures (calculated) during irradiation ranged up to 2900 C. Experiments involving the creation of lattice defects in UO₂ compacts by the addition of small amounts of calcium ions have shown that this treatment increases the rate of densification on sintering. A zirconium canned Hanford geometry uranium oxide fuel element with a 0.025-inch diameter hole drilled in the can has been flow tested in the Elmo-4 recirculating loop (260 C demineralized water) for 54 hours with no apparent effect.

A recirculating water loop to be used for study of the rate of reaction between uranium heated to 500-700 C and water at 150-300 C has been received from the fabricator. Installation of the equipment is in progress.

Study of the growth of large columnar grains in uranium in which a temperature gradient across 660 C exists has continued. It has been found that the exaggerated grain growth does not occur in a 1.5% Si alloy and occurs in a 0.4% Cr alloy only in the portion of the slug which has been cooled from temperatures above 770 C. In unalloyed uranium it was found that even upon fast cooling (upwards of 400 C per minute at 660 C) the large columnar grains were still formed.

Uranium Reduction Studies

Processes for preparing particulate uranium for use in matrix fuel materials are being studied. The effect of a high temperature soak immediately after bomb reduction of UO_3 with calcium has been investigated. The yield of particles greater than 325 mesh was found to be improved slightly. Preliminary experiments indicate that adding the fine (-325) particles produced in one bomb reduction to the charge for the next reduction results in a five to ten percent better yield of +325 material.

It has been found that addition of calcium metal and barium chloride to corroded uranium scrap and turnings will produce coalescence of the uranium metal pieces to a button upon melting. It is believed that the barium chloride flux removes the calcium oxide from the surface of the uranium particles where, ordinarily, it prevents coalescence of the tiny droplets of uranium.

RADIOMETALLURGY

Examination of low exposure production slug failures was continued on two slugs from 100-H. One was typical of the current failures, and the other was an unruptured failure. A metallographic study of several transverse sections from the rupture revealed no abnormalities in metal quality or exposure effects and that the condition of the AlSi braze and compound layers conformed to the normal fabrication quality. The metallography and chemical analysis of the can conformed to the specifications for 2S aluminum. However, in a study of the corrosion effects on the can wall it was observed that the aluminum had severe intergranular corrosion attack in the portions of the can that had apparently not been adequately cooled during pile exposure. This corrosive attack was general throughout the "hottest" area of the slug surface and was most advanced in the area of actual rupture. Other isolated spots were found where the intergranular attack had penetrated half the can wall thickness. The intergranular corrosion was not found on the portions of the slug where cooling had apparently been normal. It is felt that diminution of the coolant flow over a portion of the slug surface had resulted in surface temperatures high enough to promote the intergranular attacks which eventually penetrated the can. The unruptured slug also had surface scale formation indicative of uneven cooling. Metallographic examination showed the intergranular corrosive attack in the hotter portion of the jacket. Intergranular corrosion has not been observed on other, previously examined, cocked, warped, or ruptured slug jackets.

The examination of three cored, AlSi-bonded, natural uranium slugs from PT-105-570A was completed. The longitudinal splits which had developed in the two unruptured slugs accompanied a deformation of the uranium which caused a diameter increase in one direction with a very slight decrease in the diameter perpendicular to this direction. The slug ends, containing the plugs welded in place, had undergone the minimum deformation. A few micro cracks were found, but nothing to indicate the start of fragmentation such as occurred on the rupture.

Examination of the unbonded, point closure failure from tube 2061-C and two unruptured pieces from the same tube was begun. This tube was irradiated under PT-105-584A to an exposure of 483 MWD/T. The jacket of the rupture was severely swollen at the cap end by the formation of uranium corrosion products. Both the cap and base were nearly detached from the slug by extensive circumferential tears in the jacket, which resulted from the formation of large quantities of uranium corrosion product at both ends. Appreciable necking down was associated with the tear at the base,

but very little was noted at the cap end. Several longitudinal cracks were found in the swollen area of the jacket, all of which formed with very little plastic deformation. Sectioning of the slug approximately 1-1/2 inches from the cap end revealed relatively large areas on the jacket surfaces which appeared to be "wrinkled". These areas were located between the base and center of the slug in both cases. The mechanism involved in the formation of these "wrinkles" is not known; however, it appears that they were formed in the pile since they were not observed before or after autoclaving.

Electrical resistivity measurements were conducted on samples which were cut from an eight-inch, beta heat-treated, normally discharged slug and previously tested for bend strength, hardness values, and density. The resistivity of the sample taken 1/16" from the slug axis increased 4.5 percent over the reported non-irradiated average value. A sample taken 7/16" from the slug axis showed an increase of 2.0 percent over the non-irradiated average value. Since these values are different than those that were prognosticated, three more samples were obtained for testing.

The effect of irradiation on Zircaloy-2 containing hydrogen was studied by comparing irradiated and non-irradiated samples as a function of impact testing temperature. It appears that pre-irradiation specimens containing less than 20 ppm hydrogen were unaffected by irradiations to 1.9×10^{19} nvt. The pre-irradiation samples with 100 ppm hydrogen broke at lower impact energies and had a higher transition temperature than the unirradiated samples. Chemical analyses are now being conducted to determine the actual hydrogen content after irradiation.

Improvements to the metallographic cell operation are being contemplated that will allow more complete utilization of the shielded enclosure and provide greater ease and accuracy in obtaining metallographic data. The vacuum cathodic and electrochemical etching equipment has proven quite satisfactory, but each requires approximately one hour of processing time.

Replicas of etched surfaces of irradiated zirconium and uranium were obtained on Faxfilm and examined by the cold laboratory metallograph and by the electron microscope. Many photomicrographs of the replicated surfaces are shown.

A study of the corrosion of aluminum in the 327 Building water storage basin was completed except for pit depth measurements. Average weight loss measurements indicated that the bottom of the basin produced the least amount of corrosion.

Tools to open cans containing NaK and preformed samples of irradiated materials were fabricated and operated during this period.

CORROSION AND WELDING

Redox Phase II Corrosion Problems

A preliminary survey of the corrosion problems incurred by the Phase II modification of Redox operations has been conducted. Tests designed to evaluate types 304L, 312, and 347 stainless steels and Grade A55 titanium as materials of construction for the Redox F-2 and D-14 concentrators and the H-4 oxidizer, using seven synthetic solutions representative of these vessels, have indicated the following conclusions: (1) no serious corrosion problem exists in the F-2 concentrator if backcycle alone is used and the corrosion problem in this vessel when both backcycle and precycle are used can be overcome by using type 304L stainless steel as the base material and type 308L

as the welding alloy, (2) a serious corrosion problem exists in both the D-14 concentrator and the H-4 oxidizer which can be partially overcome by a judicious selection of operating conditions, (3) titanium used as the material of construction would eliminate the corrosion problems in all three of the subject vessels.

Field Corrosion Tests in Underground Waste Storage Tanks

In order to provide long term corrosion rates of the current construction material (SAE 1070 Carbon Steel) and candidate materials of construction (Mayari-R, Yolo, Corten, and T-1) for underground waste storage tanks, facilities have been designed, constructed, and installed in a Redox and a Purex waste storage tank which will expose the subject materials to the actual neutralized process wastes for periods ranging from four months to ten years.

Heat Transfer Corrosion Tests to Determine the Corrosivity of Synthetic 2WW Purex Waste Acid Concentrate

A continuation of the study to evaluate materials of construction for the Purex acid concentrators which is described in HW-34455 has produced quantitative corrosion rates of type 312 stainless steel exposed to synthetic Purex 2WW waste concentrate for times ranging from 64 to 2051 hours and with specimen temperatures of 145 C. These data indicate that the corrosion rate of type 312 stainless steel becomes constant with respect to time after about 1000 hours' exposure to synthetic Purex 2WW waste concentrate and has a level of approximately 0.0025 inch penetration per month.

Factorially Designed Static Corrosion Tests

Studies to correlate the static corrosion tests in sealed glass capsules with the heat transfer test units are currently being performed. The consistency and reproducibility of the two testing methods show that a correlation between them should exist.

Bearing Studies

Two grades of chromium-molybdenum-silicide and two chromium electroplatings have been evaluated as potential materials for submerged process lubricated bearing surfaces. All three grades of chromium-molybdenum-silicide exhibit sufficient corrosion resistance to boiling 65 percent nitric acid to warrant trial installations of this material.

Ultra chrome and hard chrome platings were exposed to 30 and 65 percent nitric acid at temperatures of 25, 80, and 115 C. From these tests it is questionable if the platings would serve satisfactorily in any concentration of nitric acid above room temperature unless the plating covered the entire surface exposed to the acid.

The Weldability of an Alloy and Low Alloy Steels

Specimens of Mayari-R, Corten, Yolo, and T-1 welded with low hydrogen iron powder coated electrodes were subjected to free face and root bends, tensile tests, and longitudinal weld face bend tests as a measure of their weldability. The results of these tests indicate that the welds are generally as strong and ductile as the base material and the weldments show no tendency toward brittle failure.

DECLASSIFIEDWeld Joint Design for Butt Welding Pipe

An economic survey to compare the time and cost of butt welding pipe using a conventional bevel joint and a joint designed by the G.E. Welding Laboratory has been conducted. The "G.E." joint design appears to result in the most consistently acceptable joint with the least time and expense.

Joint Design for Joining Tubes to Tube Sheets for Corrosive Radioactive Chemical Service

Joints have been developed for joining tubes to tube sheets for corrosive radioactive chemical service. The joints are corrosion resistant and can be economically and easily produced.

DECLASSIFIEDPHYSICAL METALLURGY - S. H. BushIrradiation Effects

The objectives of these programs are to evaluate quantitatively the effects of neutron flux intensity, integrated neutron flux, irradiation temperature, testing temperature, and post-irradiation annealing on the mechanical properties, physical properties, dimensional stability and crystal structure of fissionable materials. The materials used for these studies will be exposed in a HAPO pile or in the Materials Testing Reactor at various temperatures and thermal and epi-thermal flux intensities.

HAPO Irradiations - Mechanical and Physical Properties of Uranium - R. E. Hueschen, R. S. Kemper, L. J. Chockie, and W. S. Kelly

One irradiated uranium tensile specimen, which was exposed to approximately 620 MWD/T, was tested at 285 C (545 F) using the elevated temperature tensile unit described elsewhere in this report. A test run was first made at 700 F (370 C) with 347 stainless steel. The curves obtained for 347 stainless steel at this temperature are shown in Figures 3.1 and 3.2. As shown in Table 3.1, the results are in good agreement with values given in the U.S. Steel Handbook, "Steels for Elevated Temperature Service."

TABLE 3.1

COMPARISON OF TENSILE PROPERTIES
OF 347 STAINLESS STEEL AT 700 F (370 C)

<u>Property</u>	<u>U.S. Steel</u>	<u>Present Work</u>
Ultimate Tensile Str. - psi	67,000	66,000
Yield Strength (0.2% offset)-psi	32,000	32,200
Elongation - %	35	33
Modulus of Elasticity - 10^6 psi	--	26

The irradiated uranium tensile specimen was then tested at 285 C (545 F). The stress-strain curve obtained is shown in Figures 3.3 and 3.4. It is interesting to note that there was very little change in the strength and elongation values obtained at 285 C (545 F) compared to room temperature values. It is also interesting to note that less elongation resulted at 285 C (545 F) than from the tensile specimen annealed at 700 C (1290 F) and tested at room temperature (Figure 3.4). The values obtained for this one tensile test at 285 C (545 F) are given in Table 3.2. It would appear that the modulus of elasticity obtained for this specimen is low, since at 300 C (570 F) the value of the modulus for unirradiated uranium is approximately 19×10^6 psi; however, more tests must be made before any definite conclusions can be drawn. The possibility exists that the specimen was slightly bent.

FIGURE 3.1. Stress-Strain Curve of 347 Stainless Steel Obtained at 371 C (700 F).

DECLASSIFIED

DECLASSIFIED

FIGURE 3.2. Stress-Strain Curve of 347 Stainless Steel
Tested at 371 C (700 F).

DECLASSIFIED

FIGURE 3.3. Stress-Strain Curve Obtained for Irradiated Uranium Sample 2AT Tested at 285 C.

FIGURE 3.4. Stress-strain curve of irradiated uranium tested at 285 C compared with curves obtained at room temperature.

DECLASSIFIED

TABLE 3.2

TENSILE PROPERTIES OF IRRADIATED URANIUM
TESTED AT 285 C (545 F)

Ultimate Tensile Strength - psi	71,000
Yield Strength (0.1% offset) - psi	70,000
Elongation in 1 inch - %	0.7
Modulus of Elasticity - 10^6 psi	12

HAPO Irradiation PT-105-3N-Supp. A - R. E. Hueschen, R. S. Kemper, and L. J. Chockie

There is nothing new to report on Supplement A of Production Test 105-3N, involving exposure of property specimens to levels of 150, 300, 620, and 1000 MWD/T. The 69 special assemblies are still awaiting scheduling by the Manufacturing Department.

Structural Stability of Uranium Alloys - L. F. Kendall and R. G. Wheeler

The effects of irradiation on the structure of some uranium alloys are being studied; present work is with epsilonized uranium-silicon. The epsilon phase, which has the nominal composition U_3Si , is formed below 930 C (1705 F) by peritectoid reaction of uranium and U_3Si_2 . Various physical properties are being tested as a function of epsilonizing heat treatment to determine which properties are sensitive to changes in the amount of the epsilon phase present. These properties will be used before and after irradiation to determine whether changes have resulted during exposure.

Thermal expansion has not been found to be promising. Dilatometric measurements between 25 and 800 C of a specimen epsilonized by heating for 4-1/3 days at 800 C, the as-received condition, were not significantly different from those observed after heating the specimen at 955 C (1750 F) for one hour to decompose the epsilon phase. Metallographic confirmation of the epsilon decomposition by this treatment has not yet been completed; it is possible that the thermal expansion coefficient was unchanged because the structure was unchanged, but this is considered unlikely.

Properties of Depleted Uranium - R. E. Hueschen

Work has been initiated to irradiate depleted uranium at HAPO and at the MTR to various exposure levels. Present plans call for two irradiations at HAPO at approximately 150 and 300 MWD/AT, together with higher exposures and flux intensities at the MTR. Tensile, hardness, electrical resistivity, x-ray, density, dimensional stability, and metallographic data are to be obtained from three different sizes of samples. The specimens will all be obtained from one section of a depleted uranium rod. This rod was rolled to 3/4 inch diameter at Fernald from depleted billet 34 which has a U-235 content of 0.028%.

Thermal Conductivity of Uranium - R. L. Hales and R. E. Hueschen

A literature survey of methods used in measuring thermal conductivity has been completed. As a result of this survey, specifications for construction were written for a thermal conductivity test unit to determine the thermal conductivity of irradiated uranium. An attempt was made to incorporate sufficient flexibility into the equipment to allow several thermal conductivity methods to be tested. Work with the existing thermal conductivity apparatus has continued. Calibration

accuracy has been improved by using silver foil as a thermal contact and by making use of calibrated thermocouple corrections. The desired limits of five percent maximum variation in results still have not been attained. Better calibration accuracy will be sought by fabrication of a heat shield having a geometry which approaches the heat flow of the specimen. This shield will lessen radial heat loss of the specimen by allowing a more sensitive thermal control of the surrounding vacuum.

Physical Properties of Uranium Subjected to Electron Bombardment - R. L. Hales and J. M. Tobin

A literature survey on the effect of electron bombardment on the properties of materials has been completed. Recent experiments have demonstrated a change in the electrical resistivity of graphite as a result of bombardment by 1 Mev electrons⁽¹⁾. It has also been reported that copper has been heavily damaged by such irradiation at low temperatures -150 C (-238 F). The damage was removed appreciably by annealing ten minutes at -23 C (-9 F)⁽²⁾. An experiment is proposed in which uranium metal would be bombarded with beta rays while held at the temperature of liquid nitrogen. Comparative electrical resistivity data collected before bombardment and after bombardment while annealing to room temperature may give significant data. A similar experiment is being contemplated in which x-ray diffraction techniques would be used. Examples of the type of information desired are: the number of vacancy interstitial pairs produced at any particular electron flux level, the temperature range over which annealing of defects occurs and the minimum energy required to displace an atom of uranium from a lattice site to an interstitial position.

The following approximate calculations were made in order to determine the feasibility of using monoenergetic 2 Mev electrons from the Van de Graff generator for the study of radiation damage to uranium.

There are two advantages to the use of electrons for irradiation rather than the light positive ions. Fast electrons of about 2 Mev energy will produce about a single interstitial-vacancy pair. The energy transferred to the struck atom is small, reducing the probability of uranium-uranium secondary collisions. Secondly, the threshold energy for head-on displacement collisions in the case of the fast electrons is about a thousand times larger than that for a proton, for instance. Therefore, the range of the electrons is much larger, and a more uniform damage can be obtained in thicker samples. However, the threshold energy is so high that β , defined as the ratio of the particle velocity to the velocity of light, approaches unity, and the electron velocity must be treated relativistically.

In traveling through a metal, a fast electron loses its energy by both elastic and inelastic collisions. The former can lead to displacements and the production of interstitial-vacancy pairs if the recoil energy exceeds a certain minimum energy, ϵ_D , or to the excitation of lattice vibrations which eventually degenerate into heat. The inelastic collisions account for the major part of the energy loss as the electron traverses the metal. The rate of energy loss, (dE/dx) inelastic, may be computed directly or may be obtained from a relationship such as that developed by Katz and Penfold⁽³⁾ by an empirical treatment of range-energy data.

(1) All footnotes are referenced at the end of this section.

One of the results which can be obtained by the proposed experiment is a measurement of the minimum energy, ϵ_D , which is necessary to cause an atomic displacement in uranium. In the absence of a direct measurement, the value of ϵ_D may be assumed to be about 25 ev. The minimum energy, ϵ_D , implies a certain threshold energy, E_T , which is the lowest energy the incident electron may have to cause a displacement in a head-on collision. The maximum kinetic energy, E_M , an incident particle of energy, E , can impart to a struck particle initially at rest is obtained from the theory of relativistic scattering in a coulomb field⁽⁴⁾:

$$E_M = \frac{E (E + 2 \mu c^2)}{\mu c^2 \left(\frac{M}{2\mu} + \frac{\mu}{2M} + 1 + \frac{E}{\mu c^2} \right)} \quad (1)$$

where M is the mass of the struck particle, μ is the mass of the electron, c is the velocity of light and the energies E_M and E do not include rest energy.

Since the maximum output energy of the on-site generator is 2 Mev, it must be determined if $E_T < 2$ Mev. Setting $E_M = \epsilon_D$:

ϵ_D	E_T
20 ev	1.06 Mev
25	1.23
30	1.37

Thus, a considerable variation in ϵ_D yields values of threshold energy well within the available range.

If the uranium atom is initially at rest, and the electron is deflected through an angle θ , the recoil energy received by the struck atom is:

$$\epsilon = E_M \sin^2 \frac{\theta}{2} \quad (2)$$

For each value of incident particle energy, E_0 , there is a critical angle of deflection, θ_D , below which value there can be no displacement of the struck atom from the lattice. θ_D is found by setting $\epsilon = \epsilon_D$ and determining E_M from equation 1.

E_0	E_M	θ_D
1.20 Mev	24.0 ev	-
1.23	25.0	1800
1.30	27.1	147
1.40	30.5	130
1.50	34.0	118
1.60	37.7	109
1.70	41.6	102
1.80	45.7	95.5
1.90	49.9	90.1
2.00	54.3	85.4

The maximum recoil energy is plotted as a function of electron energy in Figure 3.5.

~~DECLASSIFIED~~

FIGURE 3.5. The Maximum Recoil Energy Transferred to the Struck Uranium Atom in a Head-on Collision with a Fast Electron Plotted as a Function of the Electron Energy.

The differential cross-section for deflection of an electron through an angle θ by a coulomb field into the solid angle $d\omega$ has been determined in a wave mechanical treatment by Mott(5). The equation is not too reliable for elements with large Z . (For a more accurate result, the expansion given by McKinley and Feshbach(6) may be used, and the integrations may be carried out by the methods of numerical calculus in terms of the deflection angle θ .) Integrating between θ_1 and θ_2 ;

$$\int_{\theta_1}^{\theta_2} d\sigma = \pi \left(\frac{Ze^2}{2m_0 c^2} \right)^2 \frac{(1 - \beta^2)}{\beta^4} \left[\cot^2 \frac{\theta_1}{2} - \cot^2 \frac{\theta_2}{2} - 2\beta^2 \log \frac{\sin \frac{\theta_2}{2}}{\sin \frac{\theta_1}{2}} + \frac{2\pi\beta Z}{137} \left(\sin \frac{\theta_1}{2} + \operatorname{cosec} \frac{\theta_1}{2} - \sin \frac{\theta_2}{2} - \operatorname{cosec} \frac{\theta_2}{2} \right) \right] \quad (3)$$

Setting $\theta_1 = \theta_D$ and $\theta_2 = \pi$, the following cross-sections were obtained.

E_0	$\pi \sigma_{\theta_D}$
1.23 Mev	0
1.30	$3.33 \times 10^{-24} \text{ cm}^2$
1.40	13.1
1.50	25.8
1.60	41.0
1.70	56.4
1.80	71.3
1.90	86.1
2.00	100.4

The values of cross-section as a function of incident energy for an infinitely thin uranium sample are plotted in Figure 3.6. For a sample of finite thickness, the cross-section must be reduced because of range-energy losses resulting from inelastic collisions. The rate of energy loss with electron energy is shown in Figure 3.7. The upper curve was obtained from the empirical Katz-Penfold relationship:

$$x \text{ (cm)} = 0.412 \rho^{-1} E_0^{1.265-0.0954 \ln E_0} \quad (4)$$

(E_0 in Mev)

The lower curve shows the contribution to the energy loss caused by ionization, according to Heitler(7).

By means of equations (1,2,4) the cross-section for a particular incident energy can be found as a function of the sample thickness, as shown in Figure 3.8. Integrating from the surface at $x = 0$ to the thickness of the damaged area at $x = 0.0216 \text{ cm}$, the effective cross-section for displacement, Σ_D , is found to be $9.64 \times 10^{-25} \text{ cm}^2$. The number of displacements is then:

$$N_D = N \phi \Sigma_D \quad \text{atoms cm}^{-2} \text{ sec}^{-1}$$

where N is atoms of uranium per cm^3 , ϕ is the electron flux in electrons $\text{cm}^{-2} \text{ sec}^{-1}$ and Σ_D is the effective cross-section.

FIGURE 3.6. The Displacement Collision Cross-Section for an Infinitely Thin Uranium Sample Plotted as a Function of the Electron Energy.

FIGURE 3.7. Rate of Change of Energy with Range Computed from the Empirical Katz-Penfold Relationship and Compared to the Rate of Energy Loss Due to Ionization According to the Heitler Equation.

FIGURE 3.8. Decrease in the Displacement Collision Cross-Section as the Electron Passes Through a Uranium Sample of Finite Thickness Caused by Rapid Energy Loss.

For a 2.0 Mev beam of electrons at 50 $\mu\text{amps cm}^{-2}$

$$N_D = 9.64 \times 10^{-25} \frac{\text{cm}^2}{\text{e}} \times 4.73 \times 10^{22} \frac{\text{atoms}}{\text{cm}^3} \times 3.14 \times 10^{14} \frac{\text{e}}{\text{cm}^2 \text{sec}}$$

$$= 1.43 \times 10^{13} \text{ atoms cm}^{-2} \text{ sec}^{-1}$$

A circuit can be devised for measuring an increase in resistivity, $\Delta\rho$, of 10^{-11} ohm $\text{cm}^{(8)}$. In order to study annealing effects, it would be desirable to produce a $\Delta\rho$ of about 10^{-7} ohm cm . One atomic percent of displacements will produce about 10^{-6} ohm cm increase in resistivity⁽⁸⁾. If the irradiation is carried out at -190 C (-310 F), one can assume the back-diffusion of the interstitial-vacancy pairs to be negligible. Then, it will require about 18 hours of exposure to a 2 Mev beam of electrons at an intensity of 50 $\mu\text{amps cm}^{-2}$ to produce 10^{-7} ohm cm increase in resistivity.

Dimensional Stability of Uranium - W. V. Cummings, O. K. Shupe, and E. C. Watts

The study of the effects of irradiation on the dimensional stability of preferentially oriented uranium has continued⁽⁹⁾. The remaining four assemblies of the production test PT-105-514-SI have been opened and the eighteen cylindrical specimens measured for dimensional changes and also visually inspected. As in the case of the previously opened samples, the specimens in which the (020) planes were preferentially aligned perpendicular to the axis of the cylinder of uranium had increased in length and decreased in diameter. Samples that possessed a duplex (020)(110) type of orientation in general grew in length if the (020) orientation was predominant but remained stable if the (110) orientation predominated and was approximately twice as great as the (020). Randomly oriented samples were not observed to increase in length, but in one case an elliptically-shaped cross-section was definitely observed. The measurements of the samples are given in Table 3.3. Assembly number 2, which contained the elliptical specimen, number 16, ruptured during irradiation. The remaining samples in this assembly all possessed a high (020) orientation. The failure was a combination split and end failure. The bottom of the aluminum can was badly bulged and partially separated from the can wall. In addition, two longitudinal splits, 180 degrees apart, extended approximately two inches from the damaged end of the can as shown in Figure 3.9. Examination of the contents of this assembly disclosed that the uranium sample number 16 had wedged and diffused into the aluminum insert as a result of its elliptical deformation. The remaining samples, upon elongating, were restricted because of this phenomenon and were forced in the other direction. The resultant forces were sufficient to cause the rupture. Figure 3.10 shows the opened assembly and the corroded condition of the samples.

Four of the five samples in assembly number 3 possessed the duplex (110)(020) type of orientation with the (110) predominating by an approximate factor of two. The fifth sample, number 106, had a very high (020) orientation. Figure 3.11 shows the dramatic difference in the stability of these specimens. Number 106 has grown approximately 100 percent.

Four samples in assembly number 6 showed excessive longitudinal growth, pushed the base of the can into the shape of a cone and imbedded the cap-end sample into the cap of the can. Figure 3.12 shows the badly warped and distorted conditions of the specimens. The two end samples in assembly number 9 were observed to be the characteristic dark color of oxidized uranium, while the center sample was found to be bright, exhibiting the color of the unoxidized metal. An exhaustive examination

DECLASSIFIED

ASSEMBLY #2

FIGURE 3.9. A View of the End of the Ruptured Assembly. The bottom of the aluminum can is bulged and partially separated from the can wall. Another longitudinal split exists at 180 degrees from the one shown.

FIGURE 3.10. The Opened Ruptured Assembly. The four samples at the right were highly oriented and had increased substantially in length before water penetration caused the corrosion that is seen. Sample No. 16 has an elliptical cross-section but is virtually unchanged in length.

Photographs Unclassified

DECLASSIFIED

FIGURE 3.11. The four samples at the left possessed a duplex (110)(020) type of orientation in the rolling direction with the (110) type predominating. These samples were essentially stable. The sample at the right had a lone (020) type of orientation in the same direction and has increased in length almost 100 percent.

FIGURE 3.12. The samples shown in this opened assembly indicate the distortion that results when the growth of the preferentially oriented uranium is restricted. Note the bulged cap at the left.

Photographs Unclassified

of the assembly could not account for this difference in coloration. Figure 3.13 shows the conditions of these samples.

Density measurements were made on eight specimens chosen from this test. These measurements with their respective exposures are given in Table 3.4. The densities vary from 18.78 gm/cm³ at 300 MWD/T to 17.41 gm/cm³ at 1150 MWD/T with intermediate and higher densities at intermediate exposures. These samples are also being prepared for x-ray diffraction studies along with the elliptically-shaped sample previously mentioned.

TABLE 3.3

COMPARISON OF PRE- AND POST-IRRADIATION
DIMENSIONS OF URANIUM WITH VARIOUS ORIENTATIONS

Assembly Number	Sample Number	Diameter (inches)		Length (inches)		Exposure nvt x 10 ¹⁹	Remarks
		Before	After	Before	After		
2	16	0.359	0.293 0.390	0.995	1.068	13.6	Elliptical Cross-Section
	20	0.359	--	0.995	--	13.6	Corroded
	26	0.358	--	0.992	--	13.6	"
	31	0.358	--	1.004	--	13.6	"
	35	0.359	--	1.006	--	13.6	"
3	104	0.359	0.337	1.002	1.080	12.2	Good surface
	106	0.359	0.174	0.998	2.092	12.2	" "
	112	0.359	0.335	1.003	1.086	12.2	" "
	113	0.359	0.361	1.000	1.093	12.2	" "
	131	0.359	0.384	1.005	1.089	12.2	" "
6	21	0.359	0.248	0.995	1.724	24.8	Badly distorted
	32	0.359	0.284	1.011	1.608	24.8	" "
	36	0.359	0.266	1.004	2.021	24.8	" "
	101	0.359	0.418	1.003	1.023	24.8	" "
	105	0.359	0.239	0.949	1.716	24.8	" "
9	4	0.359	0.354	1.876	2.098	35.0	Dark color
	12-C	0.359	0.299	2.004	2.271	35.0	Bright color
	13	0.359	0.348	1.977	2.292	35.0	Dark color

DECLASSIFIED

FIGURE 3.13. The two end samples in this assembly have the characteristic dark color of oxidized uranium. The center sample is very bright, exhibiting the color of the unoxidized metal.

Photograph Unclassified

TABLE 3.4

DENSITIES OF IRRADIATED URANIUM FROM PT-105-514-SI

<u>Sample Number</u>	<u>Exposure, MWD/T</u>	<u>True Density, gm/cm³ at 25 C</u>
7	300	18.775±0.002
30	570	18.585±0.004
17	585	18.897±0.002
116	585	18.533±0.002
14-B	685	18.870±0.002
28	685	18.845±0.001
8	710	18.747±0.003
29	1150	17.411±0.003

True Density of unirradiated uranium = 18.89±0.002 gm/cm³ at 25C.

Capsule Irradiations at the Materials Testing Reactor - Mechanical and Physical Properties of Uranium - R. S. Kemper

The high flux levels available at the Materials Testing Reactor have been employed in long-term, elevated-temperature exposures of uranium test specimens. A NaK-filled Zircaloy-2 capsule and specimen assembly has been described⁽⁹⁾ in which the calculated temperature in the gage section of the sample is 300-400 C (570-750 F) during exposure. Four of these capsules (GEH-3-5)⁽¹⁰⁾ were exposed to two MTR cycles for an intended exposure of approximately 4.2×10^{20} nvt (1600 MWD/T). Recent data from the MTR indicate that the flux during exposure was much lower than that requested, resulting in an estimated exposure of 1.4×10^{20} nvt (530 MWD/T). The calculated temperature in the gage section of the samples based on this lower flux is approximately 125-150 C (260-300 F). Exposure data will be obtained on individual samples as examination of the material progresses. Two of these capsules have been opened, and the examination of the material is described in another section of this report.

The requested exposure of the four additional capsules (GEH-3-11)⁽¹¹⁾ awaiting charging in the MTR was changed in an attempt to obtain the irradiation conditions originally planned. These capsules were charged June 24 and are scheduled for discharge July 11, with an estimated exposure of 800 MWD/T. The gage sections of these samples are calculated to reach 300-400 C during irradiation.

Uranium tensile specimens, uranium split sleeves, and Zircaloy-2 capsules have been prepared for irradiation of test specimens at beta or gamma phase temperatures. Additional heat transfer calculations are being made for the assembly.

The Effects of Low Temperature Irradiation on the Properties of Uranium - R. S. Kemper and R. E. Hueschen

Specimens are now awaiting charging in the reactor.

X-Ray Diffraction Studies of Radiation Damage to Uranium - W. V. Cummings

There is nothing new to report this quarter.

The Effect of Irradiation on the Properties of Thorium - L. J. Chockie

There is nothing new to report this quarter.

Zirconium Metallurgy

The study of zirconium and Zircaloy-2 has dealt with two principal fields of endeavor. The first is concerned with the in-pile aspects where gas and thermal flux levels, integrated flux, pile atmosphere (either water or gas), temperature, and time are the variables, and such factors as weight gain, growth and mechanical and physical properties are investigated. The second field of endeavor deals with the ex-pile aspects. Here the effect of various gases such as helium, carbon dioxide, air, oxygen, nitrogen, and water vapor, separately or in combination, are studied at different temperatures for various times to determine changes in weight, growth, mechanical properties, or physical properties. In addition, techniques are being developed to better evaluate such factors as change in toughness and shift in lattice parameter with gas pickup.

Effect of Irradiation on the Mechanical Properties of Zirconium - R. S. Kemper and W. S. Kelly

The objective of this test is to determine the effects of irradiation at three exposure levels for samples of arc-melted Bureau of Mines zirconium prepared with several degrees of initial cold work. The tensile properties and hardness have been determined and the microstructure observed for the material exposed in a Hanford process tube at 50-60 C (120-140 F) for exposures of 190, 490, and 780 MWD/AT (5.7×10^{19} , 1.5×10^{20} , and 2.4×10^{20} nvt thermal).

Tests were completed during the quarter for samples with the longest exposure. One of the pair of tensile specimens at each cold work level from this exposure were vacuum annealed 100 hours at 300 C (572 F) prior to testing. Tensile properties were also determined for specimens having 10, 30, and 40 percent cold work prior to irradiation with a post-irradiation vacuum anneal of 160 hours at 350 C (662 F). One hundred hours at 300 C did not completely remove the radiation damage produced in the initially fully annealed material. The post-irradiation annealing of samples with prior cold work resulted in varying degrees of recovery of both the radiation damage and cold work that was dependent upon the degree of prior cold work. No changes were observed in the microstructure of material exposed to 780 MWD/AT. These tests completed the examination of this material, and a report covering the investigation is being written.

Use of Notch Bend Test to Detect Embrittlement - R. G. Wheeler

Changes in the mechanical properties of Zircaloy-2 produced by the high temperature reaction of this material with CO₂ and air have been measured by bend testing. The gas reactions with ionized and nonionized CO₂ and air were completed at BMI(12). A 45-degree 0.008 inch deep notch with a 0.001 inch root radius was pressed into the test strips; these strips measured 0.380 inch wide by 0.060 inch thick. Oxide on the top of the specimens was fractured during notching, but oxide (or scale) on the side opposite the notch was undisturbed. Exposure conditions for the specimens are summarized in Table 3.5, and the notch-bend fracture moments and fracture angles as a function of weight gain are shown in Figures 3.14 and 3.15, respectively. From these tests it appears that the bend ductility and the bend strength depend

FIGURE 3.14. Notch Bend Test. Curves show that fracture moment of notched Zircaloy-2 specimens depend more on wt gain than on type of atmosphere or ionization

UNCLASSIFIED

FIGURE 3.15. Notch Bend Test. Curves show that fracture angle of Zircaloy-2 bend test specimens depends more on weight gain than on type of atmosphere or ionization state.

more on the amount of reaction (weight gain) than on the type of atmosphere or ionization state. All the cold worked specimens were tougher after the gas reactions than were the annealed specimens, since, at a given weight gain, both the fracture moment and fracture angle were higher for the cold-worked specimens than was the case for the annealed specimens. BMI found that the Zircaloy-2 specimens reacted with the air mixture at essentially the same rate reported for dry air in Figure 3.17, and that the rate was independent of ionization or degree of cold work. Ionization plays an important role in the reaction of Zircaloy-2 with helium-carbon dioxide mixtures as illustrated by the log weight gain versus time curves in Figure 3.16 that were taken from the BMI data. The weight gain in the first ten hours averages ten times greater in ionized CO₂ than it does in nonionized CO₂, but the slopes of the curves are such that after extrapolation beyond 100 hours, the ionized gas will cause less weight gain than the nonionized gas.

TABLE 3.5

CONDITIONS OF EXPOSURE OF ZIRCALOY-2
TESTED AT BATTELLE MEMORIAL INSTITUTE

<u>Specimen</u> <u>Identification</u>	<u>Cold-Work</u> <u>Condition</u>	<u>Atmosphere</u>	<u>Ionization</u>	<u>Exposure</u> <u>Time</u>	<u>Temp.</u>
13	Anneal	None	--	0	--
17	65% CW	"	--	0	--
12	65% CW	80% CO ₂ -20% He	Ionized	9-1/2 hrs.	500 C
18	Annealed	" " " "	"	9-1/2 hrs.	500 C
15	65% CW	" " " "	Nonionized	13 hrs.	500 C
16	Annealed	" " " "	"	13 hrs.	500 C
19	65% CW	10% air-90% He	Ionized	16-3/4 hrs.	500 C
14	Annealed	" " " "	"	16-3/4 hrs.	500 C
11	65% CW	" " " "	Nonionized	12 hrs.	500 C
10	Annealed	" " " "	"	12 hrs.	500 C

Impact Strength of Zirconium Exposed to Circulating Pressurized Water - R. G. Wheeler and W. S. Kelly

This is reported in Section 5 (Radiometallurgy) of this document.

The Effect of Present Pile Atmosphere on Uncooled Process Tube Sections - R. G. Wheeler

Zirconium and Zircaloy-2 specimens exposed to pile gas in the 100-H "X" test hole have reacted with the gas at a rate dependent on temperature and independent of pile irradiation. One Zircaloy-2 and two zirconium process tube sections, together with sheet strips of both materials, were exposed at an average temperature of 410 C (770 F) to an integrated exposure of 897 MWD/AT (27 x 10¹⁹ nvt). The tube sections contained enough surface pits and gall marks to cause reaction rates based on calculated surface area to be in error on the high side. Zirconium and Zircaloy-2 appear to react with dry air and CO₂ at the same rate, according to the results of tests of about one month's duration (9). This is taken as justification for making a direct comparison between the reaction with pile gas (50% He-50% CO₂) and the reaction with air. The in-pile weight gain after 4000 hours at 410 C (770 F) was 1.7 mg/cm² for the Zircaloy-2 test strips, 2.5 mg/cm² for the Zircaloy-2 tube

FIGURE 3.16. Reaction of Zircaloy-2 with Ionized and Unionized CO_2 -20% He Mixture at 500 C Plotted from BMI Data.

FIGURE 3.17. Extrapolated Curves for Weight Gain and Penetration Versus Time at Various Temperatures

sections, 1.0 mg/cm² for the zirconium test strips, and 1.6 mg/cm² for the zirconium tube sections. From the curves shown in Figure 3.17, it can be seen that 4000 hours in air at 400 C would cause Zircaloy-2 to gain 2.9 mg/cm². Some light colored oxide was apparent at the bottom of pits and scratches on all the in-pile specimens, but none showed evidence that the breakaway stage was reached; however, a comparison between the 2000-hour and the 4000-hour in-pile data shows the reaction rate to be nearly linear or more like the rate after breakaway. Additional sets of specimens remaining in the test hole will be discharged after a year or more exposure.

In-Pile Reaction of Zirconium Alloys with Selected Gases - R. G. Wheeler

As predicted in the previous quarterly⁽¹³⁾, weight gain was excessive for the zirconium and Zircaloy-2 specimens irradiated in wet and dry air using gamma heating. A new charge is ready for insertion in the pile.

Zirconium-Gas Reactions - L. F. Kendall

Exposure of sponge zirconium and Zircaloy-2 specimens to dry air at 300 and 400 C (570 and 750 F) has been continued to verify extrapolation of similar data obtained at 500, 600, and 700 C (930, 1110, and 1290 F). After 2100 hours, the average weight gain was 0.8 mg/cm² for Zircaloy-2 at 400 C, compared to 1.5 mg/cm² predicted by extrapolation. At 300 C, the average weight gain after 2200 hours was 0.09 mg/cm² which is also the predicted value. Figure 3.17 shows curves in the range of 200 to 500 C for weight gain as a function of time. Penetration equivalent to these weight gains is also indicated. All of these curves, except that at 500 C, are derived by extrapolation from observations made at higher temperatures.

Creep Strengths of Cold-Worked Zircaloy-2 - L. J. Chockie and S. H. Bush

Insufficient data are available to present at this time.

Structure Studies

Special investigations of techniques to increase the knowledge of the fundamental properties of pile materials both fissionable and non-fissionable are described in this section. Included are x-ray investigations of the effect of irradiation on the crystal structure of zirconium and molybdenum, the development of vacuum cathodic etching to examine the micro and macro structure of various metals, particularly uranium, development and evaluation of replica techniques capable of being applied to irradiated materials, correlation of optical and electron microscopy, extensive investigation of the microstructure of uranium, and the diffusion in-pile and ex-pile of such metal couples as U-Zr, U-Mi, U-Al, U-AlSi, and Mg-Zr. These as well as similar investigations are discussed under "Structure Studies".

Electron and Optical Microscopy - T. K. Bierlein and R. S. Kemper

The study of radiation induced changes in pile materials requires specialized, controlled experiments. One of the problems associated with such test irradiations is specimen environment and specimen container. Recent laboratory experiments have indicated that the use of liquid NaK as a heat transfer medium and Zircaloy-2 as a capsule material satisfies many of the requirements imposed by the irradiation of preformed and pre-examined specimens.

Since one of the methods being used to evaluate radiation damage in pile materials is metallography, a metallographic study was made on the system uranium, Zircaloy-2, and NaK. Metallographic specimens of uranium and Zircaloy-2 were immersed in NaK, heated to temperatures of 600, 700, and 800 C (1110, 1290, and 1470 F) for a period of eighteen days, and then re-examined at room temperature for evidence of possible interaction.

Figures 3.18, 3.19, and 3.20 contain representative photomicrographs of the Zircaloy-2 specimens before and after the NaK treatment. The micrographs clearly indicate that little interaction or corrosion has taken place. On a microscopic scale, however, it is obvious that Zircaloy-2 is etched very selectively by NaK at elevated temperatures and that a recrystallization or precipitation and subsequent growth occurs preferentially at the scratch markings and at the grain boundaries. Representative micrographs of the specimen heated in NaK at 800 C are contained in Figure 3.21 and illustrate the type and degree of attack. Although the x-ray diffraction pattern of the high temperature specimen agreed with that of zirconium, with the exception of four weak, unidentifiable diffraction maxima, an electron diffraction study of the specimen surface is being made.

Interpretation of the NaK and uranium interaction is somewhat more difficult since the growth and distortion as well as the phase transformations complicate the comparison of the initial and final metallographic surfaces. As the micrographs contained in Figure 3.22 show, very little change in the appearance of the uranium has occurred after an 18-day exposure to NaK at 600 C. Figure 3.23, however, shows evidence of considerable grain growth of the beta phase structure, and as the micrograph (c) illustrates, a peculiar type of cracking has occurred. This specimen, after the NaK treatment, was replicated with Faxfilm in order to conduct some electron microscope studies on these cracks. Figure 3.24 depicts typical electron micrographs which show evidence of intergranular cleavage and a subgrain or very fine grain structure. Previous electron microscope studies of cathodically vacuum etched, beta heat treated uranium have indicated a subgrain structure with an apparent size of ~ 0.3 micron, which corresponds very closely to the size of the subgrains depicted in Figure 3.24. Calculations of the theoretical grain size of beta heat treated uranium on the supposition that the observed x-ray diffraction peak widths are due to a particle size effect alone have yielded a value of about 0.2 micron, which is in agreement with the electron microscope results. Apparently, beta heat treated uranium has essentially three grain sizes dependent upon the etchant and examination techniques employed; namely, a macro, a micro, and a sub-micro grain size. The metallographic comparison of the uranium specimen which was heated at 800 C in NaK is given in Figure 3.25. In this case considerable surface etching has occurred. Many of the original room temperature microstructural surface irregularities have been completely obliterated. A simple yet satisfactory technique has been used to determine the room temperature micro grain structure of the specimens which have undergone extensive growth and transformation. The specimen is electrolytically polished in a phosphoric acid; ethylene glycol, and ethyl alcohol bath for 30 seconds and then permitted to air oxidize. The electrolytic polish presumably contaminates the specimen surface such that oxidation occurs quite rapidly, whereas the cathodically vacuum etched surface or NaK treated surface oxidizes only very slowly. The resultant oxidized or heat treated surface yields a multicolored grain structure which designates the final room temperature structures.

Table 3.6 lists the weights of the uranium and Zircaloy-2 specimens used in the NaK metallographic studies and indicates the weight changes which occurred.

CONFIDENTIAL

(a) Initial appearance after
swab etch in HF-HNO_3 -
 H_2O .
B.F., oblique; 250X.

(b) Appearance after 18 days
in NaK at 600 C.
B.F., oblique; 250X.

(c) Same as (b) but 750X.

FIGURE 3.18. Photomicrographs Illustrating the Effect of NaK at 600 C
on Zircaloy-2.

Photographs Unclassified

DECLASSIFIED

- (a) Initial Appearance after
swab etch in HF-HNO₃-
H₂O
B.F., oblique; 250X.
- (b) Appearance after 18 days
in NaK at 700 C.
B.F., oblique; 250X
- (c) Same as (b), but 750X.

FIGURE 3.19. Photomicrographs Illustrating the Effect of NaK
at 700 C on Zircaloy-2.

Photographs Unclassified

(a) Initial appearance after swab etch in HF-HNO₃-H₂O. B.F., oblique; 250X
(b) Appearance after 17 days in NaK at 800 C. B.F., oblique; 250X
(c) Same as (b), but 750X.

FIGURE 3.20. Photomicrographs Illustrating the Effect of NaK at 800 C on Zircaloy-2.

Photographs Unclassified

DECLASSIFIED

FIGURE 3.21. Optical Micrographs of Zircaloy-2 After a 17-day Exposure to NaK at 800 C.
Illumination: Bright field, oblique
Magnification: 340X

Photographs Unclassified

DECLASSIFIED

(b) Appearance after 18 days in NaK
at 600 C.
B.F., oblique; 250X.

(a) Initial appearance of a cathodically
vacuum etched specimen of
annealed dingo uranium.
B.F., oblique; 250X.

FIGURE 3.22. Photomicrographs Illustrating the Effect of NaK
at 600 C on Uranium.

Photographs Unclassified

(a) Initial appearance of a cathodically, vacuum etched specimen of annealed dingot uranium. B.F., oblique; 250X.

(b) Appearance after 18 days in NaK at 700 C. B.F., oblique; 250X.

(c) Same as (b), but of an area showing cracks. B.F., oblique; 750X.

FIGURE 3.23. Photomicrographs Illustrating the Effect of NaK at 700 C on Uranium.

Photographs Unclassified

DECLASSIFIED

HW-37979

(a) 4400X.

(b) Same as (a), but 13,000X.

FIGURE 3.24. Electron Micrographs of a Replica of the Specimen Heated in NaK at 700 C shown in Figure 3.23. Note the intergranular cleavage cracks and the subgrain structure.

Photographs Unclassified

SECRET

UNCLASSIFIED

(a) Initial appearance of a cathodically vacuum etched specimen of annealed dingot uranium.
B.F., oblique; 250X.

(b) Appearance after 17 days in NaK at 800 C.
B.F., oblique; 250X.

FIGURE 3.25. Photomicrographs Illustrating the Effect of NaK at 800 C on Uranium.

Photographs Unclassified

DECLASSIFIEDTABLE 3.6URANIUM AND ZIRCALOY-2 WEIGHT CHANGES CAUSED BY NaK

<u>Specimen</u>	<u>Original Wt. in g</u>	<u>Final Wt. in g</u>	<u>Wt. in g</u>	<u>Period of Immersion in days</u>	<u>Temp. of NaK(°C)</u>
Zircaloy-2	1.8365	1.8375	+0.001	18	600
Zircaloy-2	1.8865	1.8875	+0.001	18	700
Zircaloy-2	2.0480	2.0487	+0.001	17	800
Uranium	14.7095	14.7097	+0.000	18	600
Uranium	14.6545	14.6542	-0.000	18	700
Uranium	14.5430	14.5426	-0.000	17	800

Diffusion Studies - D. R. Green and T. K. Bierlein

The effect of irradiation on the maximum diffusion rates of U/Al, U/Zr, and U/AlSi couples at particular temperatures can be studied by maintaining the two carefully cleaned metal surfaces in intimate contact during an in-pile diffusion anneal. The diffusion couples now being used consist of 1/4" thick x 1/2" diameter disks which have been polished flat and cathodically vacuum etched on one side. The specimens are never exposed to air after etching but are handled in an inert atmosphere and stored in a vacuum. Temperature monitoring of the in-pile couples is accomplished by means of a disk of uranium which contains small holes into which are placed sharp-cornered fragments of near-eutectic alloys. The maximum temperature reached should be indicated by a rounding of the sharp corners of all monitors that melt.

A capsule for in-pile diffusion studies, consisting of a thick-walled Zircaloy-2 cup ironed onto the diffusion couples, has been designed for U/Al and U/AlSi couples at 200-250 C (392-482 F) and U/Zr couples at 350-450 C (662-842 F). Fluxes ranging from 4×10^{13} to 7×10^{13} nv are required to produce these maximum temperatures at the diffusion interfaces. Three of the capsules containing two diffusion couples each have been prepared and sent to the MTR and are now being irradiated for a period of one cycle (approx. 18 days). The requested fluxes which will achieve the desired temperature at the diffusion interfaces are listed in Table 3.7.

TABLE 3.7

TEMPERATURES CORRESPONDING TO FLUXES REQUESTED FOR MTR IRRADIATION
OF HAPO THICK-WALL DIFFUSION CAPSULE

<u>Title of Capsule</u>	<u>Types of Couples</u>	<u>Maximum Temperature At Diffusion Interfaces</u>	<u>Requested Flux</u>
GEH-3-9A	U/Al-U/Al	200 C (392 F)	4×10^{13} nv
GEH-3-9B	U/Al-U/Al	250 C (482 F)	5×10^{13} nv
GEH-3-9C	U/AlSi-U/AlSi	200 C (392 F)	4×10^{13} nv

Laboratory experiments have continued as a basis for interpretation of the in-pile diffusion studies. In these experiments diffusion couples identical to those used for the in-pile studies are prepared as previously described, clamped together in

a purified inert atmosphere, and annealed in a vacuum. The clamp is tightened only enough to hold the couples together and does not cause visible deformation of either metal. The U/Al couples are broken apart after the anneal, and since the fracture occurs at the diffusion zone/Al interface, the maximum penetration of the uranium into the aluminum is measured directly with a depth measuring microscope. Diffusion in U/Al couples is not uniform at low temperatures but occurs at isolated points even though the couples are carefully cleaned and handled. The results listed in Table 3.8 illustrate the effect of specimen preparation and handling on the observed diffusion rate and indicate that cathodic vacuum etching is a superior cleaning method.

TABLE 3.8

A COMPARISON OF THE EFFECT OF VARIOUS METHODS OF CLEANING AND HANDLING U/Al DIFFUSION COUPLES BEFORE VACUUM ANNEALING AT 390 C (734 F) FOR 115 HR

Experiment	Method of Cleaning U	Method of Cleaning Al	Atmosphere Used During Handling	Maximum Penetration of U into Al (mils)	Remarks
A	50% HNO ₃	20% NaOH	Air	11	Approx. 10 minutes in air before vacuum anneal
B	50% HNO ₃	20% NaOH	Purified argon	26	One point 26 mils into Al -- all other points approach 11 mils
C	Cathodic vacuum etching	Cathodic vacuum etching	Purified argon	27	Many points 27 mils into Al

Several U/Al couples have been annealed at 200 C (392 F), 250 C (482 F), and 390 C (734 F). The maximum resulting penetration of the uranium into the aluminum for each couple is shown in Table 3.9. The curves in Figure 3.26 compare the maximum

TABLE 3.9

MAXIMUM DIFFUSION OF URANIUM INTO ALUMINUM AFTER VACUUM ANNEALING AT SEVERAL TEMPERATURES

Couple No.	Time(hr)	Temp., C	Maximum Penetration $x = \sqrt{Kt}$ (mils)	K (mils ² /hr)
9	119	390	27	6.13
10	119	390	26	6.13
11	287	250	12	5×10^{-1}
12	314	200	4.8	7.4×10^{-2}
13	330	200	3.0	2.7×10^{-2}
15	330	200	1.6	8×10^{-3}
17	317	200	4.3	5.8×10^{-2}
19	329	250	9.3	2.5×10^{-1}

FIGURE 3.26. Variation of the Maximum Low Temperature U/Al Penetration Coefficient with Temperature

low temperature U/Al diffusion rates as reported in BNL-38 with the results observed at Hanford in cathodically vacuum etched couples. A value of K taken from these curves can be used in the approximate diffusion equation $x^2 = K_{\text{eff}} t$ to determine the maximum diffusion which could occur after a time at a given temperature.

Four U/AlSi couples have been vacuum annealed at 200 C (392 F) and 250 C (482 F). The uranium and AlSi disks did not adhere to each other, and no transverse cross-sections of the diffusion zone have been made. Small projections about 0.2 to 0.7 mil high on the uranium disks and corresponding holes in the AlSi were found. The couples are now being analyzed by metallographic and x-ray diffraction methods in order to determine whether these projections are diffusion product and; if so, whether they represent the total extent of diffusion of the uranium into the AlSi.

Effects of Irradiation on the Crystal Structures of Zirconium and Molybdenum -
W. V. Cummings

There is nothing new to report this month.

Influence of Specimen Geometry on Line Broadening and Peak Intensity in Uranium -
W. V. Cummings

For a precise analysis of structure damage to crystalline materials, errors in the obtaining and recording of diffraction lines must be minimized. This is especially true in a few metals, notably uranium, which are characterized by inherent crystal structure imperfections which persist even after the material has received the best known annealing treatments. In uranium, these imperfections cause higher angle diffraction lines to be broadened approximately twice as much as those obtained from most other metals. This additional broadening in uranium produces difficulties in detecting and analyzing the crystalline damage, which apparently is not induced in gross amounts, by irradiation at normal Hanford pile operating temperatures or by cold working. The major errors that lead to a false broadening of diffraction lines and which are attributable to the x-ray diffraction apparatus are: 1) the size of the x-ray source, 2) the vertical divergence of the incident x-ray beam, 3) the finite width of the slits, 4) the diffraction of the x-rays from crystals that lie at an appreciable depth below the sample surface, and 5) the use of a flat specimen rather than one with a curvature that fits the focal circle dictated by the diffraction geometry. Of these errors, the first three are inherent in the design of the diffraction apparatus, the fourth does not present a serious problem in the study of uranium since the absorption of x-rays is negligible, so apparently, the only machine error that lends itself to possible correction is the replacement of the flat surface with one possessing a curvature equal to that of the focal circle. Unfortunately, present diffraction units are designed to use flat specimens and utilize a geometry which causes the focal circle to vary with the angle of diffraction. These conditions, although not ideal, are sufficient for the diffraction studies usually made on most materials. However, to achieve the best possible results in the detection of crystalline damage to uranium, the optimum diffraction conditions must be recognized and met. A small angular region can be chosen that contains a diffraction line, or lines, from which desired information can be obtained. In this way a single sample curvature can be used for a diffraction study in a restricted region using conventional spectrometers. Such an angular region was chosen for uranium that contains three diffraction lines very close together. The (221), (004), and (202) diffraction peaks all lie between 76.7 degrees and 77.2 degrees for copper K α radiation. Normally, these lines, with their α_2 components, are not resolved without a critical loss of intensity. A specimen was prepared so that one surface

was flat and the other surface had a curvature equal to that of the focal circle at this point. A comparison of the diffraction patterns obtained from the two sides, using identical machine constants, shows that a 17 percent increase in peak intensities was obtained by the use of the curved rather than the flat surface of the sample. The (152) peak at approximately 101.5 degrees was also scanned in a similar way. The gain in intensity is indicated in Figure 3.27. In addition, when the collimating and slit systems of the machine were adjusted so that equal peak intensities were obtained from the two sides, a large increase in resolution was realized.

The results of this experiment point out the possibility of applying this technique to the study of small amounts of radiation damage to uranium. The increase in diffracted intensity and resolution could be especially helpful where special modifications of equipment, such as a double diffracting technique, result in a loss of intensity that is sufficient to cause observation difficulties at the higher angles of diffraction. The flat sample effect still is of sufficient magnitude at these high angles to be of considerable importance. The preparation of the curved surfaces from material of high radioactivity will admittedly create a major problem. This difficulty can be alleviated to a great extent by forming the curvatures prior to irradiation.

Facilities

Elevated Temperature Tensile Testing Unit for Irradiated Materials - R. E. Hueschen and D. C. Kaulitz

The elevated temperature tensile unit for testing irradiated materials has been assembled and tested in the Radiometallurgy Laboratory. Test runs have been made at room temperature and at elevated temperature and have been reported previously⁽¹³⁾ and in another section of this report. Details of various components of the apparatus will now be described.

The optical unit has been described in a previous report⁽¹⁴⁾; a photograph and an optical diagram of the unit are shown in Figures 3.28 and 3.29. In Figure 3.29 Items 1 through 6 are the viewing microscope, folded telescope, mercury arc lamp, condensing lens, mirror, micrometer drum and vernier, respectively.

The vacuum furnace was assembled on the 60,000# Baldwin tensile machine as shown in Figure 3.30. A vacuum of 1×10^{-5} mm of mercury was maintained with the furnace at room temperature and 2×10^{-5} mm of Hg when the furnace was held at 700 C. The furnace was then calibrated with three thermocouples at four different temperatures, and the results are shown in Table 3.10.

FIGURE 3.27. A Comparison of Uranium Diffraction Peaks Obtained From Flat and Curved Sample Surfaces. The machine constants were identical for both curves.

FIG. 3.28
OPTICAL DIAGRAM

UNCLASSIFIED

- 3.43 -

HW-37979

FIG. 3.29

OPTICAL INSTRUMENT FOR MEASURING STRAIN ON TENSILE SPECIMENS

UNCLASSIFIED

FIG. 3.30

PHOTOGRAPH OF TENSILE FURNACE

DECLASSIFIED

TABLE 3.10

CALIBRATION OF VACUUM TENSILE FURNACE

Control Setting, °F	Control Thermocouple*, °F	Thermocouple Temp., ** °C		
		1	2	3
572	630	312(594 F)	311(592 F)	312
670	720	359(678 F)	360(680 F)	360
870	930	472(882 F)	472	472
1295	1340	690(1274F)	689(1272F)	690

*Control thermocouple located at furnace winding.

**Thermocouple 1 located on top grip.

Thermocouple 2 located 1/4 inch below bottom gage mark of specimen.

Thermocouple 3 " " " above top " " " "

The details of the vacuum furnace are shown in Figure 3.31. The furnace is mounted from the bracket (2) attached to the column (1) of the tensile machine. The entire furnace is lowered by this bracket with a rack and pinion which is operated from outside the cell. This vertical movement of the furnace, to permit loading and unloading of the specimen, is guided by the ground Thomson rods and the ball bushings (3). The pull rods (4 and 14) are made of 310 stainless steel to which the grips (1), made of Haynes Multimet, are attached. All of the remaining metal parts are made of 347 stainless steel. The three thermocouples are brought through a vacuum seal at the top of the furnace and an air cylinder (5) provides the pressure for the seal. Bellows (6 and 12) are located above and below the furnace to insure axial loading.

After a specimen is loaded into the grips, the furnace is raised approximately 10-1/2 inches and three air cylinders (7) are activated to effect the "O" ring seal by means of tapered jaws which force the two flanges together. The furnace (8) is sixteen inches long, and the furnace was wound specifically for this application. The optical windows (11) necessitated special attention in the design of the furnace windings (9). A long bellows is attached to the vacuum flange (13) to permit the vertical movement of the furnace. The bellows extends through the lead brick cell to the vacuum pumping system.

The platinum strips are spot-welded to the irradiated specimens using the fixture shown in Figure 3.32. After the platinum is polished, the gage marks are made using the Bierbaum Microcharacter apparatus. The strips are then cut and bent to exact shape to fit the fixture. The strips are placed into the fixture, and then the fixture and strips are put into the "hot" cell. Two holes, one inch apart, are drilled into the fixture normal to the center line of the specimen. These holes are drilled to close tolerances with regard to the bottom electrode so that the fixture assembly is centered and held in position. The two platinum gages are then spot-welded to the irradiated specimens, one strip at a time. After the taper pin which holds the specimen in position is removed, the specimen is ready to be placed in the special grips for testing. These grips are shown in Figure 3.33. Grips with spherical seats were fabricated to insure axial loading of the specimen.

FIG. 3.31

CROSS SECTION TENSILE FURNACE

FIGURE 3.32. Fixture for Remote Spot-Welding of Platinum Gages to Irradiated Specimen.

FIGURE 3.33. Tensile Specimen Grips

DECLASSIFIEDReferences

- (1) Martin, A.B. and Mills, M.M., The Application of Particle Accelerators to the Study of Radiation Damage, NAA-SR-56 (SECRET).
- (2) Dienes, C.J., Annealing of Cold Work and Radiation Damage in Au-Cu and in Cu, TID-5021, p. 72 (SECRET).
- (3) Katz, L. and Penfold, A.S., Rev. Mod. Physics, 24, 28-44 (1952).
- (4) Bhabha, H.J., Proc. Roy. Soc., 164, 257 (1937).
- (5) Mott, N.F., Proc. Roy. Soc., A124, 426 (1929); Mott, N.F., Proc. Roy. Soc., A135, 429 (1932).
- (6) McKinley, W.A., Jr., and Feshbach, H., Phys. Rev., 74, 1759-62 (1948).
- (7) Heitler, W., The Quantum Theory of Radiation, Oxford University Press, London, 1936, p. 219.
- (8) Dexter, D.L., Phys. Rev., 87, 768-77 (1952).
- (9) Metallurgy Research Sub-Section Quarterly Progress Report, October, November, December, 1954, HW-34455 (SECRET).
- (10) Evans, T.W., GEH-3-5, Uranium Tensile Specimens, 12/17/54, HW-34186 (SECRET).
- (11) Evans, T.W., GEH-3-11, Uranium Tensile Specimens II, 3/15/55, HW-35766 (SECRET).
- (12) Progress Report For February 1955, EMI-985, p. 86 (SECRET).
- (13) Metallurgy Research Sub-Section Quarterly Progress Report, January, February, March, 1955, HW-36054 (SECRET).
- (14) Cadwell, J.J., Quarterly Progress Report - Metallurgy Unit - July, August, Sept., 1954, HW-33602 (SECRET).

DECLASSIFIED

FILE METALLURGY - M. J. SandersonInsulated Fuel Elements - J. E. Minor, O. K. Shupe, and P. W. Dickson

This study is designed to determine the effect of increased uranium temperature on the life of a Hanford slug, whether of solid or cored geometry. It is hoped that the increased ductility of the outer uranium surfaces may be beneficial and that the increased central uranium temperature may anneal out a portion of the radiation damage in the center of the slug.

For initial studies an anodized layer on the inside of the aluminum can will be used as the insulator. The method of anodizing and the canning technique have been described in previous reports of this series^(1, 2).

Cored uranium slugs manufactured for previous tests^(3,4) have contained a 3/8-inch axial hole counterbored at each end and plugged with uranium end plugs. Threaded and welded end plugs have been utilized. In several instances slugs containing these end plugs have cracked radially at the base of the end plug. It appears possible that this radial cracking might be eliminated by use of a straight 3/8-inch axial core containing no end plugs.

Two slugs were fabricated containing a 3/8-inch axial core without end plugs. The slugs were canned by the same technique previously used. The 0.2 inch zirconium disk at each end of the uranium prevented flow of aluminum into the axial core during closure of the aluminum can. The slugs were autoclaved, tested, and one slug shipped to the MTR for irradiation.

Notched, Cored, Unbonded Fuel Elements - P. W. Dickson, D. C. Kaulitz, and T. W. Evans

A group of longitudinally notched, cored slugs have been prepared for irradiation in the MTR Fuel Element Testing Facility. The uranium used was all obtained from one rod, furnished by O. W. Rathbun of the Fuel Technology Sub-Section, for which the roll pressure directions and the ultrasonic pattern orientation prior to beta heat treatment are known. Cored (3/8" diameter core) cylinders 3.6 inches in length were machined from the rod along with wafers for metallurgical study and for stiff end pieces for use in the cold-canning of the specimens. Five of the fifteen slugs were notched longitudinally by conventional milling methods using milling cutters whose teeth had a 45° included angle and a 0.010 inch radius. The notches were milled to a depth of 0.050 inch. Five of the slugs were left unnotched, and the remaining five were notched internally. This last operation was performed with a broach which was fabricated by sawing the teeth off of several milling cutters of the same geometry as described above. The teeth were silver-soldered in a keyway cut in a length of 3/8-inch drill rod. The teeth were set in so that each tooth made a cut 0.001" deeper than the preceding tooth. It was planned to make the internal notches 0.030" deep, but because of damage to the broach, several of the teeth had to be removed and the notches could be made only 0.018" deep. The greatest dependence of the stress intensification at a notch is on the radius at the base of the notch, rather than on the depth or the included angle. Thus, it is not felt that the experiment is prejudiced, especially since there is no quantitative evaluation at hand of the notch severity for the service conditions to which these specimens will be subjected. Comparison of the internal notch geometry with that of the external notches, as seen

(1) Superscripts refer to references which are listed at the end of this section.

at the ends of the slugs under moderate magnification, showed no difference that could be detected by eye. The notches were all fabricated at the same position relative to the ultrasonic pattern so that the preferential tendency for splitting is eliminated as a variable.

The specimens were canned in aluminum by conventional cold canning and closure techniques except that uranium rather than zirconium wafers were used at the ends of the principal uranium piece. Destructive examination of pieces canned by this method revealed that the aluminum flowed into the external notch during canning and, further, that although the uranium suffered the usual amount of upsetting during the closure operation there was no crack initiation in the notches nor did the notch geometry appear altered by the canning processes.

The specimens were machined to size, subjected to the usual fuel element inspection procedures including extended autoclaving, and then two of each type were shipped to the MTR. It has been requested that three of these specimens (one of each type) be irradiated to failure, with examination scheduled for every normal shutdown period of the MTR.

Combined Fuel-Target Element for U-233 Production - J. W. Goffard

A combined fuel-target element for the production of U-233 has been proposed. The element consists of a thorium core surrounded by an aluminum-uranium-235 alloy sleeve. The fabrication of experimental test pieces awaits the receipt of the aluminum-uranium-235 sleeves. Dummy pieces are being prepared using aluminum-uranium (normal) alloy sleeves and the cold-canning point closure technique.

Hanford Size Uranium-Magnesium Fuel Elements - J. W. Goffard and P. W. Dickson

Twenty-four uranium chip-magnesium matrix slugs have been fabricated for long term in-pile testing at HAP0. Half contain normal uranium chips and half enriched (1.75 percent 235) uranium chips. The matrix is magnesium - 1 percent silicon alloy, and the finished slugs are about 40 volume percent uranium metal. These slugs were cold-canned by the point-closure technique. The production test authorizing the loading and irradiation of these slugs to 3000 and 6000 MWD/T in a HAP0 production pile has been approved and issued. However, the loading has been delayed because of the results of a high temperature flow lab rupture test. In this test a canned matrix slug with a 0.025" hole drilled through the can wall begins to swell sufficiently at about 220 C to reduce the tube water flow. Three minutes later at about 240 C the water flow was completely blocked by the swollen matrix slug. Water stoppage at this rate in a HAP0 process tube would constitute a definite operational hazard. A second drilled matrix slug is to be run in a flow lab rupture test at about 120 C to simulate actual HAP0 operating conditions. In addition, an aluminum canned matrix slug of HAP0 geometry (enriched uranium - 1.75 percent 235) is to be run in the MTR facility at the earliest opportunity.

Thermal Stresses in Fuel Elements - K. R. Merckx

A general study of the effects of irradiation and thermal stresses on fuel element failure is a complex problem. Until a general method of solution can be developed which will consider radiation damage, stress relaxation, cycling, and plastic yielding, the effects of the various phenomena are being studied separately. Previously, two documents^(5,6) were written which described a method of determining the temperature and time dependent effects of relaxation on stresses and strains.

Though experiments on aluminum and steel indicated the assumed model⁽⁵⁾ would be sufficiently general, the analysis of creep tests conducted by BNL indicates the model does not hold for uranium over the desired temperature range of 100 C to 500 C. To evaluate the model of material behavior, the following relation between the plastic strain, E_p , and the time, t , of various creep tests was assumed:

$$E_p = Ct^b,$$

where C is a function of stress and temperature, and b is constant. The b for the 100 C and 250 C or the 400 C and 500 C creep tests is approximately a constant, but a definite change in b exists between the 250 C and 400 C creep tests. The model for material behavior must be extended to include the above effect before thermal stresses can be calculated by the proposed method⁽⁶⁾. This extension will be difficult because it must cover a transition period of material behavior where new mechanisms of slip or slip planes and directions are activated. But, the understanding of creep behavior in the 250 C to 400 C range may help explain the observations of dimensional stability under irradiation at 500 C.

In-Pile Testing of Mechanically Bonded Fuel Elements - C. L. Boyd and G. A. Last

Examination of six mechanically bonded slugs, irradiated to investigate (a) the in-pile corrosion characteristics of the point closure and (b) possible aluminum-uranium interaction under operating pile conditions, has been completed. The canning of the fuel elements involved in these studies is described in a previous quarterly report⁽¹⁾.

Visual examination revealed no unusual film conditions which would be indicative of non-uniform heat transfer conditions and no evidence of preferential attack in the closure area. Metallographic examination of the aluminum-uranium interface, near the center of the slug cap (region of highest temperature), revealed no evidence of aluminum-uranium diffusion. A report summarizing the results of the examination is being prepared.

A mechanically bonded, solid, natural uranium slug was charged in the MTR Fuel Element Testing Facility during the quarter. The slug is operating at 65 kw/ft heat generation of which an estimated 8 kw/ft is due to gamma heating. The slug has been measured twice during the course of irradiation, with the following results:

<u>Diameter After Indicated Exposure</u>			<u>Length After Indicated Exposure</u>	
<u>0 MWD/T</u>	<u>90 MWD/T</u>	<u>200 MWD/T</u>	<u>0 MWD/T</u>	<u>200 MWD/T</u>
	1.438	1.438		
1.441	1.440	1.438	4.656	4.644
	1.439	1.439		

The slug will be measured again after 650 MWD/T exposure and will be discharged at a goal exposure of 800 MWD/T.

In-Pile Testing of Unbonded Fuel Elements - J. E. Minor and D. L. Zimmerman

A large fraction of current in-pile slug failures in Hanford piles are split failures originating in the uranium core. When this type of failure occurs in a bonded fuel element, the failure of the can wall section over the cracked uranium follows. It is hoped that this situation can be improved by omitting the uranium-aluminum bond, leaving the aluminum free to move with respect to the core. In the event of core

failure the length of aluminum stressed will be greatly increased, perhaps to include the entire circumference of the jacket -- thus greatly reducing the probability of jacket failure. Unbonded slugs are being tested to determine their rupture resistance and to determine whether or not a uranium-aluminum bond is essential for heat transfer under Hanford conditions.

The following irradiations of solid unbonded slugs under PT-105-580 A⁽⁴⁾ have now been completed:

<u>No. of Pieces</u>	<u>Specific Power</u>	<u>Exposure (MWD/T)</u>
1	47 kw/ft (9.0 watts/gm)	280
1	9 kw/ft (1.7 watts/gm)	56
2	38 kw/ft (7.3 watts/gm)	490
4	42 kw/ft (7.9 watts/gm)	1015

These slugs, as viewed in the basin, showed no evidence of preferential corrosion at the closure or of non-uniform heat transfer. The four slugs irradiated to 1015 MWD/T are scheduled for further examination by Radiometallurgy.

During the previous quarter four unbonded cored slugs enriched to 1.75 percent U-235 and 36 unbonded cored natural uranium slugs were charged in one tube at C Pile. The test was terminated by rupture of one of the natural uranium slugs at a tube exposure of 280 MWD/T.

The ruptured slug from this tube has been closely examined in the Radiometallurgy facility. Figure 4.1 shows the slug as received from the pile area. The aluminum base of the can had separated from the can wall, and the uranium was split longitudinally. Note in this view that the can wall shows none of the bumping characteristic of uranium corrosion occurring underneath the aluminum can wall. Figure 4.2 shows one side of the uranium slug after removal of the can wall. Note that the uranium shows little evidence of corrosion.

Figure 4.3 shows the split surfaces and exposed axial core of the uranium. These surfaces, in contrast to the exterior uranium surface, show evidence of gross uranium corrosion. The counterbored seat which had contained the base uranium end plug has suffered severe corrosion. The extent of this corrosion can be estimated by comparing this area with the relatively smooth counterbore still visible at the right end of the slug.

These observations, coupled with the fact that the cold closure on the aluminum cap was intact and had not permitted water penetration, led to the conclusion that this slug failed by internal corrosion. Welded uranium end plugs in cored slugs sometimes give rise to difficulties in canning because of cracks in the weld or porosity in the end plug itself. These cracks or porosity lead to accumulation of pickle solution or AlSi in the core in the dip canning process. Apparently, in the case of this cold canned slug, pickle solution accumulated in the core and was not detected during inspection of the slug. This solution then gave rise to sufficient corrosion during autoclaving and subsequent pile operation to cause failure of the slug.

At the time the can wall was stripped from the irradiated slug, the can wall was found to be still unbonded except for a ring of aluminum just below the radius end of the slug. Conditions for bonding in this area probably arise during the canning process. Closure of this type of can is accomplished by failing a heavy can wall

DECLASSIFIED

HW-37979

FIGURE 4.1. Ruptured Unbonded Cored Slug.
Views of Two Halves of Uranium Core
After Separation of Split Uranium.

Mag. 1X

Photographs Unclassified

DECLASSIFIED

FIGURE 4.2. Ruptured Unbonded Cored Slug. Base End
of Can is Missing. Mag. 1X

FIGURE 4.3. Side View of Ruptured Slug with Can Wall
Removed. Mag. 1X

Photographs Unclassified

section in compression across the top of the slug. During this operation, flow of aluminum down the side of the slug occurs to some extent. In the area in question a band of scuffed-off aluminum remains on the uranium when the slug is decanned. Apparently the aluminum surface formed in this manner bonds to the aluminum can wall by thermal spike bonding during irradiation. Sectioning and metallographic examination of this bonded area showed no evidence of uranium-aluminum diffusion.

Two of the enriched slugs and two natural uranium slugs that were loaded adjacent to the ruptured slug are scheduled for examination by Radiometallurgy.

In-Pile Testing of Insulated Fuel Elements (GEH-4-3) - J. E. Minor and O. K. Shupe

Insulated fuel elements are being irradiated to study the performance characteristics of Hanford geometry fuel elements in which the uranium is maintained at elevated temperatures during irradiation. Fabrication of the test specimens has been discussed under fuel element fabrication.

Two insulated slugs were charged in the MTR Fuel Element Testing Facility on May 20. Both elements contain a 0.004-inch layer of Al_2O_3 on the inside of the can as an insulating layer. The uranium in each slug contains a 3/8-inch axial core. In one slug, ARM-17, the axial core has been plugged with welded uranium end plugs. In the other, MR-3, the axial core has been left open. Each fuel element has a 0.2-inch Zircaloy-2 disk at each end of the uranium to insulate the closure and base ends of the can.

Initial measurements of power level indicate that the slugs are operating at 65 kw/ft, including 57 kw/ft fission heating and 8 kw/ft gamma heating. Under these conditions the calculated uranium surface temperature is 380 C and the maximum uranium temperature 790 C. In these calculations no allowance has been made for the temperature drop between the Al_2O_3 surface and the surface of the uranium.

Arrangements have been made for the slugs to be periodically removed from the facility and measured and inspected. Initial results of these measurements are shown below. The measurements were made by J. B. Burnham, Jr., of the Phillips Petroleum Company. Measurements of a standard utilizing the same methods showed a reproducibility of ± 0.002 inch.

Slug No.	Diameter After Indicated Exposure			Length After Indicated Exposure	
	0 MWD/T	90 MWD/T	200 MWD/T	0 MWD/T	200 MWD/T
MR-3	1.440	1.454 (+0.014)	1.455 (+0.015)	4.831	4.863 (+0.032)
		1.475 (+0.017)	1.456 (+0.016)		
		1.475 (+0.017)	1.457 (+0.017)		
ARM-17	1.438	1.455 (+0.017)	1.455 (+0.017)	4.841	4.898 (+0.057)
		1.456 (+0.018)	1.455 (+0.017)		
		1.458 (+0.020)	1.456 (+0.018)		

It seems probable that the initial increase of about one percent in the length and diameter of the canned fuel elements arises from thermal expansion of the uranium. The absence of further expansion upon additional irradiation appears to bear out this conclusion.

The slugs have been recharged in the MTR and will be irradiated to about 650 MWD/T before they are measured again. Goal exposure presently planned for these slugs is 800 MWD/T.

MTR Fuel Element Testing Facility - T. W. Evans and D. C. Kaulitz

A testing facility for the irradiation in the MTR of experimental Hanford fuel elements under simulated Hanford reactor conditions has been described in previous quarterly reports of this series⁽⁷⁾. The fuel element specimens are irradiated in an aluminum block built to replace beryllium reflector piece B-3. Pressurized cooling water is supplied to the fuel elements in a closed system, and an extensive safety system has been provided so that a mechanical failure or failure of a specimen will not endanger the MTR or persons working around it. The testing program being carried out in this facility is designated "GEH-4" by the Phillips Petroleum Company.

Fabrication of the basket-type rechargeable B-block (see previous quarterly report) was completed during the quarter, and it was shipped to the MTR. Because the hot-waste-tank venting modifications had not been completed, it was decided to put the new block into operation as soon as possible. Accordingly, the next set of specimens to be irradiated were removed from the welded type of block in which they were awaiting irradiation and transferred to a basket.

The venting modification was completed in time for the new block to be charged during the May 9 shutdown. While flow testing the equipment prior to startup of the reactor, a water "hammer" developed and an abrupt water flow stoppage occurred. The cause of this trouble was not immediately obvious so the assembly was removed from the reactor and the next cycle of operation begun. It was subsequently found that the basket had collapsed in the heat-affected zone near a braze at the upper end of the basket. (The basket is made of 2S aluminum tubing, about 1/2 hard).

The assembly was successfully charged in the reactor during a non-routine but scheduled shutdown on May 17, and operation started on May 19 when the reactor began cycle 52b.

The water hammer and basket collapse problem has not yet been completely eliminated, but several steps have been or are being taken which should eliminate or minimize the difficulties. The back-pressure control valve had shown signs of wear and was found to be difficult to regulate. A different, better type of valve for this function has been obtained and will be installed soon. The check valves in the "Titeflex" quick-seal couplers have been removed, and the couplers have been effectively removed from the system by wiring them together in such a way that they cannot easily become uncoupled. New baskets have been fabricated without the upper spacer lugs, so that the braze is eliminated, and thus the heat affected zone is no longer present. The strength of the basket is thereby increased. If it should become necessary to do so, the use of 1/32-inch-wall thickness tubing will be abandoned in favor of heavier walled tubing.

The power generation in the GEH-4-3 group of specimens (two insulated, cored slugs and a mechanically bonded cold-closed solid slug which are described elsewhere in this report) is in the range 60 to 65 kw/ft as determined from the water flow rate and temperature rise. The gamma heating is estimated at 8 kw/ft. The operation has proceeded smoothly from May 20, except for an accident which made it necessary to remove the B-block for cycle 53b. During the shutdown preceding this cycle, the upper grid assembly accidentally struck part of the in-pile equipment causing minor damage which was corrected several days later at the end of cycle 53.

The fuel element specimens were removed from the reactor after both cycle 52 and cycle 53 for examination and dimensional measurements, the results of which are given elsewhere in this report. The rechargeability feature of the new B-block

design was found to work quite well considering the handling tools that were available and the fact that the baskets were damaged as described above whenever the water hammer or flow stoppage occurred. Tools more appropriate for handling the baskets in the MTR canal are being made.

The irradiation of the two insulated, cored slugs and the mechanically bonded solid slug is scheduled to continue without further examination for three more reactor cycles. At this time the mechanically bonded slug will be replaced by a uranium-magnesium matrix slug canned in aluminum and the irradiation continued for one more cycle. At this time the insulated cored slugs will be at about 800 MWD/T. If an insulated slug fails, the irradiation will be discontinued and the next experiment (the notched, cored, unbonded slug experiment described elsewhere in this report) started. If the mechanically bonded slug fails, it will be replaced by the uranium-magnesium slug and the irradiation continued for a total of four more cycles as planned.

The first group of slugs to be irradiated in the MTR Fuel Element Testing Facility included two lead dip canned four-inch slugs, one of which was cored, and a nickel-plated hot-press canned four-inch solid slug. The irradiation was carried out uneventfully to an exposure of approximately 700 MWD/T at a power level of 55-60 kw/ft. The irradiation period extended from August 1 to December 6, 1955. The specimens have been returned to Hanford, the hot-press canned piece being turned over to the Fuel Assembly Unit and the lead dip canned pieces delivered to the Radiometallurgy Unit for examination. The principal goal of the examination will be to determine the magnitude of the flux asymmetry that very likely existed during the irradiation.

Three lead dip canned solid slugs, two of which had longitudinal V-notches milled in the surface of the uranium, were irradiated in the MTR Fuel Element Testing Facility from December 10 to December 23, 1955, at a power level of 80-82 kw/ft. The irradiation was terminated when a failure occurred after an exposure of 125-150 MWD/T. The slugs were found to be stuck in the welded B-block which was in use at the time. By use of the power hacksaw in the MTR canal, the block was reduced to a section some 22 inches long containing the three four-inch slugs and an aluminum dummy, which has been delivered to the Radiometallurgy Building for disassembly and examination. The examination will be directed toward determining the nature of the failure that occurred.

A formal report describing the basket B-block is being written.

High Temperature Irradiation of Uranium Test Specimens - C. L. Boyd and J. E. Minor

An irradiation of straight and tapered powder metallurgy uranium rods has been conducted to determine the accuracy of heat transfer calculations and the effect of in-pile transformation from the alpha and beta phase on the centers of uranium fuel elements. Photographs and drawings of the assemblies and a description of the post-irradiation appearance of the rods were published in preceding reports of this series(1,2,8).

One tapered, cylindrical specimen, which had operated at a calculated temperature of 50 C at the base to 900 C at the rod tip, was lapped and hardness measurements obtained. The results of the longitudinal hardness survey are shown in Figure 4.4. Hardness values decreased uniformly from approximately 99 Rockwell G scale at the base (50 C) end to 93, approximately 0.5-0.6 inch from the rod tip. The calculated temperature 0.5-0.6 inch from the rod tip was approximately 780-810 C. A more

FIGURE 4.4. Variation of Hardness with Calculated Irradiation Temperature

rapid decrease in hardness, 93-88 Rockwell G scale, was observed over the remaining portion of the rod at the tip end. Pre-irradiation hardness of the uranium rod was 82 Rockwell G scale.

An attempt to delineate the structure of the rods by macroetching to show the phase transformation regions will be made as soon as cathodic etching equipment is available for the etching of larger samples. This will determine the accuracy of heat transfer calculations and correlate hardness changes with phase changes.

In-Pile Testing of the Uranium-Magnesium Matrix Fuel Material - M. D. Freshley and W. S. Kelly

The properties of a high burnup fuel material which consists of discrete uranium particles embedded in a magnesium or magnesium-silicon alloy matrix is being investigated for suitability as a reactor fuel material. This fuel material, which has been described in previous issues of the "Quarterly Progress Report - Metallurgy Unit," has properties that are desirable for successful reactor operation. The chief advantages are increased thermal conductivity, dimensional stability, and improved resistance to radiation damage.

Ten small specimens of this uranium-magnesium fuel material have been irradiated to various exposures in the MTR, and two more are still in the reactor. These samples are 0.40 inch in diameter by 1.5 inches long and are canned in Zircaloy-2 capsules. The uranium that was used for these specimens is in the form of chips which pack about 50 volume percent uranium. Uranium chips were used because spheres were not available at the time. The chips are not intended to duplicate the behavior of spheres under irradiation, but only to give an indication of the effect of irradiation on this material. Six samples contain a matrix of pure magnesium, and the other six contain an alloy matrix of magnesium - 1.4 weight percent silicon. Silicon was added to investigate the effect of a compound layer between the uranium and the matrix material which forms a bond at the interface. Four capsules, two of each matrix material, have been irradiated to 1000 MWD/T, and four have been irradiated to 5000 MWD/T. One specimen containing each matrix material has been irradiated to 10,000 MWD/T, and the two remaining capsules are currently being irradiated to 20,000 MWD/T. The exposure level of the latter two capsules is estimated to be about 16,000 MWD/T at the present time.

The examination of the capsules which were irradiated to 1000 and 5000 MWD/T indicates that the material is dimensionally stable after exposure to this level(2,7).

Examination of the two capsules which were exposed to 10,000 MWD/T has been completed by the Radiometallurgy Unit. Figure 4.5 shows these 10,000 MWD/T capsules. Visual examination of the capsules showed no dimensional changes in the samples due to irradiation and optical micrometer measurements made on the samples after decanning indicates that the material is dimensionally stable under irradiation with little or no distortion occurring after being irradiated to an exposure of about 10,000 MWD/T.

It was difficult to decan the sample that contained the pure magnesium matrix, and it was necessary to cool the capsule in liquid nitrogen before it could be removed. This sample fell in two pieces as it was removed from the can. The uranium and magnesium surfaces of the fracture appeared dark which would indicate that the sample had been broken for some time which was possibly due to a flaw in the original specimen. Figure 4.6 shows the fracture surface. About 1/2 inch was broken from the sample which made it too short for bend testing.

SECRET

DECLASSIFIED

FIGURE 4.5. Zircaloy Clad Matrix Capsules.
10,000 MWD/T Exposure.
Mag. about 0.75X

FIGURE 4.6. Fracture Surface of
the Matrix Specimen that Broke
as it was Removed from the Can.
10,000 MWD/T Exposure. Mag. 7X

FIGURE 4.7. Fracture Surface
of the Matrix Bend Test
Specimen. 10,000 MWD/T
Exposure. Mag. 10X

Photographs Unclassified

SECRET

The sample that contained a matrix of magnesium - 1.4 weight percent silicon was easily removed from the capsule and was successfully bend tested. This sample was bend tested by center loading over a one-inch gage length. The maximum load at failure was 1110 pounds, and the maximum deflection was 0.0075 inch. This curve superimposes the data obtained for the 5000 MWD/T exposure samples and follows, within experimental error, the data obtained for the 1000 MWD/T exposure samples. The fracture surfaces of the magnesium and the uranium for this sample was shiny, as is shown in Figure 4.7. The load-deflection curve is a straight line which indicates that the material is not yielding plastically as it did in the unirradiated condition. However, this effect was apparently saturated at 1000 MWD/T. Additional radiation damage to the uranium is not affecting the physical properties of the fuel material, and the property changes that did occur were caused only by the damage to the magnesium matrix material.

Two 1.00-inch diameter by 4.0-inch long matrix type fuel elements canned in Zircaloy-2 are being irradiated in the MTR(7). These slugs have accumulated an exposure of about 1300 MWD/T at the present time. Their goal exposure is 5000 MWD/T.

Thorium-Uranium and Thorium-Uranium-Zirconium Alloys - J. W. Goffard and R. H. Todd

The study of thorium rich uranium and thorium rich uranium-zirconium alloys is continuing. These alloys are being considered as high burnup fuel materials.

Metallographic studies of thorium-2 to 5 weight percent uranium alloys have indicated a solid solubility of uranium in thorium as shown in Figure 4.8. The figure does not necessarily apply to the true binary thorium-uranium system because of the large amount of impurities known to be present in the thorium metal and observed in the prepared alloys. Figure 4.9 shows the structures obtained upon soaking as-cast 2 and 5 w/o uranium alloys at 800 C. Samples of the 2 to 5 weight percent alloys are being autoradiographed in the HAPO test pile per the technique described previously(7).

A wrought three-eighths-inch rod of thorium - 2 to 2-1/2 wt % U-235 is being prepared by Nuclear Metals, Inc., Cambridge, Mass., for use by Metallurgy Research in the MTR experimental test program. Tentative plans are for fuel pins 1-1/2" x 3/8" of this material to be canned in zirconium with 1/8" walls and 1/4" end caps and exposed to a flux of 1.5×10^{14} in the MTR. Preliminary physics calculations by H. Neumann, Theoretical Physics, indicate that the approximately 0.4" diameter fuel pins will have a blackness of 0.45 and will produce a total flux depression of 20 percent. These pins will generate 18.4 kw/in³. K. R. Merckx, Metallurgy Research, has calculated the following temperature drops and gradients for the MTR test capsules:

Δ T	thorium	151 C
Δ T	zirconium	455 C
Δ T	bond	78 C
Δ T	H ₂ O film	63 C

Total gradient 747 C

Ternary alloys of thorium - 2 w/o uranium and zirconium have been prepared and examined to determine if the solubility of zirconium in thorium will decrease the solubility of uranium in the thorium. Metallographic evidence to date does not indicate a decrease in uranium solubility. Various samples of these alloys are also being autoradiographed in the HAPO test pile for further metallographic study.

FIGURE 4.8. Thorium rich end of the Thorium-Uranium System as Determined Metallographically.

Photographs Unclassified

Mag. 750X

Uranium Eutectic, 0.5% HF in Water and Air Etch.
Uranium Soaked at 800 C. Dark Phase in Thorium-
Bottom - Photomicrograph of Thorium - 5 w/o
of Impurities.

Uranium Soaked at 800 C. Note Large Amount
FIGURE 4.9. Top - Photomicrograph of Thorium - 2 w/o

UNCLASSIFIED

Samples of the ternary alloys have been dissolved for chemical analysis with great difficulty. Since this suggests a high corrosion resistance of these alloys, samples have been submitted for water autoclave tests with the following results:

<u>Sample</u>	<u>Initial Wt.</u>	<u>Final Wt.</u>	<u>% Loss</u>	<u>Test Conditions and Time</u>
<u>Thorium</u>				
Alloy base HAPO Th slugs	32.7 g	0 g	100	Distilled H ₂ O at 240 C, about 4 hrs.
Ames as-cast billet	16.8	5.0	70	Distilled H ₂ O at 240 C, 16 hrs.
<u>Ternaries</u>				
Th-2 w/o U - 6 w/o Zr	58.7	0	100	Distilled H ₂ O at 240 C, about 6 hrs.
Th-2 w/o U - 10 w/o Zr	99.4	92.0	7.5	Distilled H ₂ O at 240 C, 16 hrs.
Th-2 w/o U - 10 w/o Zr	92.0	0	100	Distilled H ₂ O at 340 C, 9 hrs.

Ternaries varying in Zr content from 4 to 20 w/o zirconium have been prepared and are being hot rolled into rods. These will be used for heat treating, corrosion and metallographic studies.

Uranium Oxide Fuel Material - W. E. Roake

A series of Zircaloy capsules containing uranium oxide for irradiation in the MTR has been described in previous reports of this series. These capsules contain UO₂ of four types: 0.67 percent U-235 in packing densities of 5.5 ± 0.01 and 7.2 ± 0.2 gm cm⁻³; 1.34 percent U-235 in packing densities of 5.6 ± 0.05 and 7.4 ± 0.2 gm cm⁻³. During the quarter, groups of four containing one of each type capsule have been exposed at the requested fluxes of 4.5×10^{13} and 9×10^{13} for one and two reactor cycles. Scheduled final discharge was June 20, 1955. The capsules are expected to be received at HAPO in the near future for examination. Internal and external dimensional changes will be measured, and the pressures and compositions of internal gases will be determined.

It is generally accepted that a high concentration of defects in a crystal lattice promotes a high rate of diffusion and material transport. One method of causing this situation is to introduce into a lattice an impurity ion of different valence so that lattice vacancies result. This technique has been shown valuable in experiments by the personnel of the ORNL Ceramics Laboratory in which the addition of calcium oxide to a thorium oxide green compact resulted in a remarkable increase in densification rate during sintering. The ionic radius of Ca⁺² (0.99 Å) is very near that of Th⁺⁴ (1.02 Å) and is able to fit into the lattice with little distortion. This serves to produce an oxygen vacancy for every calcium atom thus assimilated. The addition of calcium atoms to a U⁺⁴ (0.97 Å) oxide lattice to provide an oxide deficient structure is made somewhat difficult by the nature of uranium oxide. Uranium oxide of compositions between UO_{1.75} and UO_{2.30} crystallizes in a face-

centered cubic structure. $\text{UO}_{2.0}$ is impossible to maintain in air since the lattice chemisorbs oxygen to form higher oxides. However, $\text{UO}_{2.0}$ (prepared by hydrogen reduction of UO_3 at 1000 C and stored under argon) was mixed with ten mole percent (1.65 wt %) CaH_2 and pressed in a 0.5" diameter steel die at 100,000 psi. A similar green compact was made of unadulterated $\text{UO}_{2.0}$. The two compacts were fired simultaneously for two hours at 1800 ± 100 C in vacuo. It was intended that the hydrogen resulting from thermal decomposition of CaH_2 would maintain a reducing atmosphere throughout the green compact and that the finely divided calcium metal similarly produced would reduce adjacent UO_2 to form a metal rich defect structure. The densities attained were 7.97 gm cm^{-3} for the UO_2 and 8.64 gm cm^{-3} for the U-Ca oxide. These densities, while not the ultimate attainable, do indicate the increased rate of densification of UO_2 , under comparable conditions, obtained by doping with small amounts of calcium.

An UO_2 piece isostatically pressed and sintered to 86 percent theoretical density by the ORNL Ceramics Laboratory has been clad in an intentionally defected Zircaloy can and submitted for exposure in the Elmo-4 recirculating loop. The oxide piece, 1.350" diameter and 4.00" long, is contained in a 0.020" thick jacket having 0.220" thick end caps fusion welded to the tube. The intentional defect is a 0.025" diameter hole drilled through the can wall at a position 1.25" from the end of the oxide piece. The oxide piece is slip fit into the Zircaloy jacket with 0.005" clearance. The assembly will be subjected to recirculating deionized water at a temperature of 280 C. Unless an excess of dissolved oxygen enters the recirculating water, the oxide assembly is expected to be stable since it is reported that no additives were used in the UO_2 pressing operation. The oxide has not been examined for intergranular carbides which might cause destructive corrosion in water. At the date of this writing the assembly has been subjected to 260 C water for 54 hours with no apparent effect on the fuel element.

Fission Product Studies - T. W. Evans

To provide a basis for the planning of an experimental program aimed at the determination of the effects of the fission products on the performance of high temperature, high exposure fuel elements, a study of the fission product chains and yields has been undertaken. The present aim of the study is to develop expressions which will permit calculation of the concentrations of the individual fission products as a function of exposure for a wide range of operating conditions. The study is being made from the metallurgical point of view in contrast to the approach taken in similar studies where the aim has been either to estimate reactivity changes with fission product buildup or to determine activity levels for radiochemical purposes. Various aspects of fuel element design, as may be affected by high fission product concentrations or fission product mobility, are being explored. Very little work has been done on this study this quarter because of the demands of other programs on the writer's time.

Reaction of Fuel Materials with High Temperature Water - J. E. Minor and D. C. Kaulitz

A large fraction of recent slug failures occurring in Hanford piles have been caused by thermal stress failure of the uranium core. Failures of this type provide opportunity for the cooling water to react with the hot core of the fuel element. The object of the present study is to evaluate the hazard arising from the reaction between uranium heated to 500-700 C and water at temperatures from 150-300 C. These temperatures cover the range from the upper limit of present operating conditions to the limit of conditions likely to exist in Hanford piles within the foreseeable future.

The reaction will be carried out in recirculating water loop constructed by the Byron Jackson Company of Los Angeles. This loop and its control panel were delivered on site during June. Two views of the loop proper showing the principal operating components are shown in Figures 4.10 and 4.11. Operation of the loop has been discussed in previous quarterlies of this series^(1,8).

The high pressure gauges in the control panel were provided with threaded fittings which would not hold 2000 pounds gas pressure. These fittings have now been welded and the wiring of the control panel is being checked. As soon as electrical and plumbing connections to the loop have been completed, the loop will be hydrostatically tested and insulated for high temperature checks. The first reaction should be run in the loop early in the next quarter.

Reaction of High Temperature Uranium with Water - W. E. Roake

In the course of another investigation an uranium slug uniformly heated to 850 C was quenched by rapid immersion in water at 0 C. A small amount of hydrogen was produced which ignited an exposure to air. The burning cloud of hydrogen occupied a volume of about one liter for a period of about one second. A small amount of uranium oxide remained as a slurry. The surface of the slug appeared unchanged.

Grain growth in Uranium and Uranium-Rich Alloys - J. E. Minor and W. E. Roake

It has been previously reported⁽⁷⁾ that slow cooling of uranium from the beta phase results in large columnar grains oriented parallel to the path of heat flow.

Fast cooling experiments were carried out in which the nickel plated end of a bare uranium slug was soldered to a water cooled hollow copper box. The opposite end of the four-inch slug was heated by conduction through a silver foil from an inductively heated graphite susceptor. By simultaneously turning off the heater power and flooding the hollow copper heat sink, initial rates of cooling as high as 1000 C per minute were obtained in the uranium slug. The temperature distribution before cooling was approximately linear between 935 C at the hot end to 400 C at the cool end. Measured rates of cooling through transition temperatures were 85, 150, and 225 C per minute gamma to beta, and 85 C per minute beta to alpha. In those portions of the slug heated above the alpha-beta transition temperature, large columnar grains were formed oriented parallel to the direction of heat flow.

In an effort to observe a limiting cooling rate above which the large grain structure would not be developed, a bare uranium slug was uniformly heated to 850 C in an argon atmosphere and 1/4 inch of one end rapidly inserted in agitated ice water. Cooling rates through the phase transformations upwards of 400 C per minute were measured. Large oriented columnar grains were formed, beginning at the slug surface which was directly in contact with the quenching water.

It seems probable that under any conditions of reactor shutdown large columnar grains will be formed in uranium operating above the alpha-beta transitions temperature.

Yield and ultimate tensile strengths of uranium specimens stressed parallel to and normal to the long axis of the columnar grains were identical within the experimental limits. Dilatometer specimens have been cut from the columnar grains material in a slug cooled in this manner. The dilatation curve of this specimen will be compared with that of a specimen cut from the fine grained end of the slug.

HIGH PRESSURE LOOP - FRONT VIEW

FIGURE 4.10

FIGURE 4.11
HIGH PRESSURE LOOP — REAR VIEW

Uranium-rich alloy slugs have been subjected to oriented slow cooling from temperatures above 800 C. A 1.5 atomic percent silicon alloy slug showed no evidence of grain growth, although a line of demarcation between that material cooled from above 660 C and that cooled from below 660 C was observed on a macroetched surface. A 0.4 atomic percent chromium alloy slug exhibited columnar grain growth only in that portion of the slug which had been cooled from temperatures above 770 C.

Reduction of UO_2 with Calcium - W. B. Tolley

Experiments have continued on the reduction of uranium oxides with calcium for the production of uranium pellets. Interest in the oxide reduction stems from the development of the magnesium uranium fuel material which contains discrete uranium spheres in a magnesium matrix. Preparation of spherical uranium by bomb reduction techniques is one approach being investigated for the production of uranium for the matrix fuel element.

Studies on the precipitation of both $UCaF_6$ and $UNaF_5$ double salts from uranyl nitrate solutions were continued. Ferrous ion in the presence of fluoride ion will reduce uranyl ion to the four state, simultaneously precipitating uranium tetrafluoride or a double salt depending on the cation that has been added to the solution. The double salts are easily washed and dried under argon while uranium tetrafluoride is difficult to handle and must be dehydrated at elevated temperature in a hydrogen fluoride atmosphere. The average button yield from six reductions of the calcium double salt was 96.1 percent. Iron impurity in the metal averaged 0.61 percent. Methods of reducing the iron impurity in the double salts should be studied. The most promising attack appears to be washing the iron impurity from the precipitated salt. However, it is apparently impossible to accomplish this with water or dilute hydrofluoric acid washes. Complexing agents which have a strong affinity for ferric iron may offer a solution. Possibly the choice of another reducing agent for the reduction of uranyl to uranium four prior to precipitation will eliminate the iron contamination problem. The same problem exists with the uranium sodium fluoride double salt. Again, excellent reduction yields, over 96 percent, have been achieved while iron contamination is higher than can be tolerated. Either salt would offer a cheap simple means of recovering uranium metal from uranyl solutions. A formal report, HW-35815, on the preparation and reduction to metal of $UCaF_6$ has been written, and a final report is being prepared on the studies made on the uranium sodium double salt. Results of the precipitation and reduction studies on $UCaF_6$ are presented in Tables 4.1 and 4.2, while Figure 4.12 shows the shape and size of some of the metal buttons. The uranium recycle problem has been transferred to the Chemistry Research Sub-Section under Dr. W. H. Reas. Further development of this and related studies will henceforth appear in that quarterly report.

A number of UO_3 reductions on a thirty-gram scale have been run. Using stainless steel reduction bombs heated by induction, temperatures above the melting point of uranium could be attained. Charges were fired without booster (with a final slag composition of 40 percent CaO in $CaCl_2$) and soaked at temperatures of 1300-1400 C for periods of 40 minutes to several hours. Some improvement in the yield of +325 mesh uranium was realized in the high temperature soak. However coalescence into a button does not occur. Reductions of UO_2 on small scale appear to yield more +325 mesh uranium than the UO_3 reductions. This is true in the small charges which can be soaked at high temperatures for extended periods of time. In the UO_2 reductions the ratio of calcium oxide to uranium is reduced which apparently results in the slightly better results. However, in large charges, 300 grams or more, it becomes almost impossible to safely soak charges at temperatures above the melting

TABLE 4.1

RESULTS OF PRECIPITATIONS

Precipitation Conditions:

100 percent excess HF
 20 percent excess $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$
 30 grams uranium per run
 1 mole $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ per mole U

Run	HF Conc. Percent	UNH Molarity	Percent Waste Loss		Product Composition ^b	Actual Mole Ratio	
			Precipitation	Washing		Ca/U	F/U
1	48	0.3	1.5	0.23	$\text{UCaF}_6 \times 1.1 \text{ H}_2\text{O}$	0.92	6.21
2	48	0.5	2.6	0.94	$\text{UCaF}_6 \times 0.7 \text{ H}_2\text{O}$	0.95	5.90
3	9.5	1.48	3.0	0.43	$\text{UCaF}_6 \times 1.2 \text{ H}_2\text{O}$	0.99	6.02
4	11.5	0.9	0.64	0.59	$\text{UCaF}_6 \times 0.6 \text{ H}_2\text{O}$	1.05	6.07
5 ^a	9.1	0.9	3.4	0.11	$\text{UCaF}_6 \times 1.1 \text{ H}_2\text{O}$	0.93	6.10

a 150-gram batch of uranium.

b All compounds contain about one-half percent iron.

DECLASSIFIED

TABLE 4.2RESULTS OF REDUCTIONS

1.0 mole Ca-I₂ Booster/mole U
30 percent excess calcium, 20 grams U/reduction

<u>Run</u>	<u>Percent Yield</u>	<u>Density</u>	<u>Percent Iron</u>
1	99.0	18.32	0.75
2	98.8	18.27	0.78
3	91.4	17.38	0.41
4	99.2	18.29	0.47
5	92.6	--	0.70
6 ^a	95.3	18.19	0.66

- a Two twenty-gram reductions were made using salt prepared in run 5.

DECLASSIFIED

Uranium Buttons From UCaF_6

Actual Size

1 2 3 6

FIGURE 4.12. Rough Surface on Buttons 2 and 3 Formed by a Calcium Cap Which was Dissolved. Average Yield was 96.1 Percent.

Photograph Unclassified

point of uranium due to bomb material structural limits. With this in mind it seems that UO_3 will probably be the oxide of uranium to reduce on a large scale, since the heat of reaction with calcium or magnesium is considerably greater than that of UO_2 .

One 300-gram charge of UO_3 with a calcium chloride flux added to yield a slag of 48 percent calcium oxide in calcium chloride was fired. The charge was soaked four hours at 980 C and then was cooled and leached with ten percent acetic acid. A yield of 50 percent +325 mesh uranium was obtained. A previous run on a 300-gram scale with no soak after firing gave a yield of +325 mesh uranium of 22 percent.

The calcium chloride to calcium oxide ratio has been varied over a wide range. No real improvement in yield was noticed in reductions from slags with a final composition of 9.1 percent calcium oxide in calcium chloride. As long as the slag is molten and fluid during the reduction, the yield seems to be effected very slightly by additions of calcium chloride. Slag compositions of about 40-50 percent calcium oxide in calcium chloride are presently being employed in most reductions.

Several 30-gram reductions of UO_3 and UO_2 have been run with a seed of uranium metal fines added to the charge. A five to ten percent improvement in yield of +325 mesh uranium has been realized by this technique. In one UO_2 reduction with an addition of 20 percent - 200 +325 mesh metal fines and employing 0.5 mole of iodine-calcium booster per mole of uranium, a yield of 69 percent +325 mesh uranium was obtained. The seeding experiments indicate the possibility of developing an efficient process with only 70 to 80 percent yield of +325 mesh uranium spheres recoverable in each reduction. The 20 to 30 percent -325 mesh uranium would be added to the succeeding charge as seed material. Since the complete elimination of the -325 mesh uranium formed during bomb reduction of the oxides appears to be impossible, the cycling seed process may solve this problem. Only a few experiments with seeded charges have been run, and a true evaluation of such a reduction cannot be made as yet.

Equipment for scale-up to 3000 to 4000-gram reductions is being obtained. A 10,000-cycle, 50 kw generator is being set up for use and may be ready by the middle of July. It is hoped that the large scale reduction experiments will be under way by the first of August. A true evaluation of the reduction of uranium oxides with calcium to yield spherical shot useable in the matrix fuel element should be more readily attained from the scaled-up reduction.

Study of the Coalescence of Uranium Metal in the Reduction Process - W. R. DeHollander

Electrolytic reduction studies both above the melting point of uranium and below have been stopped pending the arrival of equipment and facilities. Meanwhile, an investigation into the role of CaO in preventing the coalescence of uranium metal has been started. The plan is to develop a reproducible method of melting uranium turnings into a button and then to demonstrate the effect of adding CaO to the melt. As a result of these experiments, a new technique believed to be patentable has been developed to recover metal from badly corroded turnings and scrap. Although not new, the method is a correlation of novel ideas into a method not obvious to the mere routinist. The philosophy of the technique is as follows:

1. All grease is removed by detergent and water and the turnings dried.
2. Metal is not pickled in any way.
3. Calcium metal is added to reduce the uranium oxide to uranium and form CaO .
4. BaCl_2 is added to dissolve the CaO which would otherwise prevent coalescence.
5. Inert argon atmosphere is maintained over the melt to protect melt and metal from oxidation.

A flux of BaCl_2 alone has not proven to be sufficiently capable of excluding oxygen from small metal pieces although it is somewhat satisfactory for large massive metal. Uranium metal heavily crusted with oxide can be successfully melted by this method at temperatures of $1300\text{ C} \pm 100\text{ C}$. The product metal is a clean, smooth, solid button. The purity of such metal has not yet been determined but should be proportional to the purity of the scrap used.

When identical melts were made with 25 mol percent of CaO already present in the BaCl_2 , almost complete lack of coalescence of metal was noticed. That coalescence which did occur could be attributed to the close physical contact of pieces of metal which were not adequately covered with oxide at the start. Also apparent was the fact that the uranium turnings, initially $1/8" \times 1/32" \times 1"$ or $2"$ were converted to very small pellets and powders which did not coalesce. The breaking up of the metal is at present attributed to an effect analogous to the breaking of a soap film, when a myriad of tiny drops is formed from a flat sheet under surface energy forces. The presence of undissolved CaO prevents the coalescence of these tiny droplets of metal.

It is hypothesized that during an oxide bomb reduction small aggregates of uranium metal are imprisoned within a CaO shell which effectively separates the uranium particles until solvent flux such as CaCl_2 or BaCl_2 can remove the CaO barrier layer. In a bomb the higher temperatures attained in the center of the charge would permit a greater solubility of CaO in the solvent flux. Hence, larger particles of metal could form. On the other hand, the layer of charge next to the crucible wall would attain a much lower ultimate temperature as a result of the bomb reaction and also would be less affected by stirring action within the melt. Hence, powders would be expected on the fringes. A cross-section of such a bomb would show large masses in the center and powders at the fringes. No effort has been made to observe this as yet. If no solvent action of flux on CaO was apparent, then particles would be expected whose maximum size was of the same order of magnitude as the grain size of the uranium oxide used as raw material. A mechanistic view of the reduction process has the oxygen atoms of the uranium oxide grain moving to an uranium oxide-calcium metal interface, leaving uranium metal behind. In subsequent stages of the continuing reaction, calcium metal adsorbs on the initial CaO layer, abstracting oxygen atoms to form new layers of CaO . Thus, the calcium oxide layer grows from the uranium oxide grain face outward, thus effectively isolating grains from each other. Unless these oxide barriers melt or are made fluid by the penetration of a solvent, coalescence of the resultant metal cannot occur. It appears therefore that the rate of the reaction and hence the peak temperature attained will be a function of the speed of migration of oxygen atoms through the uranium oxide structure. Better reductions may be expected from oxides having the greatest number of defects in structure and stoichiometry. Work is planned to further clarify this picture.

~~DECLASSIFIED~~References

- (1) Cadwell, J.J., Quarterly Progress Report - Metallurgy Unit - July, Aug., Sept., 1954. HW-33602, Oct. 29, 1954 (SECRET).
- (2) Cadwell, J.J., Quarterly Progress Report - Metallurgy Unit - Oct., Nov., Dec., 1954. HW-34455, Feb. 1, 1955 (SECRET).
- (3) Berberet, J.A. and Minor, J.E., Production Test No. 105-513-SI, Irradiation of Cored Enriched Uranium Slugs, HW-30445, Jan. 13, 1954 (SECRET).
- (4) Lang, L.W., Production Test 105-580-A, Irradiation of Unbonded Slugs with Point Closure, HW-32650, Aug. 4, 1954 (SECRET).
- (5) Merckx, K.R., A Stress, Strain, Strain-Rate, Temperature Relation Which is Readily Determined Experimentally, HW-32531, July 28, 1954 (UNCLASSIFIED).
- (6) Merckx, K.R., Time Relations for the Thermal Stresses in an Infinite Cylinder, HW-32562, July 30, 1954 (UNCLASSIFIED).
- (7) Metallurgy Research Sub-Section Quarterly Progress Report, Jan., Feb., March, 1955, HW-36054, May 16, 1955 (SECRET).
- (8) Cadwell, J.J., Quarterly Progress Report - Metallurgy Unit - Jan., Feb., March, 1954. HW-31799, May 10, 1954 (SECRET).

RADIOMETALLURGY - L. D. Turner**DECLASSIFIED**Low Exposure Failures - D. P. O'Keefe

In recent months a large number of slug failures have occurred after tube exposures of only 150-250 MWD/T have been reached. The majority of the ruptures have been of the "side failure" type in which the damage to the slug in the form of uranium corrosion is confined to a portion of the slug side and apparently results from failure of the jacket in or near the area of uranium corrosion product buildup.

The appearance of many of the recent failures has differed from that of previous side failures in that comparatively little swelling of the can and cavitation of the uranium occurred. In addition, many of the "low exposure" failures have shown evidence of uneven water flow in the pattern of film deposition on the slug surface. Failure No. 460 from Tube 0777-H, selected by Fuel Technology as representative of the low exposure failure as a type, was examined in detail. (Figure 5.1) Within the stream-lined area on the slug, intergranular corrosion of the aluminum was found to have occurred. The corrosive attack had proceeded half way through the can in spots near the ruptured area and had penetrated the can in the area of rupture. (Figure 5.2)

An unruptured slug from Tube 1587-H with a film pattern indicating a similar in-pile environment was examined and found to have suffered a similar corrosive attack in one area of the can. The only cause that could be found for the occurrence of this attack was overheating in one portion of the slug surface. The film pattern on both slugs suggested that slug cocking and consequent disruption of coolant flow caused the hot spots. Neither a material nor manufacturing defect could be found which would result in overheating in one area of the slug.

An item of interest, although it's probably not associated with the failure mechanism, was the presence of numerous micro cracks in the uranium of Failure 460. The cracks were both intergranular and transgranular, and more of them were found than have been seen in other irradiated uranium microstructures so far observed. No other defects were observed in the uranium.

Examination of Cored Uranium Slugs - D. P. O'Keefe

The examination of three cored, AlSi-bonded, natural uranium slugs which had been irradiated to a tube exposure of 1150 MWD/T was completed. In the pile test of this material, the irradiation had continued until a failure occurred. The failure and the adjacent upstream and downstream pieces were selected by Fuel Technology for examination. The failed slug had split longitudinally and had multiple cracks at each end of the slug. The uranium of the other two slugs had split longitudinally, but rupture of the jackets had not occurred. The longitudinal cracks in the slugs appeared to have started at the 3/8-inch diameter core. The uranium of the ruptured slug and the slug upstream of the rupture was examined metallographically for uranium quality and the effects of pile exposure on the uranium. The uranium quality appeared to be normal, and no evidence of beta phase heating was found in the uranium microstructure of either slug.

The longitudinal splits which had developed in the two unruptured slugs accompanied a deformation of the uranium which caused a diameter increase of 0.040 inch in one direction and a very slight decrease in the diameter perpendicular to this direction. The slug ends, containing the plugs welded in place, had undergone the minimum deformation. Failure of the one slug appeared to have resulted from the same deformation process which had cracked the unruptured slugs.

Mag. 0.6X

FIGURE 5.1. Failure No. 460, Discharged From Tube 0777-H on 5/3/55, 230 MWD/T Tube Exposure. The downstream end of the slug is on the left. The streamlined area on the slug was centered about 50° from the vertical as the slug was positioned in the tube.

50X

FIGURE 5.2. Cross-Section of Can Wall $1/4$ " From Area of Rupture. The depth of the attack in this small area was about half the can wall thickness.

Photographs Unclassified

Examination of Failed Unbonded and Mechanically Bonded Slugs with Point Closures -
D. L. Zimmerman

A recent production test^(1,2) to evaluate the in-pile performance of unbonded and mechanically bonded slugs canned by the point closure technique⁽³⁾ resulted in the failure of one of each type of slug. The mechanically bonded failure was discharged from Tube 2062-C after an exposure of 312 MWD/T, and the unbonded failure was discharged from Tube 2061-C after an exposure of 483 MWD/T. The results of the examination of these slugs are described below.

The mechanically bonded slug is shown in Figure 5.3. The swelling of the jacket resulted from the formation of uranium corrosion products. No swelling was observed between the rib marks. Four splits of various sizes were found in the swollen area of the jacket, and the largest of these may be seen near the center of the slug in Figure 5.3. The aluminum jacket at first appeared to have "necked down" before failure; however, examination of this area following cross-sectioning revealed that the reduction in thickness may have resulted from corrosion of the inside surface of the jacket in the vicinity of the crack. All of the smaller splits in the swollen area had the appearance of brittle failures.

A detailed examination of the outside surface of the jacket revealed no evidence of excessive corrosive attack. A small pit located in the center of the cap was attributed to mechanical deformation rather than corrosive attack. The cap end was removed from the slug and leak tested. The results of this test were negative.

The slug was sectioned transversely through the major jacket split, revealing one very large and two smaller radial cracks in the uranium core. These cracks are shown in Figure 5.4. The large crack was filled with uranium oxide indicating that its large size may have resulted in part from corrosive attack and the wedging action of the resulting corrosion product. The smaller cracks may also have resulted from the wedging action of the corrosion products. Metallography revealed no abnormalities in the macrostructure of this section. Examination of a second cross-section made 1-3/8 inches from the base end revealed some very small macro cracks in the center of the uranium core; however, no large cracks were observed.

The evidence observed to date would seem to indicate that failure of the slug originated in the uranium core rather than the jacket or cap. It appears that the large crack found in the core was sufficient to split the jacket, thereby allowing water to enter and cause swelling and further tearing of the jacket.

Because the point closure technique produces a jacket which is 3/4 hard, it is conceivable that the formation of a large crack in the core would produce sufficient expansion to cause the jacket to split with very little plastic deformation if it were bonded to the core. In the case of this slug, the mechanical keying produced by sizing the jacket onto an anodically roughened core may have provided the necessary degree of bonding.

The unbonded failure is shown in Figure 5.5. Severe swelling and tearing of the jacket at the cap end by the formation of uranium corrosion products can be seen. Both the cap and base were nearly detached from the slug by extensive circumferential tears in the jacket resulting from corrosion product formation. Appreciable plastic deformation of the aluminum was associated with the tear at the base, but very little was noted at the cap end.

(1) All footnotes are referenced at the end of this section.

DECLASSIFIED

-5.4-

HW-37979

FIGURE 5.3. Mechanically Bonded Point Closure Failure
From Tube 2062-C. Cap end is to the right.

FIGURE 5.4. Cross-Section Through the Longitudinal Jacket
Split of the Failure Shown in Figure 5.3.

Photographs Unclassified

Photographs Unclassified

FIGURE 5.6. Unruptured Unbonded Point Closure Slug
From Tube 2061-C. Note large "wrinkled"
area on surface of jacket.

FIGURE 5.5. Unbonded Point Closure Failure From
Tube 2061-C. Cap end is to the left.

Sectioning of the slug approximately 1-1/2 inches from the cap end revealed no macro cracks in the uranium core.

In addition to the above described ruptures, examination of two unruptured unbonded slugs from the same production test has been started. Both of these slugs are from Tube 2061-C (exposure 483 MWD/T). Both slugs exhibited "wrinkled" areas on the jacket surfaces as shown in Figure 5.6. The mechanism involved in the formation of these wrinkles is not definitely known; however, it appears that they were formed in the pile, since they were not observed during any of the pre-irradiation inspections. Both slugs showed outside diameter increases of 0.016 to 0.025 inch. Sectioning of one slug revealed pronounced macro cracks in the center of the uranium core which is probably responsible for the diameter increase of the jacket. This expansion may have been sufficient to produce the wrinkles in the jacket.

The Effects of Irradiation on the Properties of Deformed Non-Ruptured Slugs -
C. L. Boyd

An investigation on the effects of irradiation upon deformed non-ruptured uranium slugs has been initiated to provide background physical and mechanical property data for comparison with similar data obtained from ruptured slugs, preformed samples, and normal slugs.

Data on bend strength, hardness, and density were reported⁽⁴⁾ for an eight-inch, beta heat-treated, normally discharged slug, which has been selected to determine the feasibility of forming physical property test samples from whole slugs by abrasive cutting and grinding. The previously reported data are listed in Table 5.1 along with recently obtained electrical resistivity data.

Electrical resistivity measurements conducted on two samples (3/8" x 1/8" in cross-section) with center lines approximately 1/16-inch and 7/16-inch from the slug axis, gave values of 32.4 ± 0.2 and 31.7 ± 0.1 micro ohm - cm at 20 C, respectively. Measurements were made on a gage length located 2.4 to 2.9 inches from the cap end of the uranium. These values show increases in resistivity of 4.5 percent and 2 percent above an average of 31.0 ± 0.2 reported for non-irradiated beta heat-treated uranium samples oriented parallel to the slug axis. Values of the temperature coefficient of resistance showed a slight increase. Figure 5.7 shows the variation of resistivity with temperature for the two samples tested.

Metallographic studies of a wafer sectioned adjacent to the resistivity samples revealed no evidence of microcracks or large inclusions, which may have affected the resistivity values. Photomicrographs of the structure of this material, Figures 5.8 and 5.9, indicated an increase in the number of twins. Macro examination, Figure 5.10, of the uranium showed a larger grain size at the center of the slug and a maximum eccentricity of uranium with respect to the aluminum can. On one side of the fuel element the surface of the uranium core and the jacket are in contact.

The larger value of resistivity obtained for the centrally located sample contradicts the expected results. The greater annealing of irradiation induced cold work, larger grain size, and smaller concentration of fission products at the center of the slug should have resulted in a smaller value of resistivity. Three more samples, which were obtained at varying distances from the slug axis, remain to be tested before conclusions can be made concerning the indicated resistivity increase at the slug center.

TABLE 5.1

<u>Specimen</u>	<u>Distance From Slug Axis inches</u>	<u>Bend Strength psi</u>	<u>Modulus of Elasticity x 10⁶</u>	<u>Hardness R_H</u>	<u>Density gm/cc</u>	<u>Elec. Res. at 20 C Microhm-cm</u>	<u>Temp. Coeff. of Resistivity/°C</u>
Control		195,800	12.3	74-77	18.91-18.99**	31.0 ± 0.2*	0.00249 ± 0.00014*
D-1	7/16	124,160	17.4	93-94	18.866	31.7 ± 0.1	0.00251 ± 0.00014
D-3	1/16	110,670	18.7	91-92	18.860	32.4 ± 0.2	0.00254 ± 0.00011

*Values from PT-105-3N uranium per conversation with R. S. Kemper

**Reported values (5).

FIGURE 5.7

VARIATION OF RESISTIVITY OF 520 MWD/T
BETA HEAT-TREATED URANIUM WITH TEMPERATURE

Vacuum Cathodic Etch

Bright Field

250X

FIGURE 5.8. Structure of 520 MWD/T Uranium at the Edge of the Wafer Shown in Figure 5.10.

Vacuum Cathodic Etch

Bright Field

250X

FIGURE 5.9. Structure of 520 MWD/T Uranium at the Center of the Wafer Shown in Figure 5.10.

DECLASSIFIED

UNCLASSIFIED

-5.10-

HW-37979

Vacuum Cathodic Etch, 5X

FIGURE 5.10. Macrograph of 520 MWD/T Uranium Wafer. Note eccentricity of aluminum can wall with respect to the uranium core.

Photograph Unclassified

MTR Capsule Irradiation - Examination of Capsules GEH-3-5-C and GEH-3-5-D -
R. S. Kemper, W. S. Kelly, and E. C. Watts

Four Zircaloy-2 capsules containing preformed beta heat-treated uranium tensile specimens canned in NaK were irradiated in the Materials Testing Reactor to determine the effects of high exposure at elevated temperature on the tensile properties and microstructure of uranium. Two of the capsules, GEH-3-5-C and GEH-3-5-D, have been opened and the samples visually examined and photographed. The capsules were opened by twisting off the end while the capsule was immersed in butyl alcohol, after first cutting almost through the capsule wall with the parting lathe.

Both uranium tensile specimens were found to be warped (Figure 5.11). The zircaloy wire spacers which had been welded to one end of the samples prior to irradiation were broken off. These wires were apparently broken off in handling or shipping the samples rather than during irradiation.

The opening of these capsules marked the first time irradiated samples canned in NaK had been opened; however, the operation was conducted with no unexpected difficulties.

Impact Strength of Zircaloy-2 Irradiated in Circulating, Pressurized Water -
R. G. Wheeler and W. S. Kelly

The effect of irradiation on Zircaloy-2 precharged with 100 ppm hydrogen was studied by comparing the impact energy absorption of irradiated specimens with that of unirradiated specimens as a function of impact testing temperature. Hydrogen was charged into 18 cold worked impact specimens by heating them at 500 C for one hour in a vacuum apparatus, then allowing them to soak up hydrogen and homogenize for four hours at 600 C. The same annealing treatment was given to 18 identical specimens except that no hydrogen was added. On the basis of the in-pile test on these samples, it appears that Zircaloy-2 specimens containing less than 20 ppm hydrogen when exposed to 64 MWD/AT (1.9×10^{19} nvt) in recirculating water at 146 C (295 F) have Charpy "V" notch impact energies essentially the same as those measured in the control specimens exposed downstream of the active charge to 0 MWD/AT at 190 C (375 F). The irradiated specimens containing 100 ppm of hydrogen added before exposure broke at lower impact energies at the same temperatures than did identical control specimens exposed downstream out of the pile flux. Below an impact testing temperature of 175 C (347 F) the irradiated specimens containing 100 ppm broke at from 5 to 10 ft lbs and the unirradiated broke at 6 to 11 ft lbs. Transition occurred at about 230 C (446 F) in the irradiated and at about 200 C (392 F) in the unirradiated based on a criterion of one-half the sum of maximum and minimum impact energies. These data indicate that addition of hydrogen to Zircaloy-2 either accelerates the pickup of additional hydrogen during irradiation in pressurized water or the addition of the hydrogen makes Zircaloy-2 more susceptible to irradiation embrittlement.

Specimens cut from samples impact tested at room temperature and at the transition temperatures have been sent for radiochemical analysis to determine the hydrogen content of the samples after irradiation. This will resolve the question of additional hydrogen pickup during irradiation. The results of the impact tests are shown in Figure 5.12.

DECLASSIFIED

-5.12-

HW-37979

Sample GEH-3-5-D, Approx. 1X

Sample GEH-3-5-C, Approx. 1X

FIGURE 5.11. Tensile Specimens from GEH-3-5
After Removal from NaK-filled
Capsule.

Photographs Unclassified

FIGURE 5.12. Impact Energy to Fracture vs. Temperature for Zircaloy-2 Irradiated to 1.9×10^{19} nvt in Circulating Pressurized Water.

Examination of Zirconium and Zircaloy-2 Tube Sections Irradiated While Exposed to Pile Gases - PT-105-516-S1 - W. S. Kelly

A production test to obtain information concerning the changes in the properties of zirconium and Zircaloy-2 samples exposed to neutron flux and pile atmosphere at elevated temperatures is under way⁽⁶⁾. The samples from the second exposure (approximately 800 MWD/AT) were delivered to the Radiometallurgy Building for examination. The temperature of exposure was approximately 410 C⁽⁷⁾. The samples were removed from the graphite capsule in which they were irradiated and visually examined. The samples appeared unchanged by irradiation. After the visual examination was completed, the samples were weighed. Interpretation of weight gains are reported on page 3.23 by R. G. Wheeler.

Metallographic Cell - J. R. Morgan

Designing is in progress on various improvements to the metallographic cell in addition to introduction of replication and improvement on vacuum cathodic etching.

There are two principal guiding points in the design of all future equipment for the metallographic cell: (1) mount all possible equipment on cell plugs to be installed or removed independently of other equipment, and (2) arrange waste disposal equipment so that all highly radioactive waste can be collected and canned remotely for disposal via cask to avoid the personnel exposure and contamination hazards with which we are presently plagued.

New polishing equipment is being designed in keeping with this philosophy. Each unit will mount on the end of a plug and be capable of insertion or removal from the cell independent of other cell activities. Water in comparatively large amounts will replace oil as the lubricant in coarse polishing on gritcloth. Large particulate matter will be collected and placed in disposable cans inside the cell, while the effluent water will be piped directly to the hot waste system. These measures together with easily accessible cleanout provisions should virtually eliminate the liquid waste disposal problem. It has been decided that if the gritcloths themselves are still too radioactive for safe handling in cardboard boxes after use under these conditions, a coiler will be built on the end of a plug so they, too, can be canned in the cell for disposal via cask.

A macrophotographic unit soon to be built will give a continuous series of magnifications from about 1X to 25X. Integral with the unit is a sample positioner and illuminator which should make it possible to duplicate reasonably well the quality of macrographs obtained on standard non-remotized equipment. Optics of the unit will consist of a turret of three lenses, 32 mm, 72 mm, and 158 mm, mounted inside the cell and directly in front of a viewing plug. Lead shielding will protect lenses not actually in use from severe darkening by radiation. A bellows will be attached to the out-of-cell end of the viewing plug. Bench tests have indicated that stray light in the lead glass viewing plug will be one of the major problems. Light leaks too small to be troublesome in an ordinary bellows will scatter in the lead glass sufficiently to destroy quality.

The 15X and 60X objectives on the remote metallograph were changed after about 3-1/2 months' service in the cell. A rough check with an SEI exposure meter indicated the 15X objective was absorbing approximately 97 percent of the light in two passes while the 60X objective was absorbing more than 99-1/2 percent. Estimates indicate the 15X lens received some one or two million roentgen of radiation while

the 60X lens received perhaps as much as 25 million roentgen. It must be emphasized that these radiation values are little more than considered guesses arrived at by application of the inverse square law to approximate intensity readings obtained by holding the sample before a 1/2-inch diameter hole in a cell plug and taking a CP dosometer reading outside the cell.

Vacuum Cathodic Etching Procedures - G. R. Mallett

Vacuum cathodic etching of irradiated uranium samples is proving quite useful in our operations. A vacuum cathodic etch provides an etch that will give a good macro-photograph, good photomicrographs, and a surface clean enough to be replicated.

The etching conditions can be easily varied; therefore, different combinations of these conditions are being tested to see which conditions will give the best results. It has been determined that a 20-minute etch at 10 milliamps current, or 200 milliamp-minutes, provides the optimum degree of etch. Potentials of 3000, 2000, and 1500 volts have been tested. The best etch, so far, was obtained at 1500 volts. The argon pressure for all of the tests has been at 75-85 microns.

Further variations in the potential will be tested to see if 1500 volts do provide the best etch by vacuum cathodic means, using argon as the conductor.

Macroetching - G. R. Mallett

A chemical or electrochemical method for obtaining a macroetch on irradiated uranium is desired for quick and easy study of samples when no photomicrography is needed.

A method that will provide an etch capable of producing a good macrophotograph when photographed through a lead glass window is desired. Also, when a good etch is produced, some method must be used to retain the etch long enough for photographing and observation.

Several etching methods have been tested. A chemical method tested was with a hydrochloric acid etch, then cleaning in nitric acid, and rinsing in water. The electrochemical methods tested were a phosphoric-hydrochloric acid bath, a trichloroacetic-hydrochloric acid bath, a sulfuric-chromic acid bath, and an ammonium chloride-phosphoric acid bath. All of these baths, except the sulfuric-chromic acid bath were followed by a nitric acid cleaning, then a water rinse. The sulfuric-chromic acid bath was followed by a water rinse only.

The best etch was obtained with an electrochemical method using the phosphoric-hydrochloric acid bath. The bath contained 50 percent by volume of concentrated phosphoric acid and 10 percent by volume of concentrated hydrochloric acid. With the samples as an anode, an initial current density of 500 amps per square foot was applied for 30 seconds. This time can be extended if any polishing is desired. The sample is then etched at 50 amps per square foot for 10 minutes; more etching time can be used if needed. The black anodic layer, due to etching, is removed in 50 percent by volume of concentrated nitric acid; this requires a time of 30 seconds. The sample is then rinsed in water. The etch can be preserved for two or more hours by rinsing it in ethyl alcohol, then n-octyl alcohol. A representative etch of the type obtained is shown in Figure 5.13. The white dot is the contact point when etching.

Samples of 260, 600, and 1550 MWD/AT have been satisfactorily etched; therefore, no further etching studies will be made.

DECLASSIFIED

-5.16-

HW-37979

3X

FIGURE 5.13. A macrophotograph of a 600 MWD/T wafer etched in the phosphoric-hydrochloric acid bath, 500 amps/square foot for 30 seconds, then 50 amps/square foot for 10 minutes was applied. The etch was preserved for two hours by rinsing in ethyl alcohol, then n-octyl alcohol.

Photograph Unclassified

Replication of Irradiated Specimens for Optical and Electron Microscopy -
T. K. Bierlein, J. R. Morgan, and G. R. Mallett

Known correlations between changes in microstructure and physical properties of metals have resulted in the extensive use of metallography as a quick and economical tool for obtaining metallurgical data. Although extensive developmental work has been done to evolve techniques suitable for the metallography of irradiated materials such as uranium, certain aspects still require further improvement. Restrictions on equipment as well as personnel resulting from radiation and the short useful life of surface preparation are the two major technique problems currently under study.

Suitable replication of freshly etched surfaces of highly radioactive samples promises great improvements in both radiation and preservation problems. In addition, they offer various intrinsic advantages over direct specimen observation. They permit use of standard microscope equipment and normal exposure times as a result of their low radioactivity; they provide a permanent specimen record which is not subject to deterioration or storage problems; and they afford a convenient basis for conducting optical as well as high resolution, electron microscope studies.

Trial Faxfilm replicas of irradiated zirconium and uranium have been successfully applied, stripped, and manipulated with remotely operated Hanford-type slave manipulators. Contamination of these trial replicas was less than 50 mrad/hr at contact, and it was possible to conduct normal type optical and electron microscope examinations on the replicas. Since the preliminary results have substantially proved the utility of a replication process on radioactive materials, prototype manipulators and accessories are currently being designed to provide a more efficient replication system. Future major modifications of the metallographic cell will include a replication blister or chamber in which specimen and replica cleanliness can be maintained.

Figure 5.14 illustrates the optical results obtained on a zirconium specimen irradiated to 780 MWD/AT. The specimen was etched in an aqueous $\text{HF} + \text{HNO}_3$ solution and when photographed directly on a shielded metallograph yielded the micrograph listed as (a) in Figure 5.14; faxfilm replicas which were free of any contamination, although the zirconium specimen had an associated radioactivity of 5 Rad/hr/at contact, were obtained from this etched specimen, coated with aluminum, and photographed on a normal metallograph. (b) and (c) of Figure 5.14 illustrate the appearance of the replica.

Figure 5.15 illustrates the appearance of a cathodically vacuum etched specimen of uranium irradiated to 580 MWD/T as photographed with the remote metallograph, and Figures 5.16 and 5.17 illustrate the appearance of its aluminized Faxfilm replica as photographed with a normal metallograph.

Three metallographic specimens of uranium, each immersed in NaK and contained in a Zircaloy-2 capsule, are currently being irradiated in a flux of $3-4 \times 10^{13}$ nv for a period of about 36, 72, and 108 days, respectively. As the micrographs of Figure 5.18 illustrate, the specimens have been etched and marked such that the identical area may be re-examined after irradiation. The replica, an area of which is shown in Figure 5.18, should provide a simple and convenient means for comparing any portion of the original surface of the specimen with the corresponding area after irradiation. In order to conduct the high resolution, electron microscope studies on particular areas within such replicas, an area of the aluminized replica is first examined and photographed optically; the aluminum is then removed, and the base replica or a duplicate replica can be prepared for electron microscope examination and photography. In this manner, correlation of low magnification and high magnification micrographs should be possible.

(a) P.L. 260X

(b) B.F. 250X

(c) B.F. 750X

FIGURE 5.14. Photomicrographs of zirconium irradiated to 780 MWD/AT and its Faxfilm replica.

- (a) Specimen which read 5 Rad/hr at contact was etched in $\text{HF} + \text{HNO}_3 + \text{H}_2\text{O}$ and then photographed with a shielded metallograph; oblique illumination.
- (b) Replica of specimen shown in (a) as photographed on a normal metallograph.
- (c) Same as (b).

Photographs Unclassified

Macro: 3X

B.F. 250X

B.F. 250X

FIGURE 5.15. Photomicrographs of a cathodically vacuum etched uranium specimen irradiated to 580 MWD/T; oblique illumination.

Photographs Unclassified

UNCLASSIFIED

DECLASSIFIED

B.F. 250X

B.F. 250X

B.F. 75X

FIGURE 5.16. Photomicrographs of an aluminized Faxfilm replica of the irradiated uranium specimen shown in Figure 5.15; oblique illumination.

Photographs Unclassified

DECLASSIFIED

B.F. 500X

B.F. 500X

B.F. 500X

FIGURE 5.17. Photomicrographs of an aluminized Faxfilm replica of the irradiated uranium specimen shown in Figure 5.15; oblique illumination.

Photographs Unclassified

(a) B.F. 250X

(b) B.F. 250X

(c) B.F. 750X

(d) B.F. 750X

FIGURE 5.18. Photomicrographs of a cathodically vacuum etched uranium metallographic specimen, (a) and (c), currently being irradiated, and its replica, (b) and (d) oblique illumination.

Photographs Unclassified

DECLASSIFIED

A laboratory which will contain the electron microscope and necessary accessories is currently being arranged and will soon permit high resolution examination of replicas. Since specimen and replica preparation are of utmost importance to the proper interpretation of micrographs, a combined cathodic vacuum etching and evaporation chamber, shown in Figure 5.19, has been constructed. Replication by evaporated thin films immediately after the etching should avoid any possible surface contamination or oxidation which might otherwise invalidate the high resolution results; such evaporated replicas will serve as a standard for evaluating the quality of replicas which ordinarily are obtained in an air environment.

327 Basin Corrosion Study - G. R. Mallett

The rate and type of corrosion occurring with 1100 (2S) aluminum in the 327 storage basin is being determined. 200-300 ppm of sodium nitrate, as an aluminum corrosion inhibitor, has been maintained at all times. For seven to eight hours during each week day, a water flow of 30 gallons per hour is maintained to keep the basin water surface clean. The water enters at the bottom of the basin and goes out the overflow at the top. The fifteen 1100 (2S) aluminum coupons in the basin were removed on June 6, 1955, along with six control coupons, after a total of 90 days' exposure to basin conditions.

The visible corrosion, which appeared to be the same on all of the samples that were in the basin and the three in stagnant tap water, included a small amount of staining and some small white specks. The three coupons that were in running tap water were stained dark with a large amount of white corrosion product and barnacles.

The corrosion product and stains were removed by using a sulfuric-chromic acid bath. The amount of aluminum metal removed, due to the sulfuric-chromic acid bath, was determined by cleaning three 1100 (2S) aluminum coupons along with the corrosion coupons. The amount of metal removed due to cleaning was 0.40 mg/cm². The weight loss for each location was determined and is shown in Table 5.2.

TABLE 5.2

AVERAGE WEIGHT LOSS DUE TO CORROSION

<u>Location of Samples (three in each location)</u>	<u>Average Weight Loss mg/cm²/yr</u>
Storage can filled with basin water	0.29
Storage can filled with tap water	0.29
Resting in the slug storage rack	0.33
Resting on the bottom of the basin	0.14
Free of contact in the basin	1.04
Stagnant tap water	0.14
Running tap water	1.53

Pit depth measurements are being made to determine the rates of penetration due to corrosion.

FIGURE 5.19. Combined Cathodic Vacuum Etching and
Evaporation Chamber.
Photograph Unclassified

DECLASSIFIEDParting Lathe - E. C. Watts

A parting lathe for opening capsules containing irradiated preformed samples, Figure 5.20, was received from the 325 Technical Shop and set up in the Radio-metallurgy hot shop for preliminary tests. Cut-off tests were made on 2S aluminum, brass, mild steel, stainless steel, and zirconium in order to determine the most versatile parting tool design and the most suitable cut-off speeds. An 1/8-inch thick parting tool ground with a front clearance of 10 degrees and a back rake of 10 degrees proved to be the most versatile parting tool design, working equally well on all of the above metals. The machine showed a remarkable freedom from vibration and chattering and seemed to be satisfactory in all respects. The equipment has now been installed in a multicurie cell.

NaK Opening Equipment - E. C. Watts

Equipment to open capsules containing preformed samples in NaK, without metallurgical or mechanical damage to the samples, and to safely dispose of the NaK has been designed, fabricated, and installed in Cell E. This equipment consists of a remotely operated parting lathe, Figure 5.20, a stainless steel mortar, Figure 5.21, designed to accept adapters to hold various size capsules submerged in tertiary butyl alcohol. Provisions are made to maintain an inert atmosphere above the alcohol. A torque wrench with a built-in sample retriever completes this equipment. All capsules up to the size of a Hanford fuel slug can be opened and the NaK safely disposed of.

References

- (1) Duniway, R.R., Production Test 105-584A, Irradiation of Unbonded and Mechanically Bonded Slugs With Point Closures, HW-32886, Sept. 12, 1954 (SECRET).
- (2) Smith, E.A., PT-313-43-MT, Preparation of Slugs With Cold Pressure-Welded Closures for Pile Testing, HW-33016, Sept. 9, 1954 (SECRET).
- (3) Last, G.A., Point Closure Technique for Jacketing of Reactor Fuel and Target Materials, HW-33981, Feb. 9, 1955, (SECRET).
- (4) Cadwell, J.J., Quarterly Progress Report - Metallurgy Unit - Oct., Nov., Dec., 1954, HW-34455, Feb. 1, 1955 (SECRET).
- (5) Eschbach, E.A., Slug Densities Measured at Hanford, HW-29324, Sept. 11, 1953 (SECRET).
- (6) Metallurgy Research Sub-Section Quarterly Progress Report, Jan., Feb., March, 1955, HW-36054, May 16, 1955 (SECRET).
- (7) Personal Communication from R. G. Wheeler.

DECLASSIFIED

-5.26-

HW-37979

FIGURE 5.20. Parting Lathe Designed for
In-Cell Use. 1/2X

FIGURE 5.21. Stainless Steel Mortar. 1/5X

Photographs Unclassified

~~SECRET~~

DECLASSIFIED

CORROSION AND WELDING - W. R. Smith

Redox Phase II Corrosion Problems - N. D. Groves, M. C. Fraser, and W. L. Walker

The changes in stream composition brought about by the Phase II flowsheet and equipment alterations have raised the question of the potential corrosion problems involved under the new operating conditions. Past experience in the H-4 oxidizer and D-12 waste concentrator has indicated that solutions containing combinations of uranyl nitrate, aluminum nitrate, nitric acid, and sodium dichromate can be very corrosive in some circumstances.

A laboratory study has been completed wherein four candidate materials were exposed to seven potential Redox Phase II solutions representative of the F-2 concentrator, the D-14 concentrator, and the H-4 oxidizer. This study was designed to: (1) establish the presence and order of magnitude of the corrosion problem existing in these vessels, and (2) provide preliminary indications of the most suitable material of construction for repair and replacement purposes. The method of evaluation involved determining corrosion rates of wrought and weld metal specimens of AISI types 304L, 347, and 312 stainless steel and Grade A55 titanium exposed to the boiling synthetic solutions as static specimens immersed in the liquid and as heat transfer surfaces transferring heat to the liquid. The apparatus used and the types of specimens used as well as the preparation of the specimens is fully described in HW-34455⁽¹⁾. The metal temperature of the heat transfer specimens was maintained at 140 C, and the boiling point of the solutions ranged from 110 C to 115 C. The nominal solution compositions are shown in Table 6.1, along with the Redox stream which the solution represents. The duration of the exposures was 336 hours in all cases.

Since some of the solutions are highly concentrated salt solutions and tend to change composition rapidly when small amounts of water are removed, it was necessary to determine physical property curves which were used as a basis for controlling the solution composition during the test exposures. The apparatus used to determine the specific gravity vs. concentration and boiling point vs. concentration curves is shown in Figure 6.1. These curves were determined for uranyl nitrate, aluminum nitrate, and combinations of uranyl nitrate and aluminum nitrate. These curves are shown in Figures 6.2, 6.3, and 6.4.

The results of this study are tabulated in Table 6.2 and portrayed graphically in Figure 6.5. An examination of these data indicates that the presence of a severe corrosion problem potentially exists in the Redox D-14 concentrator and the H-4 oxidizer while the proper selection of construction material can reduce the corrosion rate in the F-2 concentrator to a tolerable level. A judicious selection of operating conditions appears to hold more promise for reducing the corrosion in the H-4 oxidizer than a change in material of construction until such time as titanium proves suitable for this application. It is true, however, that significant increases in service life may be realized in all of these vessels if they are constructed of type 304L or 312 stainless steel rather than type 347.

The more general conclusions which may be drawn from these tests include: (1) in these solutions, the corrosion rates of the materials exposed increases markedly when the specimens are exposed as heat transfer surfaces at an elevated temperature, (2) in the absence of free nitric acid the hexavalent chromium concentration has very little effect on the corrosivity of these solutions, and (3) titanium exhibits corrosion rates of the order of 100 times less than those of the stainless steels.

(1) All footnotes are referenced at the end of this section.

TABLE 6.1

SOLUTION COMPOSITION OF SYNTHETIC REDOX STREAMS

Solution Number	Plant Location	Operating Conditions	Constituent Molar Concentration					
			$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	$\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	HNO_3	$\text{Na}_2\text{Cr}_2\text{O}_7$	$\text{Fe}(\text{NH}_4\text{SO}_4)_2$	$\text{NH}_2\text{SO}_3\text{H}$
1	F-2 concentrator	1 stage backcycle	1.19	1.44	-0.20	0	0.002	0.003
2	F-2 concentrator	1 stage backcycle plus precycle	1.35	1.13	-0.20	0.05	0.002	0.003
3	F-2 concentrator	1 stage backcycle plus precycle	1.35	1.13	-0.20	0.02	0.002	0.003
4	D-14 concentrator	with or without precycle	2.45	0.01	-0.20	0.02	0.004	0.006
5	H-4 oxidizer	with free acid	0	2.50	0.20	0.15	0	0
6	H-4 oxidizer	neutral	0	2.50	0.01	0.15	0	0
7	H-4 oxidizer	acid deficient	0	2.50	-0.20	0.15	0	0

DECLASSIFIED

FIGURE 6.1. Continuous Concentrator for Boiling Point and Specific Gravity Determinations

FIGURE 6.2. Specific Gravity and Boiling Point Plotted Versus Aluminum Nitrate Concentration

FIGURE 6.3. Specific Gravity and Boiling Points Plotted Versus Uranyl Nitrate Concentration

FIGURE 6.4. Boiling Point and Specific Gravity Curves For an Approximately Equi-molar Solution of $\text{UO}_2(\text{NO}_3)_2$ and $\text{Al}(\text{NO}_3)_3$

TABLE 6.2

CORROSION RATES OF VARIOUS CONSTRUCTION MATERIALS EXPOSED TO BOILING SYNTHETIC REDOX PROCESS SOLUTIONS

Solution Number	Metal Temp. °C	Average Corrosion Rates (ipm)*						
		Wrought Type 304L	Type 308L Weld Metal	Wrought Type 347	Type 347 Weld Metal	Wrought Type 312	Type 312 Weld Metal	Grade A55 Titanium
1	110	0.0001	0.0001	0.0001	0.0001	Nil(3)	Nil	Nil
	145	0.0003	0.0003	0.0003	0.0004	0.0004	0.0002	Nil
2	110	0.0002	0.0002	0.0002	0.0002	0.0003	0.0003	Nil
	145	0.0014	0.0022	0.0040	0.0011	0.0016	0.0023	Nil
3	110	0.0002	0.0002	0.0002	0.0002	0.0002	0.0002	Nil
	145	0.0013	0.0008	0.0034	0.0012	0.0006	0.0012	Nil
4	110	0.0002	0.0003	0.0035	0.0005	0.0006	0.0007	Nil
	145	0.0034	0.0013	0.0069	0.0036	0.0006	0.0034	Nil
5	110	0.0075	0.0034	0.0104	0.0036	0.0028	0.0047	Nil
	145	0.0091	0.0032	0.0230	0.0124	0.0056	0.0063	Nil
6	110	0.0020	0.0019	0.0059	0.0027	0.0016	0.0018	-
	145	0.0070	0.0054	0.0140	0.0066	0.0038	0.0038	-
7	110	0.0003	0.0004	0.0003	0.0004	0.0005	0.0005	Nil
	145	0.0013	0.0027	0.0054	0.0027	0.0017	0.0004	0.0008

*Note: (1) Each test had a duration of 336 hours.

(2) All wrought material except Ti were sensitized at 1250°F for one hour and water quenched.

(3) Nil = < 0.00005 ipm.

FIGURE 6.5. Corrosion Rates of Various Alloys Exposed to Boiling Synthetic Redox Process Solutions.

Note: All wrought stainless steel was sensitized at 1250° F - 1 hr - H₂O quenched. Exposure time 336 hrs.

UNCLASSIFIED

Field Corrosion Tests in Underground Waste Storage Tanks - N. D. Groves, M. C. Fraser, and W. L. Walker

It has been recognized for several years that the limited laboratory data which served as a basis for the choice of construction material for the existing underground waste storage tanks at HAPO is inadequate with respect to predicting the service life of these tanks. Since some of the present tanks have been in service for several years and since the collection of adequate data upon which a judicious choice of replacement materials and service life prediction can be made requires several years, a long range program has been undertaken to provide such data.

The limitations of laboratory studies such as the difficulty of duplicating exact fission product concentrations and the inability of predicting and reproducing sludge formation by self-concentration on a laboratory scale indicates the desirability of collecting this data in the storage tanks themselves. Preliminary work⁽²⁾ by Endow and Sanborn has shown the feasibility of field investigations under the present operating conditions and indicated some of the difficulties which may be encountered.

In order to conduct corrosion tests in the underground waste storage tanks, three exposure units or facilities have been designed, built, and installed. These units essentially consist of tubes, inserted through the top of the storage tanks and reaching to the bottom of the tank, which have been perforated at the points of sample exposure to allow free circulation of the stored wastes. (See Figure 6.6). Each tube contains one set of samples exposed to the liquid wastes and one set of samples exposed to the vapors above the liquid waste. A unit containing two tubes has been installed in the Hot Semi-Works self-concentrator where the wastes from Purex studies in the Hot Semi-Works are being stored under controlled conditions to simulate Purex waste storage practices. It is intended that the samples from these tubes will be removed at the end of one and two years' exposure, respectively.

Two similar units, each containing seven tubes, have been installed in a Redox and Purex underground waste storage tank, respectively. It is intended that the samples from these tubes be removed after exposure intervals of 4, 8, 16, 32, 64, and 128 months with the seventh tube of each unit containing spare samples.

The materials being evaluated are the present material of construction, SAE 1020 carbon steel; two low alloy, high strength steels, Mayari-R and Corten; and an alloy steel, Corilloy T-1. Each set of samples includes wrought, welded, and stressed specimens of each of the materials as well as specimens of the weld metal normally used to fabricate each of these materials.

A formal report⁽³⁾ describing the details of design and installation of these exposures is being issued and due to the nature of this program, progress will be reported only at such times as sample removals are made.

Heat-Transfer-Corrosion Tests to Determine the Corrosivity of Synthetic 2WW Purex Waste Acid Concentrate - M. C. Fraser

A program is in progress to determine the corrosivity of boiling synthetic 2WW Purex waste acid concentrate by quantitative, semiquantitative, and qualitative methods upon types 304L, 347, and 312 stainless steel and type A-55 titanium under heat transfer conditions. The three-fold purpose of this program is: (1) the determination of the existence and order of magnitude of the corrosion problems which may be expected in the Purex acid concentrators; (2) the collection of engineering data

FIGURE 6.6. Waste Storage Tank Corrosion Test Facility

UNCLASSIFIED

to facilitate the judicious selection of replacement or repair materials and methods; and (3) the establishment of the effects of time and temperature on corrosion for operational data. The equipment and solution being used are described in HW-34455(1).

Quantitative data have been collected on the corrosive effects of boiling synthetic 2WW Purex waste acid concentrate upon wrought type 312 stainless steel heat transfer wafers. The specimens were maintained at an average temperature of 145 C for periods which ranged from 64 hours up to 2051 hours. These data are tabulated in Table 6.3.

TABLE 6.3

THE EFFECT OF TIME UPON THE CORROSION RATE OF WROUGHT TYPE 312 STAINLESS STEEL EXPOSED TO BOILING SYNTHETIC 2WW PUREX WASTE ACID CONCENTRATE UNDER HEAT TRANSFER CONDITIONS

<u>Specimen Temperature</u> °C	<u>Exposure Time</u> Hours	<u>Average Corrosion Rate</u> ipm
145	64	0.0014
"	132	0.0011
"	269	0.0013
"	549	0.0021
"	1045	0.0027
"	2051	0.0029

These data show that the corrosion rate of wrought type 312 stainless steel at 145 C increases slowly for approximately the first one thousand hours of exposure to boiling synthetic 2WW and remains essentially unchanged over the second thousand hours of exposure. This behavior and visual inspection of the corroded areas of the wrought type 312 specimens shows that the corrosion is relatively uniform over the exposed area.

Factorially Designed Static Corrosion Tests - W. L. Walker

Efforts to correlate the factorially designed static corrosion tests, in which a four or five component system is sampled statistically and the results from the exposures extrapolated to cover the entire system, with the heat transfer corrosion studies are still in progress. A detailed description of the techniques and apparatus involved in the two types of corrosion testing methods is available in HW-34455(1).

The most recent study in the factorial test involved AISI type 304L stainless steel in Purex 2WW. Time versus corrosion rate at 145 C and temperature versus corrosion rate from 80 C to 170 C curves were obtained. The time versus corrosion rate curve showed that the corrosion rate was constant with respect to time throughout the entire testing period, 10 days, and that the corrosion rate reached equilibrium within 24 hours; indicating that this method of study can provide usable corrosion data in much shorter periods of time than any of the other current methods. The temperature versus corrosion rate curve showed an exponential increase of corrosion rate with respect to temperature. The same types of curves have been obtained from the heat transfer test units, but the magnitude of the corrosion rates is different, and the difference is not constant with respect to temperature.

Efforts to determine the composition of the vapor phase inside the capsules were discontinued since the latest study indicates that this is not a controlling factor in corrosion of the liquid phase sample. Solution temperature appears to be the rate controlling factor, and studies are planned to isolate and evaluate this factor.

Bearing Studies - N. D. Groves

As a continuation of the search for a suitable bearing material to be used in Purex submerged rotating equipment, three grades of chromium-molybdenum-silicide compacts, produced by the Borolite Corporation and assigned the grade numbers of 1101, 223, and 226, have been exposed to boiling 65 percent nitric acid as a screening test. The following table indicates the comparative resistance of these grades to this media.

TABLE 6.4

CORROSION RATES OF CHROMIUM-MOLYBDENUM-SILICIDE
IN BOILING 65 PERCENT NITRIC ACID

<u>Sample Grade</u>	<u>Exposure Time (hrs)</u>	<u>Corrosion Rate (ipm)*</u>
223	240	0.0011
226	240	0.0014
1101	240	nil**
1101	500	nil**

*ipm = inches penetration per month

**nil = 0.0001 ipm

Since all three of these materials exhibit acceptable corrosion characteristics under the above conditions, it is intended that (1) the corrosive stability of this material be determined under conditions of intense radiation, and (2) full scale bearings be procured to determine the corrosion resistance under actual operating conditions.

Preliminary tests designed to compare Hard Chrome plating and Ultra Chrome plating have been completed. Twelve sections of type 304L stainless steel, measuring 1/4-inch diameter by 1-inch long, were plated with chromium to a thickness of 0.003 inch on the outer surface, exclusive of the ends. Six of the samples were plated by the Industrial Electroplating Company using the Hard Chrome process and six of the samples were plated by the Ultra-Chrome Corporation using the Ultra Chrome process.

One sample from each of the above groups was exposed to 30 and 60 weight percent nitric acid at room temperature, 80 C, and the boiling temperature. The exposures consisted of placing a passivated weighed sample in the desired media for 48-hour periods until 240 hours had elapsed. Quantitative determination of the corrosion rates on these specimens is impossible since the ends and interior of the specimens were not plated. The following qualitative breakdown is based on visual examination and experience gained during previous tests which revealed the mode of corrosion damage to be a rapid attack of the boundary layer between the chromium plating and the base metal.

TABLE 6.5

CORROSION RATES OF CHROMIUM PLATING EXPOSED TO NITRIC ACID

Nitric Acid w/o	Plating Type*	Temperature C		
		25	80	115
30	Hard Chrome	Satisfactory	Questionable	Unsatisfactory
65	Hard Chrome	Satisfactory	Unsatisfactory	Unsatisfactory
30	Ultra Chrome	Satisfactory	Questionable	Unsatisfactory
65	Ultra Chrome	Satisfactory	Unsatisfactory	Unsatisfactory

*Note: All samples were made using type 304L stainless steel as base metal.

It is significant to note that no apparent attack occurred on the plating but rather on the boundary layer between the plating and the base metal. Thus, these platings might serve satisfactorily in applications where the entire exposed surface could be plated.

The Weldability of an Alloy and Low Alloy Steels - G. W. Riedeman and L. C. Lemon

As part of a program to evaluate candidate construction materials for future underground waste storage tanks, three low alloy, high strength steel plates and one alloy steel plate, all one-half inch thick, were welded without preheat or postheat with low hydrogen iron powder coated electrodes of appropriate physical properties to match the respective steels. The three low alloy steel plates tested were "Mayari-R" (Bethlehem), "Corten" (USS), and "Yoloy" (Youngstown Sheet and Tube) and the alloy steel was "T-1" (USS). The electrodes used were "Atom Arc - 7016" (Alloy Rods) for the low alloy steels and "SW-91" (A.O. Smith) for the alloy plate.

The resulting weldments were subjected to a series of tests to evaluate the weldability of the plates and the strength of the welds. Standard free face and root bends and tensile tests were made as well as a special longitudinal weld face bend test. This latter test possesses considerable merit in demonstrating weldability and base plate notch sensitivity. The test involves loading the weld face in longitudinal tension (see Figure 6.7). If a crack that opens up in the weld propagates through the base plate in a brittle manner, the plate is considered to be "notch sensitive".

An examination of the results of these tests reveal that with one minor exception, the weld was at least as strong and ductile as the base material, and the weldments showed no tendency toward brittle failure.

Weld Joint Design for Butt Welding Pipe - G. W. Riedeman and L. C. Lemon

The evaluation of weld joint designs for butt welding pipe by the tungsten inert gas process (TIG) has continued through the last quarter. The object of this evaluation is to find a joint design that will consistently produce a high quality joint in the least time and can be non-destructively inspected.

Comparative tests have been performed on three types of joint, and the individual factors that comprise the total weld fabrication time and cost were recorded. The three types of joint are the conventional bevel joint, the EB consumable insert joint, and a new joint designed by the GE welding laboratory (see Figure 6.8). Carbon

FIGURE 6.7. Longitudinal Weld Face Bend Test

FIGURE 6.8

DECLASSIFIED

steel as well as stainless steel pipe of various sizes and schedules were employed in these tests. Although a systematic evaluation of these data has not been completed, the preliminary examination indicates the time and cost of the "GE" joint and the conventional bevel joint are comparable; whereas, the EB consumable insert joint is more tedious, time consuming, and costly. The advantages of the "GE" joint over the conventional bevel joint are the consistently reproducible quality of the joint with a minimum of operator skill and its ability to be inspected with ease and without destroying the joint. Additional cost reduction in the latter joint has been accomplished by developing a form cutting tool which reduces the joint preparation to two fast, simple operations. The tool will prepare the joint on the ends of all pipe having a wall thickness of greater than 1/8 inch.

The dissolved gases in rimming and semi-killed carbon steel has caused some difficulty in welding pipe fabricated from these steels, and several methods have been proposed to eliminate this problem. It is intended that the current testing program will be continued to evaluate the more promising of these methods.

Joint Design for Joining Tubes to Tube Sheets for Corrosive Radioactive Chemical Service - W. R. Smith

Joining tubes to tube sheets in the fabrication of tubular heat exchangers is a fabrication problem which is particularly acute in the fabrication of equipment for corrosive radioactive chemical service. Joints have been developed which are considered to be corrosion resistant and to be economically and easily produced. A report⁽⁴⁾ has been issued which describes the joints which have been developed for the case where the corrosive media is on the inside of the tubes and for the case where the corrosive media is on the outside of the tubes. This latter joint was described in a previous quarterly report⁽⁵⁾.

References

- (1) Cadwell, J.J., Quarterly Progress Report - Metallurgy Research Sub-Section - October, November, December, 1954, HW-34455, Feb. 1, 1955 (SECRET).
- (2) Endow, N. and Sanborn, K.L., Field Corrosion Test - SAE 1020 Carbon Steel in Redox Process Waste Solution Tank No. 104-S, HW-32755, Aug. 11, 1954 (SECRET).
- (3) Groves, N.D., Fraser, M.C., and Walker, W.L., Field Corrosion Tests in Redox and Purex Underground Storage Tanks, HW-37642, June 28, 1955 (OUO).
- (4) Smith, W.R., Joint Design for Joining Tubes to Tube Sheets for Corrosion Radioactive Chemical Service (to be issued).
- (5) Metallurgy Research Sub-Section Quarterly Progress Report, Jan., Feb., March, 1955, HW-36054, May 16, 1955 (SECRET).