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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING SEPTEMBER, 1957

by

Russell W. Dayton
Clyde R. Tipton, Jr.

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BMI-1220 "Progress Relating to Civilian Applications During August, 1957", by Russell W. Dayton and Clyde R. Tipton, Jr. (Declassified version to be made available by TISE.)
A. DEVELOPMENTS FOR ZIRCONIUM-CLAD FUEL ELEMENTS

F. R. Shober

Construction of the equipment for measuring the thermal conductivity of irradiated uranium rod clad with Zircaloy 2, with NaK as the heat-transfer medium, has been completed and calibration will be made using standard unirradiated specimens. Apparatus designed to measure the thermal conductivity of UO₂ is being assembled to evaluate the design prior to completion of remote-operation equipment. A series of creep tests at elevated temperature has been started using 15 per cent cold worked Zircaloy 2 as test material.

Burst testing of welded zirconium and Zircaloy 2 tube has been completed at room and elevated temperatures. Evaluation of the bond strength of zirconium cladding to electropolished uranium produced by hot hydrostatic pressing is being evaluated.

Thermal Conductivity of Uranium and UO₂

H. W. Deem and C. F. Lucks

The effect of irradiation on the thermal and electrical conductivities of uranium and on the thermal conductivity of uranium oxide is being investigated.

Uranium

Exploratory work on unirradiated uranium will determine the feasibility of making thermal-conductivity and electrical-resistivity measurements on irradiated uranium rods that are clad in Zircaloy 2 with NaK as the heat-transfer medium. The steady-heat-flow method will be used. A regulated-voltage power source has been completed. The apparatus is being assembled and a calibration run, using an Armco iron specimen, should be completed by the end of October.

Uranium Dioxide

A detailed study has been made on the proposed design of the thermal-conductivity apparatus. Because of the small specimen size, very small heat flows are involved. Experience indicates that temperatures and heat flows will be difficult to measure to the required accuracy. Proper guarding will also be difficult to maintain.

The apparatus will be assembled to evaluate the design without all the remote-operation equipment. It will be checked with specimens of known thermal properties.

In the next work period the apparatus will be assembled and evaluation runs started.
The creep properties of Zircaloy 2 and Zircaloy 3 are being studied in the 300 to 650°F range and burst strengths of welded zirconium and Zircaloy 2 tubes are being determined. The creep tests are part of an investigation to study the variation of mechanical properties with individual lots of Zircaloy 2 and to determine to what extent the cold-worked properties of Zircaloy 2 can be retained during creep at elevated temperatures.

The results of the creep tests in progress are given in Table A-1. The creep rates for those test in progress over 1000 hr are less or equal to those of previous lots of material at comparable stresses. However, total deformation obtained from the current lot at comparable stresses are higher by approximately a factor of 2. Another series of tests has been initiated on 15 per cent cold-worked material.

The burst strength of 0.50- and 0.625-inch-diameter welded tubes has been investigated for temperatures up to 650°F and data are reported in Table A-2. Burst strengths of welded tubes appear to be generally higher for comparable tube diameters, wall thicknesses, and temperatures. This completes the testing program on tubes.

**Zirconium Cladding**

F. R. Shoher and R. F. Dickerson

Evaluation of the metallurgical bond formed between electropolished uranium and zirconium cladding by hot hydrostatic pressing is continuing. Equipment is being prepared to evaluate the bond strength by welding a threaded zirconium stud to the cladding and measuring the load necessary to separate the cladding from the core.

An electropolished specimen was canned and pressurized at 10,000 psi for 1 hr at 1350°F. The specimen was sectioned and examined. The bond area was discontinuous in approximately 2 per cent of the area. A very thin diffusion zone existed. This bond will be evaluated further by water quenching from 730°C and the determination of bond strength by the method previously described.

**Preparation of Aluminum, Molybdenum, and Zirconium Single Crystals**

J. A. De Mastry, F. R. Shoher, and R. F. Dickerson

The preparation of single crystals of molybdenum and zirconium for irradiation studies is continuing.

Growth of single crystals of molybdenum in a modified Andrade furnace is being attempted. A small amount of grain growth in 1/4-inch-diameter rods has been observed; however, optimum conditions necessary to obtain maximum grain growth have not been determined. Future work will include investigation of time and temperatures for growth of single crystals.
TABLE A-1. CREEP PROPERTIES OF ZIRCALOY 2 AND ZIRCALOY 3 AT ELEVATED TEMPERATURES

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Temperature, °F</th>
<th>Stress, psi</th>
<th>Time at Test, hr</th>
<th>Load On</th>
<th>50 Hr</th>
<th>100 Hr</th>
<th>500 Hr</th>
<th>1000 Hr</th>
<th>1500 Hr</th>
<th>2000 Hr</th>
<th>Load Off</th>
<th>Creep Rate (a) Per Cent, per hour</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zircaloy 2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1-7</td>
<td>450</td>
<td>20,000</td>
<td>1775(b)</td>
<td>0.25</td>
<td>1.680</td>
<td>1.695</td>
<td>1.702</td>
<td>1.705</td>
<td>1.718</td>
<td>--</td>
<td>--</td>
<td>&lt;0.00001</td>
</tr>
<tr>
<td>15-5</td>
<td>550</td>
<td>17,000</td>
<td>1500(b)</td>
<td>0.38</td>
<td>1.065</td>
<td>1.075</td>
<td>1.085</td>
<td>1.086</td>
<td>1.087</td>
<td>--</td>
<td>--</td>
<td>&lt;0.00001</td>
</tr>
<tr>
<td>15-3</td>
<td>550</td>
<td>15,000</td>
<td>1550(b)</td>
<td>0.16</td>
<td>0.470</td>
<td>0.475</td>
<td>0.480</td>
<td>0.480</td>
<td>0.490</td>
<td>--</td>
<td>--</td>
<td>&lt;0.00001</td>
</tr>
<tr>
<td>34-7</td>
<td>550</td>
<td>14,000</td>
<td>1525(b)</td>
<td>0.16</td>
<td>0.375</td>
<td>0.382</td>
<td>0.392</td>
<td>0.395</td>
<td>0.402</td>
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<tr>
<td>15-4</td>
<td>550</td>
<td>12,500</td>
<td>1500(b)</td>
<td>0.15</td>
<td>0.22</td>
<td>0.223</td>
<td>0.230</td>
<td>0.240</td>
<td>0.240</td>
<td>--</td>
<td>--</td>
<td>&lt;0.00001</td>
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<tr>
<td>15-6</td>
<td>650</td>
<td>17,850</td>
<td>1500(b)</td>
<td>1.00</td>
<td>1.40</td>
<td>1.410</td>
<td>1.475</td>
<td>1.490</td>
<td>1.570</td>
<td>--</td>
<td>--</td>
<td>0.00002</td>
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<tr>
<td>15-1</td>
<td>650</td>
<td>15,000</td>
<td>2034.3</td>
<td>0.35</td>
<td>0.65</td>
<td>0.658</td>
<td>0.703</td>
<td>0.733</td>
<td>0.750</td>
<td>0.775</td>
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<tr>
<td>20-5</td>
<td>650</td>
<td>14,000</td>
<td>325(b)</td>
<td>0.128</td>
<td>0.148</td>
<td>0.164</td>
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<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
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<tr>
<td>20-4</td>
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<td>12,500</td>
<td>1250(b)</td>
<td>0.145</td>
<td>0.200</td>
<td>0.21</td>
<td>0.25</td>
<td>0.275</td>
<td>--</td>
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<td>--</td>
<td>0.00005</td>
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<tr>
<td>7-11(c)</td>
<td>650</td>
<td>30,000</td>
<td>100(b)</td>
<td>0.315</td>
<td>0.537</td>
<td>0.592</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Zircaloy 3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>3-11</td>
<td>300</td>
<td>20,000</td>
<td>1600(b)</td>
<td>0.17</td>
<td>1.24</td>
<td>1.37</td>
<td>1.625</td>
<td>1.75</td>
<td>1.76</td>
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<td>--</td>
<td>0.00025</td>
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<tr>
<td>3-12</td>
<td>300</td>
<td>17,500</td>
<td>1676(b)</td>
<td>0.14</td>
<td>0.75</td>
<td>0.84</td>
<td>1.035</td>
<td>1.10</td>
<td>1.11</td>
<td>--</td>
<td>--</td>
<td>0.00015</td>
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</table>

(a) Based on creep deformation between 500 and 1000 hr.
(b) Tests in progress.
(c) Cold work 15 per cent, longitudinal specimen.
<table>
<thead>
<tr>
<th>Specimen</th>
<th>Material</th>
<th>OD, in. Nominal</th>
<th>OD, in. Measured</th>
<th>Wall Thickness, in. Nominal</th>
<th>Wall Thickness, in. Measured</th>
<th>Cold Work, per cent</th>
<th>Burst Pressure, psi</th>
<th>Burst Stress (a), psi</th>
<th>Circumferential Elongation, per cent</th>
<th>Remarks</th>
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<td>273-8(b)</td>
<td>Zircaloy 2</td>
<td>0.625</td>
<td>0.625</td>
<td>0.049</td>
<td>0.047</td>
<td>10-15</td>
<td>11,700</td>
<td>62,160</td>
<td>10.0</td>
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<tr>
<td>273-8(b)</td>
<td>Ditto</td>
<td>0.625</td>
<td>0.625</td>
<td>0.049</td>
<td>0.048</td>
<td>10-15</td>
<td>11,650</td>
<td>61,160</td>
<td>10.0</td>
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</tr>
<tr>
<td>328-8(b)</td>
<td>Ditto</td>
<td>0.500</td>
<td>0.500</td>
<td>0.035</td>
<td>0.035</td>
<td>10-15</td>
<td>11,100</td>
<td>68,185</td>
<td>53.4</td>
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<tr>
<td>328-8(b)</td>
<td>Ditto</td>
<td>0.500</td>
<td>0.499</td>
<td>0.035</td>
<td>0.035</td>
<td>10-15</td>
<td>11,100</td>
<td>65,825</td>
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<td>485-1-4</td>
<td>Zircaloy 2</td>
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<td>0.625</td>
<td>0.050</td>
<td>0.0515</td>
<td>10</td>
<td>11,400</td>
<td>58,790</td>
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<td>Pin hole failure in weld</td>
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<td>0.050</td>
<td>0.051</td>
<td>10</td>
<td>12,200</td>
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<td>0.500</td>
<td>0.035</td>
<td>0.035</td>
<td>10</td>
<td>11,600</td>
<td>72,485</td>
<td>53.6</td>
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<td>484-1-6</td>
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<td>0.625</td>
<td>0.625</td>
<td>0.035</td>
<td>0.035</td>
<td>10</td>
<td>11,800</td>
<td>72,485</td>
<td>57.4</td>
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<tr>
<td>650 F</td>
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<td></td>
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<td>0.625</td>
<td>0.050</td>
<td>0.051</td>
<td>10</td>
<td>10,000</td>
<td>51,590</td>
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<td>0.625</td>
<td>0.050</td>
<td>0.051</td>
<td>10</td>
<td>9,900</td>
<td>50,070</td>
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<td>0.600</td>
<td>0.50</td>
<td>0.035</td>
<td>0.035</td>
<td>10</td>
<td>6,500</td>
<td>39,785</td>
<td>1.0</td>
<td>Pin hole failure</td>
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<td>484-1-2</td>
<td>Ditto</td>
<td>0.600</td>
<td>0.50</td>
<td>0.035</td>
<td>0.035</td>
<td>10</td>
<td>9,500</td>
<td>58,360</td>
<td>51.8</td>
<td></td>
</tr>
</tbody>
</table>

(a) Calculated by Barrows formula, \[ S = \frac{P_d}{2t} \] where \( S \) = hoop stress, \( P \) = internal pressure, and \( t \) = wall thickness.

(b) Seamless tubes.
A-5 and A-6

A variation of the strain-anneal technique is being tried on small zirconium rods. Tensile bars of 0.05- and 0.125-in. diameters were prepared. These bars had grain size of 0.011 mm at 100X before any work was started.

The 0.50-in. bars were elongated in the range of 1.5 to 3.0 per cent. The annealing temperature of these bars was increased from 1150 to 1550 F in 100 steps at 24-hr intervals. No appreciable grain growth was obtained. The annealing time at 1550 F is to be increased for several specimens to determine the effect of annealing on grain growth.

The 0.125-in. bars were elongated from 2.0 to 13.0 per cent and the same annealing schedule as used for the 0.5-in. bars was followed. Some grain growth was noted in the more highly strained specimens. After examination, these specimens were elongated an additional 2.0 to 13.0 per cent and the step anneal repeated. This second straining and annealing resulted in grain growth from 0.011 mm at 100X, the original grain size, to greater than 0.500 mm at 100X.

However, some hydride formation is evident in these specimens. They will be vacuum annealed to remove these hydrides since it is believed that hydrides inhibit grain growth. Further studies on the dehydrided specimen will continue. A third elongation is planned along with lengthening of the annealing time.

Preparations for straining at temperatures of 800 to 850 C are being made.
B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

The "right-angle" extrusion-cladding assembly is expected to be completed by the end of October. Meanwhile, work has continued toward improvement of the extrusion-cladding process, with particular attention being paid to reducing extrusion pressure. The four-port mandrel, a new six-port streamlined mandrel, and an older six-port mandrel recently streamlined have been tested during the past month. The pressures were somewhat lower than had been achieved previously.

The evaluation of a group of air-melted billets of aluminum-45 w/o uranium produced by centrifugal-casting techniques has been completed. Ingots produced in molds rotating at a speed of approximately 975 rpm and poured at a temperature slightly below 2500 F exhibited good surfaces, little porosity, and a tendency to segregate from end to end. It is believed that the uranium-content variations from end to end can be reduced by the utilization of a suitable metal-distributing device.

The construction of a mechanical extensometer which is expected to possess sufficient sensitivity and range to study the transverse mechanical properties of uranium tubes is nearly complete. This extensometer will be calibrated through the use of standard gage blocks, and correction curves obtained for use in the study of the tube specimens.

Extrusion Cladding of Flat Plates

R. J. Fiorentino, C. J. Slunder, and A. M. Hall

Work was continued toward the improvement of the extrusion-cladding process, with particular attention paid to reducing extrusion pressure. The four-port mandrel (Tip 6), a new six-port streamlined mandrel (Tip 7), and a six-port mandrel (Tip 3) in which the shape was revised to reduce the angle at which the aluminum contacts the core were tested in September. Tested under similar conditions, the pressure required with these three tips was in the range from 26,000 to 28,000 psi. This pressure range is somewhat lower than has been achieved thus far. It is planned to continue the testing of these tips under different conditions and also the modifying of their shapes in attempts to lower the pressure still further.

Mandrel Tip 5 was designed with a heavy center web that was intended to reinforce the mandrel slot. This web restricted the flow of metal to the center of the plate. Therefore, the web has been cut down stepwise to improve the metal flow. Increased metal flow in the center region has been achieved with no apparent sacrifice of support for the mandrel slot.

The "right-angle" extrusion cladding assembly is expected to be completed by the end of October. Meanwhile, the necessary auxiliary equipment for the assembly is being prepared.
Study of the Liquidus in Aluminum-Uranium Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

The investigation of the liquidus line of the aluminum-uranium system is continuing. Included in this study is a determination of the effects of additions of silicon and magnesium on the liquidus.

A tentative liquidus line has been determined up to 30 w/o uranium for the binary system. Differential-thermal-analysis breaks have been recorded at 780 C for a 17.2 w/o uranium alloy, at 885 C for a 20 w/o uranium alloy, and at 1068 C for a 30 w/o uranium alloy. With the exception of the 885 C liquidus temperature for the 20 w/o uranium alloy, the data are in agreement with previously published data for 16 w/o and 24.5 w/o uranium alloys (BMI-1066). Chemical analysis may prove the composition of the 20 w/o alloy to be higher in uranium than intended.

Thermal analysis has also confirmed the peritectic temperature at 732 C. To date, no effect of silicon or magnesium on the liquidus has been detected. However, silicon appears to have a refining effect on eutectic UAl$_4$ while having no effect on peritectic UAl$_4$.

Alloys containing 40 and 50 w/o uranium are being prepared for study. Meanwhile, the investigation into the effects of silicon and magnesium on the liquidus temperature at the lower uranium compositions is continuing.

Preparation of Aluminum-Uranium Alloys

N. E. Daniel, E. L. Foster, and R. F. Dickerson

Aluminum-uranium alloys of relatively high uranium content are of interest as possible fuels for low-temperature water-cooled reactors. Because of this interest, a program of research concerned with the melting and casting of such alloys containing up to 50 w/o uranium is in progress. The ultimate objective of this program is the development of production techniques capable of yielding sound homogeneous extrusion billets in the form of relatively heavy-walled cylinders. An investigation of the effects of magnesium and silicon in amounts up to 1 w/o on the casting properties of the alloys and the determination of the upper limit of uranium that can be incorporated into an acceptable extrusion billet by conventional air-melting techniques are also under way.

The evaluation of a group of air-melted ingots containing 45 w/o uranium produced by centrifugal-casting techniques has been completed. Variations in uranium content were noted from end to end and from the inside to outside surface of ingots poured at 2500 F into molds rotating between 550 and 750 rpm. The surface of these castings were rated fair. Ingots produced in molds rotating at a speed of approximately 975 rpm and poured at slightly lower temperatures exhibited better surfaces, and uranium segregation from inside to outside surfaces was decreased significantly. Little improvement in uranium homogeneity from end to end was noted with these changes in conditions.
Further work will be concerned with the evaluation of the variables encountered in centrifugal air casting. It is believed that the uranium-content variation from end to end can be reduced by the utilization of a suitable metal-distributing device.

A portion of the program has been devoted to the preparation of sound homogeneous alloy castings by vacuum melting and casting techniques. While the use of this technique for production of extrusion billets is acknowledged to be cumbersome, it represents an alternate method by which the billets could be obtained. A very sound homogeneous ingot containing 45 w/o uranium was produced with this technique by melting in a zirconia crucible and pouring into a graphite mold heated to 1000 F. The pouring temperature was 2450 F and care was exercised to prevent the pour from streaming down the mold surface. Good surfaces were obtained on both inside and outside. Additional examination by radiographic and sectioning techniques failed to reveal any areas of extensive porosity. The chemical analyses of this ingot showed the maximum uranium concentration of 45.9 w/o to be located at the top inside and the lowest uranium concentration of 44.7 w/o to be in the bottom inside.

Development of a Mechanical Extensometer

J. A. DeMastry, F. R. Shober, and R. F. Dickerson

The development of a mechanical extensometer with sufficient sensitivity and range to study the transverse mechanical properties of uranium tube is continuing. Construction of an extensometer is nearly complete.

The extensometer consists of a spring-loaded divided expansion ring, inter-sliding rods and tubes attached respectively to the halves of the ring, and provisions for either cantilever or loop application of electrical strain gages. Knife-edge supports on the ring are so positioned that any change in the diameter of the specimen is measured by the strain gages through the relative displacement of the rod and tube. The output of these gages can either be read directly from a strain-gage indicator or automatically recorded on an appropriate recorder.

This extensometer will be calibrated through the use of standard gage blocks, and correction curves obtained for use in the study of the tube specimens.
C. PLANT ASSISTANCE TO MCW

A. A. Bauer

Studies of structural and compositional variations of the uranium oxides are continuing. Investigation of the constitution of active and inactive UO₂-oxygen systems shows the systems to be identical below UO₂,25. Above this composition changes in phase behavior are noted. Attempts to confirm the effect of nitrogen on the reoxidation rate of UO₂ were unsuccessful. However, nitrogen in UO₂ is continuing to receive study; initial tests to determine nitrogen in UO₂ by a colorimetric method have indicated that a difference between the nitrogen content of several samples exists.

Examination of WAPD UO₂ samples before and after micronizing reveals no evident differences in structure.

Corrosion tests of Type 304 ELC stainless steel in fluoride- and chloride-contaminated nitric acid solutions of 5, 18, 30, 38, and 45 w/o have continued. No pronounced changes in corrosion rate over these previously reported have been noted. Tests in 38 w/o nitric acid solutions containing fluoride, chloride, and aluminum additions, with and without iron additions, have continued. Iron is found to be beneficial in decreasing the rate of corrosion.

Initial tests have been conducted to measure the rate of hydrogen permeation through molten slag. Experimental difficulties have necessitated limited design changes which will be retested shortly.

A Brookfield viscometer has been selected for viscosity-measurement studies of magnesium fluoride slag. Samples of the slag are to be contained in a tantalum crucible for the determinations.

Investigation of Uranium Oxides

D. A. Vaughan, J. R. Bridge, and C. M. Schwartz

Structural and compositional variations in uranium oxides are being investigated as assistance in the production of chemically reactive uranium dioxide and in improving metal quality. Previous reports have related the structural and compositional variations of UO₃ and UO₂ to the method of pyrolysis of uranyl nitrate hexahydrate. Colorimetric methods were developed for the analyses of nitrogen in UO₃.

During the past month work was completed on the inactive UO₂-oxygen system and the active UO₂-oxygen system. Below the composition of UO₂,25 the inactive UO₂ and active UO₂ oxygen systems are essentially identical. However, above UO₂,25 the inactive oxide forms a tetragonal oxide, U₃O₇, while the active oxide remains cubic to a composition of UO₂,40. The lattice parameter of this cubic oxide does not change with composition, implying either that the oxygen is physically adsorbed or that a second phase, possibly U₃O₈, is being formed but not detected. The thermal expansion of this
cubic oxide is linear \( (\alpha = 9.27 \times 10^{-6}) \) over the temperature range 25 to 700 C while the cubic oxide \( \text{U}_4\text{O}_9 \) is linear \( (\alpha = 11.8 \times 10^{-6}) \) only over the range 300 to 700 C.

An attempt was made to confirm the observation, indicated in BMI-1205, that reoxidation of \( \text{UO}_2 \) is accelerated with decreasing nitrogen content of \( \text{UO}_3 \). Preparations of \( \text{UO}_3 \) with various nitrogen contents were made by the procedure outlined in BMI-1205. These were reduced, after which the reoxidation in room-temperature air was determined over a 3-week period. The spread in rate of \( \text{UO}_2 \) reoxidation for each nitrogen level of the \( \text{UO}_3 \) precludes finding any systematic trend.

Although no trends were observed in the above work, the retention of nitrogen in \( \text{UO}_2 \) may not be directly related to the nitrogen content of the prior \( \text{UO}_3 \). Previous attempts to determine the nitrogen content in \( \text{UO}_2 \), by the Kjeldahl method, revealed no differences. However, a colorimetric method, which proved reliable for the analysis of nitrogen in \( \text{UO}_3 \), is being applied to \( \text{UO}_2 \). The initial tests have indicated a difference in the nitrogen content of several \( \text{UO}_2 \) preparations.

Mallinckrodt experiments indicate that micronizing WAPD \( \text{UO}_2 \) improves the sinterability of this oxide. A sample of WAPD \( \text{UO}_2 \) before and after micronizing was examined at Battelle for structural differences. The tests and results are given in Table C-1. There was no significant difference between the two samples in these tests except for the oil odors present in the micronized samples.

**TABLE C-1. STUDY OF THE EFFECTS OF MICRONIZING WAPD \( \text{UO}_2 \)**

<table>
<thead>
<tr>
<th>WAPD ( \text{UO}_2 ), Lot 291-106-D</th>
<th>Micronized WAPD ( \text{UO}_2 ), Lot 291-106-E</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tap Density, g per cm(^3)</td>
<td>2.50</td>
</tr>
<tr>
<td>Oxygen-Uranium Ratio, As Received</td>
<td>2.064</td>
</tr>
<tr>
<td>Oxidized 48 Hr, 180 C</td>
<td>2.24</td>
</tr>
<tr>
<td>Structure of Oxidized Phase</td>
<td>Tetragonal</td>
</tr>
<tr>
<td>Lattice Parameter, As Received, A</td>
<td>5.468(_g)</td>
</tr>
<tr>
<td>Appearance</td>
<td>Dark brown</td>
</tr>
<tr>
<td>Odor</td>
<td>None</td>
</tr>
<tr>
<td>Particle Size, ( \mu )</td>
<td>3.5</td>
</tr>
<tr>
<td>Crystallite Size, ( A )</td>
<td>&gt;3000</td>
</tr>
<tr>
<td>Conductivity, As Made, ohm-cm(^{-1})</td>
<td>7 x 10(^{-4})</td>
</tr>
<tr>
<td>Conductivity, Annealed 200 C, ohm-cm(^{-1})</td>
<td>3.2 x 10(^{-3})</td>
</tr>
<tr>
<td>Carrier Type</td>
<td>P</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The Electrical Properties of Uranium Dioxide

J. W. Moody, R. K. Willardson, and H. L. Goering

Experimental studies are proceeding at a limited rate. There are no new data to report.

The Corrosion Resistance of Selected Stainless Steels

C. L. Peterson, W. C. Baytos, and F. W. Fink

Investigations have been continued on the corrosion of Type 304 ELC stainless steel by chloride- and fluoride-contaminated nitric acid solutions similar to those that might be expected to occur in the nitric acid reconcentrator at the Weldon Springs uranium refinery.

The prolonged exposure tests in 5, 18, 30, 38, and 45 w/o nitric acid solutions described in Table C-2 of BMI-1220 are still in progress. There has been no radical change in results from those listed in this table. There are indications that the chloride concentration is not being retained during the prolonged boiling of the 38 and 45 w/o nitric solutions. The rate of this loss of chloride will be studied and appropriate additions of chloride or changes of solution will be made to insure that the proper test conditions are maintained.

A series of seven exposure tests conducted for 48-hr periods have been completed with 38 w/o nitric acid solutions containing fluoride, chloride, and aluminum additions, with and without additions of iron. The results show that iron additions are definitely beneficial in decreasing the corrosion caused by these conditions. However, more investigations will be necessary to determine whether the severe corrosion noticed in some cases in the absence of iron is due entirely to chlorides, or perhaps to chlorides and fluorides, since the 6-to-1 weight ratio of aluminum which was added to complex the fluoride may not have been adequate in this concentration of nitric acid.

Gas-Metal Studies

J. B. Bodkin, W. R. Hansen, M. J. Trzeciak, and M. W. Mallett

A study is being conducted on two phases of research related to production of dingot uranium metal. One phase is concerned with determining means of lowering the residual hydrogen content of the uranium. This involves evaluation of the relationships among hydrogen, feed materials, and products. Presently, the permeation of hydrogen through magnesium fluoride slag is being measured.
The other phase is concerned with measuring the effect of various additives on the viscosity of magnesium fluoride slag. The viscosity is believed to be pertinently related to the uranium metal yield. Within limits, the lower the viscosity of the slag the higher should be the uranium metal yield. A summary of progress on the two phases follows.

**Permeation of Hydrogen**

A tentative experimental procedure for measuring the permeation of hydrogen through molten magnesium fluoride slag was reported in BMI-1220. An initial run was made in which it was found that the cylindrical molybdenum tube carried a sufficient amount of heat away from the heat zone to melt the Apiezon "W" wax in a taper joint holding the tube in place. Therefore, it was necessary to provide a cooling coil around the tube just below the wax joint. In an experiment made with the modified tube, the wax joint was found to hold satisfactorily. However, molten slag leaked from the bottom of the reaction tube because of a loose fit between the molybdenum diaphragm and the reaction tube. Apparently the molten slag is more fluid than had been expected. The diaphragm fitting has been redesigned and will be tested in the next run.

**Viscosity of Molten Slag**

Initially it was planned to use a system similar to that of Nicholls and Reid* for measuring viscosity. However, a literature search has indicated that the viscosity of molten salts is of the order of 1 centipoise which is less than the sensitivity of this equipment. Therefore, a Brookfield viscometer, an instrument of greater sensitivity, has been selected for making isothermal viscosity measurements. The sample of magnesium fluoride will be contained in a tantalum crucible held under argon and heated by induction. Determinations will be started in the near future.

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D. SOLIDIFICATION OF URANIUM

E. L. Foster, C. K. Franklin, B. Schwartz, and R. F. Dickerson

Battelle is involved in a theoretical study on the solidification of uranium metal. The objective of this program is a fuller understanding of the problems inherent in the production of large uranium castings. Hence, on the basis of such knowledge, any future considerations in varying factors such as ingot shapes and mold material could be properly evaluated as to their role in obtaining optimum casting results.

The over-all program includes a literature survey, mathematical analyses of heat flow, and experimental determinations of physical properties as required, in addition to the confirmation of the theoretical studies by small-scale experiments.

Work to date has been concerned with the literature survey and evaluation of the metallurgical factors involved in the solidification study. Also being scrutinized are results of studies conducted in the past by uranium producers. Of particular note has been the effect of melting and casting variables on the occurrence of character of ingot defects. In relation to the mathematical treatment of heat flow, the initial formulation of the problem was undertaken. The relationships established at this point for analytic purposes were a set of simultaneous equations showing the transient heat flow in each of the cells into which the casting and mold is divided. These equations describe both the conductive and radiation flow of heat, and convective heat transfer can also be included if necessary. This formulation indicates which thermal and physical constants are required for a preliminary analysis, and these are being assembled from available data. This initial formulation of the problem is not regarded as final and it is expected that it will be constantly improved as the analysis progresses. Programming of these equations for solution by the computer is the next task.

Future work will be involved with the role of metallurgical factors in the formulation and analysis of the heat flow in uranium casting. The determination of physical properties not available will be initiated as the requirements of the heat-flow analysis are resolved.
E-1

E. GENERAL FUEL-ELEMENT DEVELOPMENT

F. A. Rough and D. C. Carmichael

In this section, fuel-element development programs supported by the AEC Division of Reactor Development are reported. Stainless steel-clad specimens containing UC or UN fuel dispersed in a stainless steel matrix have been prepared for irradiation testing. Similar samples have been fabricated for tensile tests and additional specimens have been roll clad to determine the influence of the rate of reduction during rolling on the resulting core structure.

Techniques are being investigated for the preparation of cermet fuel elements with loadings of 60 to 90 volume per cent fuel compound dispersed in a metal matrix. Preliminary work on UO₂ stainless steel mixtures has indicated that hot pressing is preferable to sintering or infiltration methods for preparing these cores. A more detailed investigation of hot-pressing variables is being made on this system and cores of UO₂ dispersed in molybdenum and UN in stainless steel are being hot pressed.

An investigation is being conducted to study the feasibility of using the hydride of a uranium-zirconium alloy as a fueled moderator. An apparatus is being constructed which will be used to determine phase relations in the hydrogen-zirconium-uranium system.

Development of Dispersed UC and UN Fuel Elements

S. J. Paprocki, D. L. Keller, and G. W. Cunningham

Fabrication of irradiation specimens has been completed. Specimens of 24.2 w/o UN and 24.3 w/o UC dispersed in stainless steel and clad with Type 347 stainless steel will be irradiated to burnups of 10, 15, and 20 a/o uranium-235 at 1500 F.

A limited number of specimens of these compositions have been roll clad and will be machined into tensile specimens. The ultimate tensile strength will be measured at 1100, 1600, and 1700 F.

In addition, a series of specimens has been rolled to determine the effect of total reduction rate on the core structure. Metallographic examination has not yet been made.

Fabrication of Cermet Fuel Elements

S. J. Paprocki, D. L. Keller, and G. W. Cunningham

Methods for preparing cermets with a high uranium loading (60 to 90 volume per cent uranium compound) are being investigated. In addition, bend tests will be run and
electrical-conductivity measurements will be made. Also, a limited amount of work will be directed toward the development of cladding techniques.

All preliminary work has been conducted on the UO$_2$-stainless steel system, and indications are that the hot-pressing method results in structures which are stronger and more dense than sintered specimens. The sintered specimens have densities of approximately 75 per cent of theoretical as compared with hot-pressed densities of 90 per cent of theoretical. In addition, the stainless steel skeleton which remains after removal of the UO$_2$ with boiling nitric acid appears to be stronger, and there appears to be more growth across individual particle surfaces in the case of hot-pressed specimens. Uniform dispersions are obtained by both methods.

Furthermore, infiltration procedures do not appear promising. Sintering of mixtures above the melting point of stainless steel results in a poor structure, since the stainless steel does not wet the UO$_2$. However, it may be possible to improve the density and structure of the hot-pressed specimens with a final sinter at approximately 2500 F. Hot-pressed specimens are being sintered to determine the feasibility of such an operation.

A more detailed investigation of the hot-pressing procedure is in progress. Specimens of UO$_2$ and Type 302B stainless steel will be sealed in picture-frame packs and pressed at 50 tsi and temperatures of 1800, 1900, 2000, and 2100 F. Variables to be investigated include the particle size of UO$_2$ and stainless steel, and the UO$_2$ loading in the range 60 to 90 volume per cent. Densities, bend-test breaking strength, and electrical conductivity will be measured and compared with values for high-density UO$_2$.

In addition, specimens of 80 volume per cent UO$_2$ in molybdenum and 80 volume per cent UN in stainless steel will be hot pressed and examined.

Fueled Zirconium Hydride Moderator

H. E. Bigony and J. B. Vetrano

The apparatus designed for use in measuring hydrogen-sorption isotherms on a zirconium-1 w/o uranium alloy and a similar but less extensive preliminary study on a zirconium-50 w/o uranium alloy, referred to in BMI-1220, is not yet completed. Major components are ready for assembly and will be assembled during the coming period.

Samples of both alloys were submitted for vacuum-fusion analysis to determine the oxygen, hydrogen, and nitrogen content. A spectrographic analysis is also being made.

It is known that silicon contamination of uranium-zirconium alloys will occur during hydriding in a Mullite chamber if specimens are not adequately protected. This leads to erroneous results in dissociation-pressure measurements. Recent information, however, indicates that it may be possible to protect specimens against silicon contamination, if hydriding is carried out in a Mullite chamber, by wrapping the samples in Nichrome foil.
Inconel will be investigated to serve as a reaction chamber in the preliminary work on the zirconium-50 w/o uranium alloy along with Mullite to evaluate the two materials before making a choice for work on the zirconium-1 w/o uranium alloy.
F-1

F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS

F. A. Rough and A. W. Hare

Various uranium-containing fuels are being studied under sponsorship of the Division of Reactor Development.

A temperature-controlled irradiation-test capsule, BMI-8-5, which contains two specimens of uranium-50 w/o zirconium alloy, has now operated successfully in the MTR for one complete cycle. This test is continuing.

Ternary alloys of uranium containing about 23 w/o zirconium and 5 w/o niobium have been found to exhibit exceptional corrosion resistance in 680 F water.

Current plans for the work on the preparation of uranium-niobium alloys are reported.

Data are reported on the uranium corrosion rates in boiling aqueous solutions of various electrolytes.

Radiation Stability of Uranium-Zirconium Alloys

A. W. Hare, A. E. Austin, A. A. Bauer, and R. F. Dickerson

Studies of irradiation effects on uranium-zirconium alloys are being continued. A temperature-controlled capsule, designated as BMI-8-5, which contains two specimens of uranium-50 w/o zirconium alloy, has now been operating satisfactorily in the MTR A-39NW test hole for one complete cycle. Specimens are being irradiated in a thermal-neutron flux of from 0.5 to 1.0 x 10^{14} neutrons/(cm^2)(sec) at a core temperature of 675 C.

A noncontrolled capsule which contains three specimens of zirconium-22 w/o uranium alloys, each given one of three different heat treatments, has been shipped to the MTR and is now awaiting available test space. It is believed that this capsule will be inserted in the MTR in October.

Preliminary work has been initiated on a literature survey for the purpose of summarizing all of the work to date on the uranium-zirconium alloy system. It is planned that the report will be a follow-up of BMI-1030. Kinetics of transformation, mechanical properties, corrosion studies, and irradiation effects will be the various areas covered in this report on the uranium-zirconium alloy system.
Development of Gamma-Phase Uranium Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

The investigation aimed at developing a corrosion-resistant, gamma-phase alloy is continuing. Ternary and quaternary alloys of uranium and chromium, molybdenum, niobium, ruthenium, vanadium, and zirconium are being studied for this purpose.

A preliminary examination of these alloys is being conducted in an attempt to establish the trend an alloying addition has in stabilizing the gamma phase or improving corrosion resistance. Alloys have been cast and fabricated and data should be available shortly from tests now under way.

Corrosion data have been obtained on a series of alloys prepared for a prerunner of this program where the objective was the development of a corrosion-resistant 70 w/o uranium alloy. Alloys containing 23.2 w/o zirconium and 4.84 w/o niobium and 23.8 w/o zirconium and 6.1 w/o niobium have been found to exhibit exceptional corrosion resistance for a uranium-zirconium-base alloy. Alloy specimens of these compositions have been on test for 408 hr, bare, in 680 F water and include specimens which were air cooled from 800 and 900 C and specimens which were subsequently annealed 24 hr at 500 C. In the latter condition, the alloys exhibit corrosion rates of 0.10 and 0.040 mg/(cm^2)(hr), respectively. Uranium-50 w/o zirconium specimens normally exhibit rates on the order of 0.20 mg/(cm^2)(hr).

Clad and artificially defected specimens of these alloys have not stood up as well to corrosion in 680 F water although it is uncertain whether this is occasioned by bond-line attack or initially poor bonding. However, several specimens in the as-clad and gamma-quenched conditions have been on test for 408 hr with only a blister approximately 1/8 in. in diameter appearing at the defect. Binary uranium-26.6 w/o zirconium specimens, isothermally transformed at 500 C for 10 and 100 sec, have been on test for a similar period of time with similar results. Additional tests of these alloys are planned.

Preparation of Uranium-Niobium Alloys

N. E. Daniel, E. L. Foster, and R. F. Dickerson

Although the entire uranium-niobium alloy system has not been extensively investigated, the work done to date indicates that a number of alloys have possibilities as reactor fuels. Extensive investigations of the binary alloys in the range of niobium content below 15 w/o have been and are currently in progress; however, there are relatively few available data on alloys containing niobium in amounts above 15 w/o. Because of the attractiveness of the system and the lack of a complete knowledge of the alloys in the higher niobium range, it would be desirable to perform a comprehensive study of the entire system. Such a study would be coordinated in a way that data obtained as a result of other programs would be incorporated, thus eliminating duplication of effort.
F-3 and F-4

The initial phase of this over-all program consists of a comprehensive literature survey. The objective of this survey is to collect all available data on the alloy system, evaluate this information, and plan an experimental program which will be of maximum value. This survey is nearing completion. In addition, work has been initiated on a study concerned with the preparation of high-purity uranium-niobium alloys.

The consumable-electrode arc-melting technique has been chosen as the best method of producing alloys of the desired purity. Since the problems connected with melting alloys containing 10 w/o niobium are known and since little is known about the difficulties that may be encountered in the preparation of relatively large ingots of alloys containing higher niobium contents, it has been decided to study the melting characteristics of alloys containing 20 and 80 w/o niobium. These alloys have been cast and they will be carefully evaluated by analytical, radiographic, and metallographic techniques. The results of these examinations will guide the future direction of the melting study.

Since high-purity alloys are desirable for a study of the type discussed in previous paragraphs, the purity of alloy ingredients is of utmost importance. In view of this fact, niobium of the various grades that are available from commercial suppliers is being evaluated. It is hoped that, as a result of this evaluation, a source of supply for high-purity niobium will be determined.

Mechanism of Aqueous Corrosion

J. B. Schroeder, C. M. Schwartz, W. E. Berry, P. D. Miller, and F. W. Fink

The mechanism of aqueous uranium corrosion is being studied to aid in the development of a corrosion-resistant fuel alloy.

Some additional samples have been run in various electrolytes to select a suitable electrolyte for charging experiments. From the data listed in Table F-1, and those reported in BMI-1220, sodium acetate, sodium sulfate, and acetic acid appear suitable. The effect of charging will be studied using sodium acetate and sulfate solutions.

<table>
<thead>
<tr>
<th>Electrolyte</th>
<th>Duration of Test, hr</th>
<th>Corrosion Rate, mg/(cm²/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01 N CH₃COOH</td>
<td>24</td>
<td>2.55</td>
</tr>
<tr>
<td>0.01 N CH₃COOH</td>
<td>24</td>
<td>2.71</td>
</tr>
<tr>
<td>0.01 N CH₃COOH</td>
<td>24</td>
<td>2.74</td>
</tr>
<tr>
<td>0.01 N CH₃COOH</td>
<td>24</td>
<td>3.26</td>
</tr>
<tr>
<td>0.05 N Na₂SO₄</td>
<td>24</td>
<td>2.50</td>
</tr>
<tr>
<td>0.05 N Na₂SO₄</td>
<td>24</td>
<td>2.50</td>
</tr>
</tbody>
</table>

Plans are being made to study the nature and composition of the corrosion films formed at temperatures above 100 C in the presence of water.
The objectives of this program are to obtain basic fatigue information on Inconel and also to establish a quantitative relationship between the variables of temperature, stress, strain, time, and cyclic frequency for Inconel which is subjected to repeated stressing.

In the current phase of this program, stress-lifetime data are being obtained for Inconel at 1600 F. Lifetime is defined for several criteria which include 0.1, 0.2, 0.5, 1, and 5 per cent strain and fracture.

During September a series of tests was run at a load ratio of 0.25 and a cyclic frequency of 10 cps. Tests have now been run at the four load ratios selected (A = 0.25, 0.7, 2.0, and $\infty$) and at the two frequencies of 1 and 10 cps at the temperature of 1600 F. A preliminary analysis of these eight series of tests discloses where additional tests are needed at the 1600 F temperature. It is expected that testing at 1600 F will be completed during October. At the conclusion of testing, all data will be reported.

Along with analysis of data at 1600 F it is expected that testing will gradually be shifted to the 1200 F level for the next several months.
Various programs of study supported by the AEC Division of Reactor Development are reported in this section.

Data on the oxidation resistance of niobium-vanadium, niobium-molybdenum, and niobium-zirconium alloys are reported.

Experimental work is being continued on the investigation of valence effects in solid solutions in an attempt to confirm the operative mechanism suggested in the literature as responsible for the formation of "anomalous" solid solutions.

The preliminary design of apparatus for the testing of rubbing surfaces in a sodium environment is well under way, and mock-ups of individual components of the system are now being checked.

It is reported that in the analytical studies of pressure bonding, considerations are being given to the effects of deformation of the surface of contact under pressure. Sample specimens are now being fabricated by various methods to attempt to determine satisfactory welding and sealing techniques for the aluminum specimens which are presently being used for the experimental studies of pressure bonding.

Oxidation-Resistant Niobium Alloys

W. D. Klopp, C. T. Sims, and R. I. Jaffee

This program is a study of some of the factors which determine the oxidation behavior of niobium and niobium-base alloys. Current work includes microexamination of scales formed on niobium alloys, a study of the microstructures of niobium-alloy strips, and the preparation of new alloy samples for oxidation testing.

Metallographic examinations were made of oxidized niobium-vanadium, molybdenum, and zirconium alloys. Scales on the niobium-5 to 12.5 a/o vanadium alloys spalled off on cooling from the test temperature, making examination difficult, but small pieces of gray adherent scale showed no evidence of more than one scale phase. Porosity was noted in the outer portions of the scales. However, no thickness measurements could be made on these specimens. The niobium-25 a/o vanadium scale was thick, porous, and acicular. This sample had oxidized rapidly and completely. The reaction may have been accelerated by the presence of a small amount of liquid $V_2O_5$ which acted as a flux. Niobium-2.5 to 7.5 a/o molybdenum alloys were similar to the vanadium alloys in that most of the scales had spalled off and the remaining adherent scale appeared single phase. The niobium-10 a/o molybdenum scale adhered. It was a single phase although a solid subscale 1.1 mils thick could be distinguished from the porous 9-mil outer scale. The solid subscale may have been forming during the first 0.6 hr of oxidation, when a parabolic reaction curve was observed. Mechanical breakdown of the solid protective scale beyond a limiting
thickness could account for the linear reaction observed after 0.6 hr, with diffusion of oxygen through the constant subscale limiting the rate of oxidation. The small amount of scale which adhered on the niobium-10 a/o zirconium alloy was single phase and porous. Scales on the niobium-35 and -45 a/o zirconium alloys were also single phased and porous, but adherent. Thicknesses of the scales were 17.8 and 11.5 mils, respectively.

Metallographic examinations were made on rolled and annealed (1 hr at 1500 C) strips of niobium containing binary additions of 10 to 35 a/o titanium, 2.5 to 10 a/o molybdenum, 5 to 25 a/o vanadium, and 10 to 45 a/o zirconium. The alloys with titanium, molybdenum, and vanadium exhibited equiaxed single-phase structures, indicating that these elements are completely soluble in niobium up to at least 35, 10, and 25 a/o, respectively. The niobium-zirconium alloys were single phased up to 25 a/o, while the 35 and 45 a/o zirconium alloys had a eutectoid-type second phase at the grain boundaries. These data are in accord with published phase diagrams, which indicate that molybdenum and vanadium are mutually soluble with niobium, titanium is soluble to about 75 a/o, and alloys containing over 20 a/o zirconium transform from single- to two-phase structures at 600 to 1000 C.

Hardness data, given in Table H-1, on these strips indicate that titanium hardens niobium least, while molybdenum is the most potent hardener on an atomic per cent basis. Vanadium hardens niobium most on a weight per cent basis. Grain-size data indicate that annealed grain sizes increased with increasing titanium and zirconium contents, suggesting that these two elements decrease the recrystallization temperature.

Conversion of the continuous-weighing oxidation apparatus from manual to automatic recording has been discontinued. Reaction rates on new samples are now being measured manually.

Valence Effects of Oxide Solutions in Uranium Dioxide

W. B. Wilson and C. M. Schwartz

The objective of this project is to investigate the mechanism and physical principles involved in the preservation, under oxidizing conditions, of the fluorite structure of UO₂. Normally UO₂ transforms in an oxidizing atmosphere to the more volatile and less dense U₃O₈. It had been suggested that the addition of divalent and trivalent oxides in solid solution might stabilize the fluorite structure by some process of compensation for the valence change of the uranium ion during oxidation.

A recent literature check has shown evidence* of the formation of a fluorite-type structure in "anomalous" solid solutions of uranium oxide containing trivalent cation oxides. These solid solutions, which exist over a broad homogeneity range, were formed from mixed oxides during firing in air at 1200 C.

TABLE H-1. HARDNESS AND GRAIN-SIZE DATA FOR NIOBIUM-ALLOY STRIPS ANNEALED 1 HR AT 1500 C

<table>
<thead>
<tr>
<th>Alloy Composition (Balance Niobium), a/o</th>
<th>Hardness, VHN</th>
<th>Grain Size, mils</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 Ti</td>
<td>127</td>
<td>4.5</td>
</tr>
<tr>
<td>20 Ti</td>
<td>154</td>
<td>5.6</td>
</tr>
<tr>
<td>25 Ti</td>
<td>162</td>
<td>7.7</td>
</tr>
<tr>
<td>30 Ti</td>
<td>160</td>
<td>10.7</td>
</tr>
<tr>
<td>35 Ti</td>
<td>169</td>
<td>13.2</td>
</tr>
<tr>
<td>5 V</td>
<td>143</td>
<td>2.6</td>
</tr>
<tr>
<td>7.5 V</td>
<td>169</td>
<td>3.3</td>
</tr>
<tr>
<td>10 V</td>
<td>191</td>
<td>3.2</td>
</tr>
<tr>
<td>12.5 V</td>
<td>210</td>
<td>2.2</td>
</tr>
<tr>
<td>25 V</td>
<td>314</td>
<td>2.2</td>
</tr>
<tr>
<td>2.5 Mo</td>
<td>126</td>
<td>1.7</td>
</tr>
<tr>
<td>5 Mo</td>
<td>146</td>
<td>1.7</td>
</tr>
<tr>
<td>7.5 Mo</td>
<td>205</td>
<td>1.8</td>
</tr>
<tr>
<td>10 Mo</td>
<td>219</td>
<td>1.3</td>
</tr>
<tr>
<td>10 Zr</td>
<td>199</td>
<td>0.8</td>
</tr>
<tr>
<td>25 Zr</td>
<td>266</td>
<td>1.3</td>
</tr>
<tr>
<td>35 Zr</td>
<td>306</td>
<td>3.3</td>
</tr>
<tr>
<td>45 Zr</td>
<td>302</td>
<td>6.8</td>
</tr>
</tbody>
</table>

The present program is intended to confirm the effect of such additions and to extend the understanding of the physical processes involved. It is planned to study systematically the effects of solute valence and ionic size upon oxide stability, and to obtain information on valence of the uranium ion and on the type of lattice defect. To do this, it will be necessary to prepare a series of fired mixed oxides, selected according to size and valence, determine their phase compositions, and measure oxidation stabilities. Calculated and measured densities will be compared as an indication of type of lattice defect. Electrical-conductivity and thermoelectric-power measurements will be made to obtain information on valence states and type of carrier.

Samples of UO₂ containing varying additions of CaO, BaO, MgO, La₂O₃, and Sc₂O₃ have been or are being prepared. Work has been started on electrical measurements of the completed samples.
High-Temperature High-Pressure Solid-State Studies

W. B. Wilson and C. M. Schwartz

Research is continuing on high-temperature high-pressure solid-state studies. The final stages of construction are now being approached on the first die design. The second "geometrical-advantage" type design will be started in October and will be finalized after the initial testing of the first die has been completed.

Rubbing Surfaces in Sodium Environments

J. W. Kissel, R. W. Endebrock, and W. H. Goldthwaite

A study of factors affecting the nonhydrodynamic friction between sliding surfaces in molten sodium up to 1500 F is under way. This program has as its over-all objective the establishment of engineering data for sliding-surface components in high-temperature sodium-flow systems. Such components may be characterized either by low rubbing speeds (e.g., check-valve pivots and valve-stem guides) or by high rubbing speeds (e.g., face-type seals and journal bearings). Although it is recognized that the significance of parameters probably will not be the same for different speed situations, an investigation of slow-speed phenomena will constitute the initial work in this program. Such an approach has a distinct advantage from the viewpoint of utilizing basically simple laboratory apparatus to probe for effects which may be meaningful even for circumstances involving higher relative speeds of rubbing members.

The experimental rig will permit the sliding of a spherical specimen over a plane, the contacting surfaces being submerged in molten sodium. By carefully measuring force and linear displacement of the moving specimen (which is to be the plane) the frictional behavior of specially selected materials will be obtained under controlled conditions of temperature, atmosphere, speed, and surface treatment. The choice of specimen configuration was dictated by the desire to minimize problems of surface alignment such as other types of configurations might present (for example, edge-loading effects and nonuniform loading conditions caused by nonuniform flatness are known trouble areas for plane-plane specimens).

The preliminary design of the test apparatus for the phase of research outlined above is well under way. To assist in the final design, several mock-ups of components such as the specimen heating system, the specimen pivot and displacement mechanism, and the specimen drive are being checked out in the laboratory. As soon as design details have been completed, construction of the apparatus will start. During the construction period selection of specimen materials will be made, and experimental procedures will be established.
An investigation is being conducted to determine and analyze the phenomena involved in pressure bonding, a technique in which metal surfaces are welded through the application of hydrostatic pressure at elevated temperatures below the solidus.

The present emphasis is on a determination of the conditions under which the surfaces will deform sufficiently to insure the required intimate contact.

**Analytical Studies**

Consideration is being given to the deformations of the surface of contact under pressure. An analysis along the lines of Hertz's well known theory of contact is satisfactory as long as the contacting bodies are fully elastic. It has been determined that inelastic behavior begins at pressures of much smaller order of magnitude than will be required to obtain complete contact. Once yielding begins, deformations will occur at lower pressures than would be required for the same deformation if the material remained elastic. However, the required pressures have not yet been determined.

**Experimental Studies**

This phase of the program is directed toward obtaining experimental data for testing the validity of certain simplified mathematical assumptions which might be used in predicting plastic deformations in pressure bonding. In order to obtain a correlation between the theoretical predictions and the physical phenomena, and to investigate the influence of time, to which no theoretical consideration has been given, specimens are designed, fabricated, and pressure bonded under various conditions as prescribed by the analytical studies.

Samples have been fabricated by various methods to attempt to determine satisfactory welding and sealing techniques for the aluminum specimens presently being used in the investigation. Procedures that produced specimens which contained no leaks at room temperature were used to prepare several specimens for pressure bonding. Two specimens pressurized at 500 F and 4000 psi for 3 hr developed leaks in the evacuation tubes after considerable deformation of the specimens had taken place. Measurements of the specimens indicate that the size and geometry used are feasible for the intended study, since the hole remaining in the specimen after deformation is approximately circular in cross section, and since the increased resistance to deformation caused by the welds at the ends of the specimens extends only a short distance from the ends. However, the partial failure of the specimens prevented the data from being used to test the theoretical calculations. An additional specimen has been sealed using modified techniques and was pressurized under the same conditions. This specimen will be evaluated to determine if the modified sealing technique is suitable for preparing specimens for this investigation.
I. PHYSICAL RESEARCH

F. A. Rough

Various programs of physical research in metallurgy and ceramics are reported in this section. These programs are supported by the AEC Division of Research.

In the study of bonding fundamentals, emphasis is presently directed toward understanding the effects of bonding load and the strain energy of cold working in the bonding process.

In the study of physical properties of uranium compounds, progress is reported for the preparation of samples of UBe$_{13}$, UC and UC$_2$, and UB$_2$. Data are reported for the reaction of UC$_2$ with oxygen and nitrogen. A report of progress on the measurement of heats of formation of uranium compounds is also included.

Initial rate experiments on the reaction of niobium and hydrogen to produce hydrogen-niobium ratios of 0.05, 0.10, and 0.50 have begun. The surface preparation and experimental conditions are very important in this work, and it is found that annealing of the specimens at 800°C retards the reaction apparently by formation of a thin oxygen film. Further tests on unannealed samples are being carried out.

Research on various other programs is continuing, but report sections are not included this month. These programs include studies of the constitution of uranium alloys, the solid-state characteristics of UNC, the structure of UC$_2$, and the migration of hydrogen in zirconium hydride.

Study of Bonding Fundamentals

F. C. Holden and J. B. Melehan

The purpose of this research is to express the phenomenon of solid-state bonding in terms of material physical properties and experimental conditions. Experiments are conducted on a simulated single-asperity bonding system employing a gold needle with a hemispherical point bonded to a flat gold surface. The normal force required to rupture the bond has been selected as the principal measure of degree of bonding. Results obtained from a systematic study of bonding conditions (temperature, time, and pressure) suggest that a power relation exists between bond strength and bonding time at constant temperature. The general form of the relation is

\[ F_t = F_0 + K(T)t^n, \]

where \( F_t \) and \( F_0 \) are, respectively, the force to rupture the bond at time \( t \) and at time 0. \( K \) is a temperature-dependent quantity, and \( n \) is a constant with an experimental range of about 0.3 to 0.4. This equation can be expressed in terms of the theoretical expression derived by Kuczynski* for the sintering of small spherical particles to flat spheres.

---

surfaces. This relation expresses contact interface dimensions as a function of temperature, time, and several physical properties as follows:

\[
x^5 = \left( \frac{40\sigma \delta^3 a^2 D_v t}{KT} \right),
\]

where

- \( x = \text{radius of bonded area} \)
- \( \sigma = \text{surface tension} \)
- \( \delta = \text{interatomic distance} \)
- \( a = \text{radius of contacting sphere} \)
- \( D_v = \text{coefficient of volume self-diffusion} \)
- \( t = \text{time} \)
- \( K = \text{Boltzmann's constant} \)
- \( T = \text{temperature, } K. \)

Assuming that the breaking force is proportional to the bonded area, \( \pi x^2 \), and combining constants, the expression for bond strength becomes

\[
F = F_0 + S\pi \left( \frac{40\sigma \delta^3 a^2 D_v}{KT} \right)^{0.4} t^{0.4},
\]

in which \( S \) is the tensile strength of gold at the temperature of testing. Thus, the empirical constant \( K(T) \) is represented by the term

\[
s\pi \left( \frac{40\sigma \delta^3 a^2 D_v}{KT} \right)^{0.4}.
\]

The experimental values for the exponent \( n \) are in reasonable agreement with the predicted value of 0.4. The value of \( K(T) \), calculated from the expression

\[
s\pi \left( \frac{40\sigma \delta^3 a^2 D_v}{KT} \right)^{0.4},
\]

is not in agreement with experimental results, but is several orders of magnitude too low. A study was initiated to determine the probable cause for this discrepancy. In the development of the Kuczynski equation, a number of assumptions were made which do not apply to the experimental conditions used in the present work. These are listed as follows:
The Kuczynski equation was derived for the case of a sphere in contact with a flat surface without an applied load; in the present work, the needle tip is forced into contact with the flat, so that an initial area of contact exists.

A number of simplifying assumptions were made in the Kuczynski development regarding the geometry of the section at contact; these assumptions become less valid as the area of contact is increased.

Although the tip of the needle is approximately spherical, the actual cross section is closer to a parabola, whereas the Kuczynski equation was developed for a sphere.

The effect of deformation on the diffusion coefficient, \( D_v \), has not yet been evaluated. It is quite possible that a considerable increase in \( D_v \) may result from the initial deformation of the system.

Modification of the equation to include the differences in geometry have not shown an appreciable improvement in the value of \( K(T) \). At the present time, it appears that the most promising approach to this problem lies in evaluating the effects of strain on the diffusion coefficient. Efforts are being continued along these lines.

Experiments are in progress to study the effects of applied bonding load and the strain energy of cold working on the bonding process. Bond-strength data obtained under varying conditions of applied load are presented in Table I-1. Bond-strength values increase as the applied and maintained load is increased. When the results of these tests are plotted as curves of breaking strength versus time, it appears that the increase in applied load has only a transient effect on the bond growth. That is, at longer times, the strength-versus-time curves for different applied loads are approximately parallel.

**TABLE I-1. NORMAL BREAKING FORCE OF SINGLE-ASPERITY GOLD CONTACTS UNDER VARIOUS APPLIED LOADING CONDITIONS AT 700 C**

<table>
<thead>
<tr>
<th>Applied Bonding Force, g</th>
<th>Breaking Force (a), g, at Indicated Time, min</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>100</td>
<td>50</td>
</tr>
<tr>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>200</td>
<td>0</td>
</tr>
</tbody>
</table>

(a) Breaking force measured at bonding temperature.
A similar transient effect was observed in previous experiments on the time
dependence of the hot hardness of gold. From the hot-hardness experiments, it was
observed that the area of the hardness impression could be expressed as the following
function of time:

\[ A_t = A_0 (1 + ct^n), \]

where \(0.12 < n < 0.16\).

An investigation has been initiated to study the effect of strain energy of cold work
on bonding behavior. It was necessary to modify the testing procedure for this study
because of the high rate of recrystallization of cold-worked gold. At 400, 600, and
800°C, recrystallization is complete in 1 min, 15 sec, and 5 sec, respectively. Speci-
men contact and recrystallization are made to occur simultaneously by establishing
contact immediately before heating begins. In order to minimize the heating interval
between room temperature and the testing temperature, the specimen size (conse-
quently the heat capacity) was reduced and a furnace was constructed for rapid heating
rates; e.g., 25 to 600°C in less than 10 sec. Although the effectiveness of the changes
in experimental techniques has not been definitely established, preliminary test results
are reported in Table I-2 for experiments using work-hardened needles and flat speci-
mens. The results are compared with those for annealed specimens. More complete
data will be required before an evaluation is possible; however, at 600°C the bond
strengths for annealed specimens are slightly lower than bond strengths of the cold-
worked specimens. At 400°C there is no apparent difference between the annealed and
cold-worked materials.

TABLE I-2. NORMAL BREAKING FORCE OF SINGLE-ASPERITY GOLD CONTACTS
DETERMINED FOR ANNEALED AND COLD-WORKED SPECIMENS

<table>
<thead>
<tr>
<th>Temperature, °C</th>
<th>Specimen Condition</th>
<th>0</th>
<th>2</th>
<th>5</th>
<th>10</th>
<th>20</th>
<th>40</th>
<th>60</th>
</tr>
</thead>
<tbody>
<tr>
<td>400</td>
<td>Cold worked(b)</td>
<td>0.18</td>
<td>0.36</td>
<td>0.30</td>
<td>0.29</td>
<td>0.42</td>
<td>0.73</td>
<td>1.02</td>
</tr>
<tr>
<td>400</td>
<td>Annealed(c)</td>
<td>--</td>
<td>0.30</td>
<td>0.28</td>
<td>0.42</td>
<td>0.45</td>
<td>0.57</td>
<td>0.70</td>
</tr>
<tr>
<td>600</td>
<td>Cold worked(b)</td>
<td>0.17</td>
<td>0.28</td>
<td>0.30</td>
<td>--</td>
<td>0.47</td>
<td>0.70</td>
<td>0.78</td>
</tr>
<tr>
<td>600</td>
<td>Annealed(d)</td>
<td>0.20</td>
<td>--</td>
<td>--</td>
<td>0.33</td>
<td>0.45</td>
<td>0.57</td>
<td>0.66</td>
</tr>
</tbody>
</table>

(a) Breaking force measured at bonding temperature.
(b) Needle in as-received condition; flat cold rolled to 90 per cent reduction.
(c) Needle and flat annealed for 5 min at 800°C.
(d) Needle in as-received condition; flat annealed 1 hr at 1000°C.

Efforts will continue toward determining the effects of bonding load and energy of
cold work on bonding. Continued analysis of the Kuczynski expression will be made in
an attempt to resolve the present disagreement between theoretical and experimental
values.
Preparation, Fabrication, and Physical Properties of Uranium Compounds

A. B. Tripler, Jr., M. J. Snyder, and W. H. Duckworth

Methods of preparation and fundamental properties of some refractory uranium compounds which are potential nuclear fuels are being investigated.

Work was continued on the preparation of UBe$_{13}$ by heating UH$_3$ and beryllium powder to 1600 C in an argon atmosphere. Chemical analysis of the specimen prepared in August in which the starting materials were mixed in the stoichiometric proportions to form UBe$_{14}$ showed an excess of beryllium. The specimen had a bulk density which was 90 per cent of theoretical. Hardness measurements showed the sintered specimen to have an average Knoop hardness of 1188. In an attempt to obtain a purer sample, the center portion of the specimen, which was the soundest, was ground to minus 100 mesh, the powder was compacted, and the compact was resintered. The bulk density of the resintered compact was 110 per cent of theoretical, indicating loss of beryllium on the second sintering.

The sound, vacuum-sintered specimen of UC$_2$, the preparation of which was described in BMI-1220, was examined metallographically. The material was mostly UC$_2$. A small, but unestimated, amount of UC was present, and also some other material, possibly U$_2$C$_3$. No free carbon was observed. The porosity was estimated to be about 10 per cent, which checks well with the bulk density which was 89 per cent of theoretical. Gas kinetic studies were made on the sound UC$_2$ specimen, and the results are described in the following section of this report.

Metallographically homogeneous samples of UC and UC$_2$ were prepared by arc melting. They had bulk densities of 104 per cent and 96.5 per cent of theoretical, respectively. The UC had a Knoop hardness of 857 and the UC$_2$ had a Knoop hardness of 618 (both with 100-g loads). Gas reaction studies will be made on these specimens in October.

UB$_2$ was prepared by solid-solid reaction of high-purity boron and uranium hydride in a vacuum furnace at 1300 C. The resultant specimen had a bulk density which was 60 per cent of theoretical. This result duplicated that of previous experiments, and it was concluded that this is the highest density obtainable by the method used. In order to densify the material, the specimen was melted in an arc-melt furnace. The material so obtained had a bulk density which was 99.6 per cent of theoretical. Metallographic analysis showed UB$_2$ to be present with a uranium-rich phase along the grain boundaries. There also was a small amount of what appeared to be a eutectic phase. A eutectic has been reported between uranium and UB$_2$. The combination of solid-solid reaction, sintering, and arc melting will be repeated using an amount of boron somewhat higher than the stoichiometric amount.

The preparation of the vacuum-induction furnace which is to be used in making thermal-conductivity specimens has been delayed by material failures.
Reaction With Gases

W. M. Albrecht and B. G. Koehl

A study is being made of the reactions of certain uranium compounds with nitrogen, oxygen, and water vapor.

Initial reaction-rate experiments were made with the sound compact of UC$_2$ described in the previous section of this report. Rate data were obtained with nitrogen in the range 400 to 700°C and with oxygen in the range 150 to 300°C. For reaction times of 3 to 4 hr the rates followed very nearly a parabolic law,

$$w^2 = kt,$$

where

- $w =$ weight gain, g per cm$^2$
- $k =$ rate constant, $\text{(g/cm}^2\text{)}^2$/sec
- $t =$ time, sec.

For parabolic behavior, a plot of the logarithm of $w$ against the logarithm of $t$ is a straight line with a slope of 0.5. The values of the parabolic rate constants and the log-log slopes are given in Table 1-3. The data show that the reaction of oxygen with the sintered UC$_2$ is more rapid than the nitrogen reaction at the same temperature. UC$_2$ reacted with nitrogen at 600°C at about the same rate as with oxygen at 250°C. At 300°C the oxygen reaction proceeded anisothermally with the temperature increasing rapidly.

<table>
<thead>
<tr>
<th>Temperature, C</th>
<th>Slope of Log-Log Plot</th>
<th>$k$, \text{(g/cm}^2\text{)}^2$/sec</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Nitrogen</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>400</td>
<td>0.46</td>
<td>16.1</td>
</tr>
<tr>
<td>600</td>
<td>0.55</td>
<td>856</td>
</tr>
<tr>
<td>700</td>
<td>0.51</td>
<td>3330</td>
</tr>
<tr>
<td><strong>Oxygen</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>150</td>
<td>0.61</td>
<td>6.09</td>
</tr>
<tr>
<td>200</td>
<td>0.58</td>
<td>75.4</td>
</tr>
<tr>
<td>250</td>
<td>0.52</td>
<td>897</td>
</tr>
<tr>
<td>300\text{(a)}</td>
<td>--</td>
<td>--</td>
</tr>
</tbody>
</table>

(a) Sample reacted anisothermally.
Equations for the variation of the rate constant with temperature were obtained from semilog plots of rate constant versus temperature. The equations for the best straight lines through the points are, for the nitrogen reaction,

\[ k = 5.27 \times 10^8 \exp\left(-\frac{23,130}{RT}\right) \]

and for the oxygen reaction,

\[ k = 5.68 \times 10^{11} \exp\left(-\frac{21,180}{RT}\right) \]

The energies of activation are 23,130 and 21,180 cal per mole for the nitrogen and oxygen reactions, respectively.

It was found that specimens having been reacted with nitrogen crumbled when exposed to air. X-ray analysis showed only the presence of UC2 and UO2 in these specimens. Apparently the freshly formed nitride reacted with the air to produce UO2. Specimens not exposed to air did not crumble and X-ray analysis showed the presence of UC2 and UNx, where x is probably greater than 1.5.

X-ray analysis of specimens reacted with oxygen showed the presence of UC and UO2 as reaction products.

Further rate data are being obtained for the reaction of UC2 with water vapor.

Heat of Formation Measurements


Heats of formation of refractory uranium compounds are being determined as part of the program on evaluating these materials. Heats of formation are being determined from measured heats of combustion of the compounds and known heats of combustion of the constituent elements.

Two approaches have been followed in attempting these measurements. An oxygen-bomb calorimeter has been used for the combustion of the more readily burned compounds such as UC. A sodium peroxide bomb has been used with compounds such as the silicides where combustion in oxygen leads to the formation of fused SiO2, which prevents complete combustion. Although some success has been achieved with both techniques, no final numbers have been obtained. The difficulties encountered mainly result from the extreme accuracy required because of the fact that the heat of combustion of the compounds is not greatly different from the heat of combustion of the constituent elements.

In one part of the program, attempts have been made to use a sodium peroxide calorimeter to find the heat of combustion of USi2 using benzoic acid as a combustion aid. Results on the combustion of benzoic acid alone scattered by about 0.1 per cent. This degree of precision was not attained with USi2, however. Two related difficulties, incomplete combustion and inability to analyze for unreacted sample, plagued the experiments.
The normal procedure was as follows: A USi₂ sample was mixed with about 13 g of Na₂O₂, a small amount of benzoic acid as a combustion aid, and KCIO₄ as a "promoter". The mixture was placed in a capped, nickel fusion cup and was fired, in the calorimeter, by an electrically heated fuse wire.

With this procedure, the mixture remained molten for about 2 min. Apparently, some of the heavy USi₂ sank to the bottom of the Na₂O₂ bath and was there cooled by the massive nickel cup sufficiently to prevent complete reaction. At the completion of the experiment, the fused mass was dissolved in water and the unreacted sample was filtered out and weighed. Reaction was usually about 95 per cent complete. Since the reaction continued in the presence of water, however, the exact amount of unreacted sample could not be accurately determined.

One attempt was made to contain the mixture in a liner fashioned from nickel foil inside the fusion cup so that the fused mass could be removed from the nickel cup. In this experiment, however, the foil became so hot that it was destroyed by the peroxide mix. Thus it seems that, to avoid excessive attack, the nickel must be kept cool; but this prevents the reaction from going to completion. Further work with the peroxide fusion calorimeter has been suspended pending the development of a method for removing and analyzing the product or for conducting the reaction in a thin, attack-resistant container.

The heat of combustion of UC is being determined in an oxygen-bomb calorimeter. The combustion of UC in 30 atm of oxygen produces enough heat to fuse the combustion product, U₃O₈, to sublime some of it to the walls of the bomb, and to melt the platinum crucible containing the sample. In order to reduce the temperature of the combustion, a procedure has been developed whereby the sample is mixed with ten times its weight of UO₂, and the oxygen pressure is 10 rather than 30 atm. In this way the sample can be contained in a platinum crucible and the product recovered and weighed.

At one stage in the development of a workable procedure, the sample was contained in a Mylar bag to protect it from oxidation prior to the ignition. Also, a little water was placed inside the bomb to absorb any HNO₃ formed from the trace of nitrogen in the bomb. At the lower oxygen pressure, however, no HNO₃ was formed, so that the presence of water was not necessary. Furthermore, the heat of adsorption of water on the U₃O₈ product constituted an unknown source of error. With the Mylar bag technique, difficulties were also experienced in accounting for the small changes in weight of the product upon ignition to U₃O₈ at 800 °C. The amount of unburned sample and the exact formula of the product must be known very accurately. For these reasons it seemed essential to eliminate all traces of water from the bomb process, including the water-producing Mylar and cotton-thread fuse. Experiments to this end are currently in progress. Although elimination of the Mylar left the sample unprotected from oxygen, it has been found that under the conditions of the experiment very little oxidation takes place prior to combustion.

With the difficulties described, the precision of the UC combustion has not been much better than 1 per cent. However, the precision of four calibration experiments with benzoic acid was about 0.04 per cent.

Combustion experiments with UO₂-UC mixtures and with UO₂ will be continued.
Niobium-Hydrogen Reactions

W. M. Albrecht and W. D. Goode

A fundamental investigation of the kinetics and mechanisms of the reaction of hydrogen with niobium is being made. The kinetics of the formation of solid solutions of known hydrogen compositions are being determined on the basis of the niobium-hydrogen equilibria previously reported. Initial rate experiments are being made at the equilibrium temperatures and pressures to produce hydrogen-niobium ratios of 0.05, 0.10, and 0.50.

The experiments were made using dry-abraded niobium samples. A sample was placed in the reaction chamber of a modified Sieverts apparatus and vacuum annealed for 1 hr at 800°C at a pressure of 1 x 10^-5 mm of mercury. The sample was then cooled to the temperature of the run. Hydrogen was added to the reaction chamber and rate data were obtained. In all cases, the reactions were initially linear and stopped when the equilibrium compositions were attained. Typical rate curves obtained for annealed samples at 500 and 600°C to produce a composition with hydrogen-niobium ratio of 0.1 are shown in Figure I-1. The initial, linear reaction rates of hydrogen with the annealed niobium are given in Table I-4. In general, it can be seen that the rates obtained under the same experimental conditions are in poor agreement. It was believed that the inconsistencies in these experiments resulted from the presence of very thin oxide films on the niobium samples. Probably the oxide formed on the niobium during the vacuum annealing, even at a pressure of 10^-5 mm of mercury.

**TABLE I-4. SUMMARY OF LINEAR RATE CONSTANTS FOR THE REACTION OF HYDROGEN WITH NIOBIUM**

<table>
<thead>
<tr>
<th>Temperature, C</th>
<th>Composition, Hydrogen-Niobium Ratio</th>
<th>Linear Rate Constants for Indicated Pretreatments</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Annealed at 800°C</td>
</tr>
<tr>
<td>800</td>
<td>0.05</td>
<td>9.2 x 10^-2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.9 x 10^-2</td>
</tr>
<tr>
<td>700</td>
<td>0.05</td>
<td>3.3 x 10^-2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.8 x 10^-2</td>
</tr>
<tr>
<td>600</td>
<td>0.05</td>
<td>7.3 x 10^-3</td>
</tr>
<tr>
<td></td>
<td>0.1</td>
<td>6.7 x 10^-3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8.7 x 10^-3</td>
</tr>
<tr>
<td>550</td>
<td>0.1</td>
<td>7.1 x 10^-3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.4 x 10^-2</td>
</tr>
<tr>
<td>500</td>
<td>0.1</td>
<td>1.3 x 10^-3</td>
</tr>
<tr>
<td>450</td>
<td>0.5</td>
<td>3.8 x 10^-3</td>
</tr>
</tbody>
</table>
FIGURE I-1. REPRESENTATIVE RATE CURVES FOR THE REACTION OF HYDROGEN WITH NIOBium TO PRODUCE HYDROGEN-NIOBIUM RATIO OF 0.1
A second group of rate experiments was then made in which the vacuum anneal at 800 °C was omitted. Results of typical rate experiments at 500 and 600 °C to produce a composition with hydrogen-niobium ratio of 0.1 in unannealed samples are also shown in Figure I-1. These rate curves are of the same general shape as those obtained for annealed samples; however, the initial linear rates are about four to ten times faster for the unannealed samples. Linear rate constants for the reaction of hydrogen with unannealed samples are also shown in Table I-4. Additional data are being obtained with unannealed samples.

Also, a high-temperature X-ray study is being initiated to identify and establish the structures of the phases present in the niobium-hydrogen system.
J. CORROSION PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS

C. L. Peterson, P. D. Miller, E. F. Stephan, E. L. White, W. D. Beasley, T. E. Snoddy, and F. W. Fink

The corrosion problems associated with some of the various processes for the recovery of uranium from spent reactor fuel elements are being investigated.

Molybdenum, platinum, and gold show good resistance during short-term exposure to the conditions of the liquid-phase (boiling AlCl₃-NH₄Cl) Zircex hydrochlorination. Both Type S-816 and Haynes 25 look promising after 55 cycles in the cyclic test for the evaluation of materials for the Zircex vapor-phase hydrochlorination.

Titanium is still performing well in evaluation tests for the Darex process. However, the buildup of a hard scale, apparently resulting from the dissolution of stainless steel, has been observed in the dissolver. This might create a problem in actual operation.

Difficulty is still being experienced in operating tests in which HF is passed through molten ZrF₄-NaF. The one test which has been completed showed a corrosion rate of 21 mils per month for an Inconel specimen.

The Zircex Process

In the Zircex process, uranium is separated from zirconium by making use of the volatility of zirconium tetrachloride. After the fuel element has been hydrochlorinated, the chloride is stripped from the uranium solution by means of concentrated nitric acid and the uranium is recovered by conventional solvent extraction. Presently two routes (liquid phase and vapor phase) are being considered for the hydrochlorination step.

The screening of materials for use in the construction of a container vessel for liquid-phase hydrochlorination has continued. The metal specimens are exposed both submerged and in the vapors above the boiling double salt, AlCl₃-NH₄Cl while HCl is passed over the surface of the liquid. The exposure period has been set for 24 hr during the initial screening tests. The corrosion rate of the specimen submerged in the liquid is usually greater than that of the vapor-phase specimen by at least tenfold. Of the metals tested, platinum, gold, and molybdenum show good resistance; Hastelloy B, C, and W, and Type S-816 have rates of 10 to 20 mils per month, and the Haynes alloys, 25, 30, and 36 are in the range of 40 to 50 mils per month in the liquid. Several metals remain to be screened for this particular application.

The cyclic tests, designed to evaluate materials of construction for a vapor-phase hydrochlorinator-dissolver, are under way. Both Haynes 25 and Type S-816 have low corrosion rates even after 55 cycles in 3 and 5 M HNO₃ solutions. The corrosion rates in 15 M HNO₃ solutions are steadily increasing for both Type S-816 and Haynes 25 and have reached the values of 8 and 10 mils per 100 cycles, respectively, after 55 cycles. In the solutions of lower HNO₃ concentration, the corrosion rates for Haynes 25
have been lower than those for Type S-816 and have remained at or below 2 mils per 100 cycles. Considering that mils per 100 cycles might be thought of as units of approximately four times the values as mils per month, these are very acceptable corrosion rates.

The exposure of specimens to the hot cycle only has been continued, but no evaluation has been made during the month. Data from tensile specimens exposed to the hot cycle only are also being collected.

**The Darex Process**

In the Darex process, uranium is separated from fuel elements containing stainless steel by solvent extraction of the nitric acid solution which results after the fuel element has been dissolved in a dilute mixture of nitric and hydrochloric acids and the chlorides stripped by concentrated nitric acid.

A total of 2483 hr of exposure of titanium, tantalum, and Haynes 21 has been completed in the presence of dissolving stainless steel in a flowing system. No deleterious effect on any of these metals has been noted from the limited examination given the specimens thus far. An appreciable buildup of a hard, glasslike solid occurs on the specimens and on the glass walls of the dissolver. This deposit is quite difficult to dislodge or dissolve and may result in part from the silica content of the stainless steel. Exposure of the specimens mentioned above will be discontinued in favor of exposing various titanium specimens which contain stressed areas, crevices, or weldments.

The exposure of the titanium specimen to boiling dilute nitric and hydrochloric acid solution, while impressed with an external EMF similar to that which it would attain if contacted by dissolving uranium, was discontinued after 2 months. The specimen was covered with a hard, adherent scale. Rough calculations show a corrosion rate of about 1.7 mils per month. This rate does not seem excessive and it is possible that this coating may offer high resistance to the galvanic currents, thus becoming self-stifling. Further measurements for embrittlement and hydrogen pickup remain to be made.

**Fluoride-Volatility Process**

Fuel elements containing zirconium as an alloying element or cladding are hydrofluorinated in a molten bath of ZrF₄-NaF by a stream of HF as the first step in the fused salt-fluoride volatility process. A program for the evaluation of materials of construction for this process is being carried out.

A 96-hr exposure run was completed using an Inconel specimen. A corrosion rate of 21 mils per month was calculated. The test equipment has not yet been perfected to the point where long-term exposures can be completed without plugging of the lines. Procedures to alleviate the situation are being examined.
K. EVALUATION OF A REFLECTOR-CONTROLLED HETEROGENEOUS BOILING REACTOR

J. W. Chastain

This program is being conducted to investigate the feasibility of controlling a boiling-water reactor system by varying the height of the radial reflector. This work has been carried out in two phases. The first phase consists of a critical experiment to measure reactivity worth of the reflector as a function of position for a variety of core conditions. The second phase is the simulator study which couples a complete mock-up of a hydraulic system to an electronic reactor simulator.

During September, experimental work terminated on both phases of the research. Topical reports are being prepared and will be issued in the near future.

Critical Experiment


The purpose of this experiment was the evaluation of the effectiveness of a reflector system of control in a boiling power reactor. An evaluation of the long- and short-term transient behavior of the reactor system was to be obtained by using the results from the critical assembly as input data in the reactor simulator.

During the month of September, the effect of the boron carbide-impregnated plastic sheet between the inner and outer radial was investigated. With the inner radial reflector removed, the critical mass with the poisoned plastic was 7863 g. For a similar reflector condition without the poisoned sheet the critical mass was 6940 g.

In the final nonuniformly voided case the upper axial reflector was voided to 45 per cent. To obtain a symmetrical reactor condition the upper axial reflector void was reduced to 20 per cent to correspond to that of the lower axial reflector. No measurable change in critical mass or flux distribution was found.

This research is now concluded and a topical report is being prepared.

Simulator Studies

B. B. Gordon and J. J. Stone

During September, experimental work on this project was completed. The system was operated using different numbers of plates in the damping portion of the system to determine an optimum damping for the system.
A topical report containing a detailed description of the work performed and the results obtained is being prepared.
L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT ELEVATED TEMPERATURES

J. H. Stang

During the coming year, the research being conducted in association with LAMPRE I (the Los Alamos sodium-cooled fast reactor featuring tantalum as the core material) will be along the three lines listed below; during September, emphasis was placed on various planning details pertaining to forthcoming experiments.

1. Flow-loop studies to investigate the strength characteristics of tantalum specimens during exposure in polythermal sodium environments (1200 °F peak temperature for the present) will be initiated. This program will utilize loop apparatus similar to that developed for tantalum-sodium corrosion studies conducted during the past year.

2. Conventional static-creep experiments involving tantalum, with particular attention paid to impurity effects, will be performed. This investigation is essentially an extension of the past year's work in the mechanical-property area.

3. Welding studies to pinpoint factors which lead to weld porosity in tantalum and to devise methods to reduce grain size in tantalum welds will be undertaken. According to past studies, these problems are of prime importance in the tantalum-welding picture.

Major points drawn from experiments in progress during September are listed below.

1. After a 38-day exposure to 1200 °F peak-temperature sodium (oxygen level about 80 ppm), tantalum specimens lost weight corresponding to a maximum of 2 mils per year of removed surface metal. This rate of loss ties in well with past corrosion information.

2. Static-creep tests (helium atmosphere, 1200 °F) of tantalum specimens which had been reduced in oxygen content from about 270 ppm to about 30 ppm by prior exposure to sodium displayed strength nearly the same as displayed by the material before sodium treatment.

Tantalum-Sodium Compatibility Studies

G. E. Raines, C. V. Weaver, and J. H. Stang

As pointed out in BMI-1220, emphasis in the polythermal forced-convection flow-loop studies of the behavior of tantalum in 1200 °F peak-temperature sodium is being
shifted from an investigation of the corrosion problem to an evaluation of mechanical properties exhibited by tantalum during exposure to sodium. The purpose here is to determine (1) changes in creep strength attributable to the slight attack of the material by sodium and the lowering of its oxygen content, which are established effects of sodium exposures on tantalum (provided low-oxygen sodium is the test fluid), and (2) effects of stress on the rate of corrosion attack. The design of the flow-loop systems to be used in the contemplated experiments is now well into its final stage; construction of test assemblies should start in October. These loops will be similar to those developed for the corrosion work, retaining Type 316 stainless steel as the principal material of construction and featuring suitable sodium-purification facilities. However, they will be instrumented to obtain creep data for stressed tantalum specimens immersed in flowing sodium in an isothermal (1200°F) region.

During September, the final experiment (Test 8) now contemplated in the corrosion-test series terminated a 38-day run; the oxygen level of the sodium was approximately 80 ppm. The only data available from this run to date are the specimen weight changes presented in Table L-1. In general, these correlate fairly well with corresponding values from another 80-ppm run (Test 2, 8 days of exposure; data for this exposure were presented in BMI-1220).

<table>
<thead>
<tr>
<th>Specimen Designation</th>
<th>Estimated Average Specimen Exposure Temperature, F</th>
<th>Mg per In.²</th>
<th>Corresponding Mils per Year Removed From Specimen Surfaces</th>
</tr>
</thead>
<tbody>
<tr>
<td>G-9</td>
<td>1200</td>
<td>-56.0</td>
<td>-2.0</td>
</tr>
<tr>
<td>G-1</td>
<td>1200</td>
<td>-45.5</td>
<td>-1.6</td>
</tr>
<tr>
<td>G-7</td>
<td>1150</td>
<td>-39.0</td>
<td>-1.3</td>
</tr>
<tr>
<td>G-2</td>
<td>1100</td>
<td>-22.1</td>
<td>-0.8</td>
</tr>
<tr>
<td>G-5</td>
<td>1050</td>
<td>-7.0</td>
<td>-0.2</td>
</tr>
<tr>
<td>G-8</td>
<td>1000</td>
<td>-0.1</td>
<td>-0.04</td>
</tr>
<tr>
<td>G-6</td>
<td>800</td>
<td>+0.1</td>
<td>+0.03</td>
</tr>
<tr>
<td>G-3</td>
<td>700</td>
<td>+0.05</td>
<td>+0.02</td>
</tr>
</tbody>
</table>

Also, during September, the final inspections of the flow loop systems that have been used were started. This is to obtain information relative to tantalum mass-transfer effects and to the corrosion resistance of the loop material in the particular environment investigated. Information now on hand for one system that had a cumulative exposure of 1400 hr (with <10 ppm sodium) indicates the following:

1. A section of the Type 316 stainless steel pipe from a 1200°F sodium zone displayed slight intergranular attack to about 1/2 mil.

2. Minute deposits, identified as being rich in iron, were noted on the pipe wall in the 1200°F zone. The presence of such deposits, which must have been transferred from the steel, in a peak-temperature zone is somewhat surprising.
(3) Tantalum deposits have not been found although the colder zones of the system (minimum temperature of 700 F) have not been completely examined.

High-Temperature Mechanical Properties of Tantalum

G. F. Huber, C. J. Slunder, J. G. Dunleavy, and A. M. Hall

Creep testing at 1200 F was continued on low-oxygen-content tantalum. The low-oxygen material was obtained by exposing specimens (fabricated from sintered sheet) to an environment of flowing sodium at a peak temperature of 1200 F for 33 days. The sodium was continuously purified by gettering with heated zirconium; the nominal oxygen content of the sodium during the run was <10 ppm.

Table L-2 includes the hardness and analytical values for this material obtained before and after exposure to the sodium, while Table L-3 presents postexposure creep data now on hand for it. It is important to note that the strength, as given in Table L-3, for this low-oxygen material appears to be about the same as that exhibited by the original material; creep rates of about 0.001 and 0.00001 per cent per hr are indicated at stresses of 15,000 and 10,000 psi, respectively. These results suggest that loss of oxygen may not appreciably reduce the creep strength of tantalum when hydrogen and nitrogen are present in the amounts shown in Table L-2. The effects of hydrogen and nitrogen on creep strength will be studied in experiments now in the planning stage.

TABLE L-2. DESCRIPTION OF TANTALUM EMPLOYED IN CREEP EXPERIMENTS COVERED BY TABLE L-3

<table>
<thead>
<tr>
<th>Before Exposure to Sodium</th>
<th>After Exposure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Hardness, VHN (5-kg load)</td>
<td>170</td>
</tr>
<tr>
<td>Oxygen, ppm</td>
<td>260; 285</td>
</tr>
<tr>
<td>Nitrogen, ppm</td>
<td>120; 140</td>
</tr>
<tr>
<td>Hydrogen, ppm</td>
<td>1.6; 1.9</td>
</tr>
</tbody>
</table>

TABLE L-3. CREEP DATA FOR ANNEALED TANTALUM AFTER EXPOSURE TO SODIUM AT 1200 F FOR 33 DAYS

<table>
<thead>
<tr>
<th>Stress, psi</th>
<th>Time in Progress, hr</th>
<th>Time, hr, To Reach Indicated Percentage Deformation</th>
<th>Total Deformation, per cent</th>
<th>Minimum Creep Rate, per cent per hr</th>
</tr>
</thead>
<tbody>
<tr>
<td>15,000</td>
<td>1000</td>
<td>0.1 0.2 0.5 1.0 2 5</td>
<td>3.8</td>
<td>0.0015</td>
</tr>
<tr>
<td>10,000</td>
<td>675</td>
<td>385</td>
<td>-- -- -- -- --</td>
<td>0.12</td>
</tr>
</tbody>
</table>
Weldability of Tantalum for High-Temperature Systems

S. M. Silverstein, R. P. Sopher, and P. J. Rieppel

During September, experimental work was not performed in this research. However, plans were made for future research which will probe into two major problems now known to be associated with the weldability of tantalum. These problems are: (1) porosity in welds in some lots of tantalum, and (2) large grains in the cast structure of weld deposits. Both of these conditions could lead to a service failure of an all-tantalum core such as is contemplated for LAMPRE I.

In order to solve the first problem, it will first be necessary to concentrate the unidentified inclusion material which is common to unweldable lots of tantalum. X-ray diffraction analysis can then be used in attempts to identify the concentrated material. Subsequent to these studies, attempts will be made to determine the maximum amount of this material that can be tolerated without producing porous welds in tantalum.

Two approaches are planned for reducing the grain size in the cast structure of tantalum welds. These are: (1) the use of ultrasonic techniques in conjunction with the presently used argon-shielded tungsten-arc process, and (2) the use of resistance welding. The first approach will be utilized by inducing ultrasonic waves into tantalum which is being welded. It is believed that the energy induced in the tantalum during welding will help prevent excessive grain size on crystallization of the molten weld metal. Both a mechanical- and a liquid-coupling ultrasonic vibration system will be designed and evaluated for effectiveness in coupling high-energy vibrations to weld areas during the tungsten-arc welding process. After the systems are evaluated, welds will be made during ultrasonic excitation and will be compared with normal welds to determine the effectiveness of the technique.

Resistance welding is another process that offers promise for joining tantalum without excessive grain growth in the bond area. It is anticipated that a number of joint designs for resistance welding will be studied. However, since grain growth can be controlled (by accurate time and temperature control and by rapid cooling and/or quenching) it is believed that the process, regardless of the joint design, will produce welds with a finer crystalline structure than is presently produced by conventional tungsten-arc welding.
Further studies have been made of the effect of design changes on mixing in a model of the PWR reactor. Three lower-plenum configurations were studied, two of which improved mixing.

Components for the X-ray photometric detection apparatus have been received and are being assembled.

A study of the pressure bonding of PWR plate-type uranium oxide fuel elements is in progress. Difficulties have been encountered in obtaining consistently good bonds on pickled or vapor-blasted surfaces, although not on machined surfaces. The difficulties appear to be caused by contamination of the surfaces, and various finishing methods are being studied to determine the cause of difficulty. Reaction occurs between uranium oxide and Zircaloy 2, producing a corroducible layer a mil or two thick. Barrier layers for preventing such reaction are being investigated.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Air-flow studies are being conducted in a quarter-scale flow model of the PWR reactor to determine the effect of lower-plenum changes on mixing and flow distribution for Core 2. Past work includes preparation of the model for the present studies, core-flow-distribution studies, and mixing studies involving various geometric changes in the lower plenum. Mixing was not improved by any previous changes.

In September, three more lower-plenum configurations were studied to determine their effect on mixing. For two of the three configurations, mixing was improved substantially. A conical extension of the flow baffle, extending to within 1/4 in. of the bottom dome, was installed. Four openings, 6-1/2 by 2-3/4 in., were placed at the bottom of the extension, midway between the inlets. This change did not improve mixing. The second alteration consisted of installing semicircular flow deflectors around each inlet to direct the flow tangentially around the bottom dome to form a large vortex in the lower plenum. The deflectors were placed 1/2 in. from the edge of each inlet, fastened to the pressure vessel, and contoured to fit the conical surface of the flow baffle. Each deflector was positioned to direct flow directly beneath the adjacent inlet. With this configuration mixing was improved. The maximum amount of fluid from the traced inlet found in any fuel assembly in the core was 61 per cent. The minimum was 6 per cent. Without changes in the lower-plenum geometry, as much as 90 per cent of the fluid in some fuel assemblies came from the traced inlet. With perfect mixing all fuel assemblies would receive 25 per cent of their flow from the traced inlet. The flow deflectors were then repositioned to direct flow horizontally to impinge on the adjacent flow deflector. A further improvement in mixing was found.
The maximum amount of fluid from the traced inlet found in any fuel assembly was 51 per cent and the minimum was 11 per cent.

In October a mixing study will be made with the flow deflectors directed slightly upward to direct flow between the adjacent flow deflector and the bottom of the thermal shields.

Pressure loss and core-flow distribution will then be studied for those configurations in which mixing is improved.

X-Ray Photometric Examination of Fuel Elements

J. B. Schroeder and C. M. Schwartz

A nondestructive production-test apparatus is being developed to detect weld porosity and uranium segregation in alloy fuel elements.

The fine-focus X-ray tube and constant-potential generator have been received and installed. A lathe bed has been ordered to serve as the feed mechanism.

Pressure Bonding of Zircaloy 2-Clad Fuel Elements Containing Compartmented Oxide Fuel Plates

S. J. Paprocki, E. S. Hodge, D. C. Carmichael, P. J. Gripshover, and H. D. Hanes

An investigation is being conducted to determine the feasibility of preparing by pressure bonding a Zircaloy 2-clad uranium dioxide plate-type fuel element, which is being considered for the PWR Core 2. This process utilizes gas pressure at elevated temperatures to achieve bonding. The program consists of two major efforts; one is concerned with the development of an optimum pressure-bonding process and the other effort is involved with the preparation of irradiation-test specimens. Primary emphasis has been shifted to the development phase of this program.

A series of specimens has been pressure bonded to determine if satisfactory bonding can be achieved using lower pressure-bonding temperatures and a final vacuum anneal at 1832 F to produce recrystallization and grain growth across the interface. The results of this series indicate that specimens can be consistently prepared by pressure bonding at a temperature of 1450 to 1550 F and then vacuum annealing at 1832 F for 15 min. Since the removal of the cans and Ti-Namel spacers from the specimens by pickling is time consuming, the use of barrier layers such as graphite and molybdenum disulfide between the specimens and the spacers is being investigated. Additional spacers are being placed in the ends of the cans to facilitate removal of the specimen, and stainless steel rather than Ti-Namel cans are now being used, since the former are more easily removed and appear to yield more consistent specimens.
Specimens are being prepared to investigate the preparation of cold-rolled Zircaloy 2 surfaces for pressure bonding. Each of the specimens consists of two plates of Zircaloy 2 and will be pressure bonded at 1550 F and 10,000 psi for 4 hr. Abrasive finishing, pickling, and vapor-blasting techniques will be considered. Production methods of abrasive polishing will be investigated and evaluated. Contamination has apparently been introduced during the pickling of components and a study is being directed toward identification and elimination of the contamination. Since vapor blasting with quartz previously resulted in surface contamination, specimens have been prepared to investigate the use of an alumina vapor blast. Zirconia will also be considered as a material for vapor blasting. Bonds will be examined metallographically and will be subjected to corrosion and mechanical evaluation tests.

The reaction occurring between the uranium dioxide cores and the Zircaloy 2 cladding during pressure bonding has led to the investigation of the use of barrier layers between the core and cladding to prevent reaction. Components have been machined which will be coated with various oxides, graphite, molybdenum disulfide, and metal coatings and pressure bonded at 1550 F for 4 hr at 10,000 psi to evaluate the effectiveness of each barrier layer in preventing reaction between cladding and core during pressure bonding. Methods of applying the coatings are being determined, and the thickness of the coating required to minimize reaction will be studied. The oxides to be considered initially are those of calcium, thorium, and magnesium. If difficulty is experienced with graphite or molybdenum disulfide coatings contaminating the Zircaloy 2 bonding surfaces, techniques for preventing the transfer of material to the bonding surfaces will be studied. The feasibility of using metallic barrier layers, such as chromium, iron, molybdenum, nickel, and niobium, are being investigated. After preliminary screening of the specimens, sprayed, plated, and evaporated coatings will be considered for more promising metallic barrier layers. Effectiveness of the barriers will be evaluated by metallography, X-ray techniques, corrosion, and mechanical-property tests.