MELTING AND FORMING
OF
SER FUEL RODS

AEC Research and Development Report

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MELTING AND FORMING
OF
SER FUEL RODS

By
P. S. DRENNAN

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ABSTRACT

Sixty-one metal alloy rods were fabricated for the SNAP Experimental Reactor (SER), a compact nuclear power source. These rods were 1 in. in diameter by 10 in. long, and consisted of an alloy containing 7 wt% highly enriched uranium and 93% zirconium. Melting of the alloy was done by generally accepted consumable arc melting techniques. Extrusion of alloy slugs into rods was carried out, unclad, at 850°F. Suitable controls were developed to insure uniformity of products and reproducibility of batches. The procedures developed for this application are considered adequate for future requirements.
I. INTRODUCTION

The SNAP Experimental Reactor (SER) was designed as a preliminary operable prototype of a reactor concept for a Systems for Nuclear Auxiliary Power (SNAP) unit. The SER consists of a core of self-moderated uranium-zirconium hydride alloy rods, compactly arrayed and beryllium reflected. NaK is circulated through the core and is used as the primary and secondary heat transfer medium. Since power conversion is not a consideration in this experiment, heat is removed from the secondary loop by means of a forced-air heat exchanger.

The core of the SER consists of 61 fuel elements, arranged in a hexagonal array (Figure 1). The individual fuel elements (Figure 2) consist of the fuel rod and beryllium reflector end pieces, clad in a ceramic coated, thin-walled stainless steel tube. The ceramic coating prevents the loss of hydrogen at the operating temperature of 1200°F. The fuel rod is an alloy of uranium in zirconium, containing 7 wt % of highly enriched uranium (93% $^{235}$U) and hydrided to an $N_H^2$ of 6 ($6 \times 10^{22}$ atoms of hydrogen per cubic centimeter).

Generally accepted methods and equipment were used to develop procedures for the production of massive uranium-zirconium alloy ingots and to extrude these ingots to cylindrical shapes, from which fuel rods were machined. Alloying was accomplished by arc melting of consumable electrodes, followed by remelting to obtain homogeneity and soundness. Massive alloy ingots were reduced to cylinder form by cold extrusion. Suitable controls were developed to insure uniformity of product and reproducibility of batches. Some of the control methods were developed primarily for use with the SER. It is anticipated that these methods will be refined further, and used extensively in subsequent similar applications.

This report embodies only the operations of alloying (formation of massive alloy ingots), extruding of ingots to cylinders, and machining of fuel rods for the SNAP Experimental Reactor. The subsequent operations of conversion of zirconium in the alloy to zirconium hydride, and the final cladding of the fuel rods in thin walled, ceramic coated, metal containers, are discussed in other reports.$^1,^2,^3$
Figure 1. SER Core

Figure 2. Fuel Element
II. ALLOYING OF MATERIALS

A. RAW MATERIALS

1. Uranium Metal

Two types of uranium metal were used in the fabrication of alloys for the production of SER fuel elements and prototypes: natural uranium, and highly enriched uranium (containing 93 wt % U\textsuperscript{235}). Natural uranium was used for all melting development and for the production of prototype elements used in developing hydriding and cladding procedures.

Enriched uranium metal was obtained from the Union Carbide Nuclear Company at Oak Ridge, Tennessee, in the form of cut wire, 1/8-in. diameter and approximately 3/16 in. long. A typical analysis is given in Table I, along with the vendor's certification.

2. Zirconium Sponge

Zirconium was obtained from the Wah Chang Corporation in Albany, Oregon, as Kroll process, magnesium-reduced sponge. The specification required that all of the sponge pass a 3/8-in. screen and be retained on a 20-mesh screen. The only specifications stipulated, other than size, was that the zirconium be of reactor grade and have a preferred hardness of about 120 Brinell. A typical analysis and the vendor's certification are given in Table II. It may be noted that most of the sponge used had surface contamination of a carbonaceous-appearing material, and also contained a significant quantity of magnesium chloride, both of which volatilized upon melting. This presented minor difficulties, such as impaired visibility into the furnace during melting, and hygroscopic residues on the primary ingots and in the furnace body. These resulted in excessive pumpdown time and impaired operation of some of the valves.

3. Zirconium Metal

Zirconium metal bars of high purity were used to fabricate connectors for attaching the alloy electrodes to the stinger tip in the arc melting furnace. These connectors were cleaned and reused many times during the fabrication period. However, it is certain that, at times, small quantities of the connectors were melted into the alloy.
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*U^{234} = 1.09 wt %, U^{235} = 93.19 wt %, U^{236} = 0.45 wt %
## TABLE II

**SPECTROGRAPHIC ANALYSIS OF ZIRCONIUM SPONGE**

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Figure 3a. 500 Ton Hydraulic Press and Glove Box Used for Weighing and Pressing Electrodes

Figure 3b. Die Used for Pressing Zirconium Sponge for Primary Electrodes
B. COMPACTING

To make one compact, 63 g of uranium pellets were blended with 837 g of zirconium sponge in a hardened steel die and compacted into a bar, 1 in. by 1 in. by 10 in. long. Blending was accomplished by pouring in half of the sponge, lightly compacting, adding uranium pellets uniformly along the length of the die, and then adding the remaining half of the sponge. Compaction was effected with a hardened steel punch, pressed into the die at a force of 500 tons by means of a hydraulic press (See Figure 3). The press was totally enclosed and vented to insure safety of use with radioactive material.

Eighteen compacted bars were required for each massive arc melted ingot. The bars were welded end-to-end, in groups of three, and were welded to a zirconium connector for bolting onto the furnace stinger.

It was important that the uranium and zirconium be blended and compacted as uniformly as possible, in order to obtain good dispersion of the uranium throughout the alloy. It was further desirable to disperse the zirconium sponge fines evenly throughout the compacts, in order to obtain maximum strength.

C. WELDING

After the raw materials were compacted into bars, they were welded end-to-end, to form three-bar electrodes (Figure 4) with a bolting connector welded to one end. Welding was done in a special glove box (Figure 5), in an argon atmosphere, to prevent oxidation of the alloy. A conventional, 300-amp dc Heliarc welding machine was adapted for use in the glove box. A 3/16-in. tungsten non-consumable tip was used for fusing the electrodes together. Electrode components were placed in the glove box and the air was evacuated. After a brief degassing period, the box was back-filled with argon to atmospheric pressure, and the glove ports were opened. Alignment of the compacts was facilitated with a steel angle-iron jig, and the components were fused together, using about a 200-amp current. It was essential that the electrode components be welded in straight alignment because of the relatively close tolerances of the electrode in the crucible, with the subsequent danger of arcing to the side wall during melting.
Figure 4. Compacted Bar Electrode

Figure 5. Welding Glove Box
The same procedure was followed in welding the primary melted ingots together to form an electrode (Figure 6) and for remelting the alloy. The only other considerations involved were those of mass and shape, as discussed in Section II-E.

Figure 6. Primary Melted Electrode

D. ARC MELTING

The alloying of compacted raw materials, and the remelting to form massive ingots, was done in a modified consumable electrode arc melting furnace (Figures 7 and 8), manufactured by the Oregon Metallurgical Corporation, Albany, Oregon. The furnace consisted of a body with viewing ports, a controlled, movable cathode (stinger), and detachable, water-cooled, copper melting pots (anodes). The cathode assembly consisted of a chrome plated, copper, water-cooled shaft, shown in Figure 8, which passed through a gland at the top of the furnace body. A drive screw, attached to the upper end of the shaft, passed through a gear train activated by an amplidyne drive. Predetermined arc voltage limits (a function of arc length) were used to control the direction of the amplidyne drive. By these means, continuous controlled feed of the electrodes was obtained.

Copper crucibles having removable bottoms (Figure 9) were attached to the bottom of the furnace body, using a heavy resilient rubber gasket to effect a seal and clamps to facilitate rapid changing. A water jacket, having a solenoid wound along its exterior surface for stirring of the molten alloy, surrounded the crucible. A flow of about 40 gpm of water was used to cool the crucible and
Figure 7. Arc Melting Furnace

Figure 8. Schematic of Arc Melting Furnace
freeze the melt. Two sizes of crucible were used for melting the SER core: a 2.4-in. ID by 16-in. deep crucible for melting the 1 in. by 1 in. by 30 in. compacted electrodes, and a 4-in. ID by 16-in. deep crucible for remelting the primary alloy ingots. The smaller crucibles were tapered to facilitate removal of the cooled ingot; this was unnecessary for the larger ingots.

Figure 9. Melting Crucible

A vacuum system, consisting of a 130-cfm Kinney Model KDH-130 mechanical pump, a 1300-cfm Stokes Model 900-150-2, 6-in. ring jet diffusion booster pump, and a 5-cfm Welch Model 1402 mechanical holding pump (Figure 8), was attached to the furnace. This vacuum system was capable of pumping the furnace down to about 0.4 μ in a relatively short time and of maintaining a pressure of between 5 and 30 μ during the vacuum melting of a 4-in. ingot.

Power for arc melting was supplied by a bank of 6 Miller Electric "Bumble Bee" selenium rectifiers (Model 1000), and provided a combined maximum output of 6000 amp. From the busses, power was brought into the furnace by flexible cables attached to the top of the movable cathode shaft and to "ears" on the melting
crucible top flanges. The individual rectifiers were preset at incremental percentages of their output, and were controlled as Off or On by remote toggle switches located on a control console. In this manner, arc current could be selected by cutting in or out any combination of rectifiers. All of the process variables, except inert gas flow and pressure, were controllable from the console.

Melting of an ingot began with the opening of the furnace, which was normally maintained assembled and under vacuum to prevent absorption of moisture from the air. A crucible, containing a 150-g blend of uranium and zirconium sponge as starting charge, was placed in position on the bottom of the furnace body and the system evacuated. After a normal pumpdown period of 15 min, the main vacuum valve was closed and a small bleed valve opened, admitting argon into the bottom of the furnace body at a rate to establish a pressure of 300 mm Hg. The amplidyne circuit was closed, and the electrode was started downward into the crucible. When the electrode touched the starting charge, an arc was established and automatically adjusted to the desired length (about 1/2 in.) by the preset voltage controller. As soon as a pool was formed, the amplidyne action was more or less stabilized and melting of the electrode proceeded smoothly at about 1600 amp. Intimate stirring of the molten pool, which was about 2 in. deep, was accomplished by means of a magnetic field induced by the solenoid around the outside of the cooling jacket; this rotational force field also served to control the direction of the arc and minimize arcing to the side of the pot. When all the electrode had been melted, as evidenced by visual observation and/or by the distance traveled by a pointer attached to the stinger, the current was shut off and the stinger retracted. After cooling, a second electrode was attached and melted onto the top of the first, following the aforementioned procedure, except that no starting charge was used. These two compacted 30-in. electrodes thus produced an ingot, 2.4-in. diameter by about 12 in. long, weighing about 5 kg. When three of these primary melted ingots had been produced, their ends were faced off in a lathe, they were welded end-to-end, and a connector attached to form an electrode (Figure 6) for remelting into a larger ingot.

E. REMELTING

Remelting of the single alloy electrode, into a 4-in. diameter crucible 16 in. deep, was carried out without interruption. This produced a 4-in. diameter by
about 12-in. high ingot weighing about 15 kg (Figure 10). All remelting was done at about 28 v and 3300 amp, under the highest vacuum possible (from 5 to about 30 μ), using a 350-g starting charge. When all but about 3 in. of a 2.4-in. diameter electrode had been melted, the current was shut off and the electrode was retracted about 2 in. After a brief period to allow entrapped gases to escape and the pool to partially solidify, the amplidyne was reactivated and the remainder of the electrode melted. This procedure, known as "hot-topping," materially reduced the size of the shrink-cavity normally left at the top of a vacuum arc melted ingot, caused by the release of gases from the molten metal and by shrinkage upon solidification. The efficiency of the arc melting process was determined largely by the amount of usable material obtained from a finally melted ingot.

Considerable work has been done on controlling the physical quality of arc melted ingots. The principal considerations are surface conditions, uniformity, and grain structure. These factors are under the control of arc current and magnetic stirring field. Other important variables are arc voltage, current density, atmosphere, and pressure.
Figure 11. Photomicrographs of Zirconium-Uranium Alloy
Actual forming of fuel rods started with the double-melted ingot. After heat treating for 2 hr at 1480°F, the ingots were machined into slugs for extrusion into rods. The extruded rods were then machined into "raw" fuel rods for hydriding. Elaboration of each of these steps follows.

A. HEAT TREATING

Cast ingots were cropped and sampled at the top end to remove the sleeve and shrink cavity and to provide a sample of the cast structure for determining hardness and/or other physical properties. Hardness of the cast ingots ranged from 92 to 96 on the Rockwell B scale. In general, the hardness of a transverse section increased from the outside to the center. It was found that the working properties of the alloy were improved by heat treating in an argon atmosphere at 1480°F for at least 2 hr, followed by furnace cooling over a period of 16 hr; a certain degree of grain refinement was obtained, as illustrated in Figure 11b. Negligible changes in hardness values were measured after heat treatment. Extension of the time beyond 2 hr did not significantly improve the physical properties.

B. MACHINING OF SLUGS

After heat treatment, the ingots were machined into slugs for extrusion. All rough end cutting was done with a power hack saw, and all finishing of the slugs was done on a lathe. The extrusion slug is a right circular cylinder, 3.75-in. diameter by about 10 in. long, having one end shaped to a blunt cone. All machining operations of the alloy were carried out in enclosed and vented equipment with the benefit of coolant, since the small chips were quite pyrophoric.

C. EXTRUSION

Extrusion of the slugs into rods was done at Bridgeport Brass Company in Riverside, California, using a Lake Erie horizontal press having a capacity of 3500 tons. The slugs were heated in an electric furnace to 850°F, coated with a proprietary lubricant, and extruded through a 1-in. die, preheated to about 500°F. Ram speed of the press was about 4 in./min. Pressure during extrusion
averaged about 1100 tsi; initial pressures were a little above this value. No followup slug was used in these extrusions, hence a heel was left from each slug.

In general, all slugs were extruded successfully to give straight smooth rods. However, in some cases, there was a breakthrough in the lubricant which resulted in light galling of the die and slight striations along the surface of the rod (See Figure 12). During the production of prototype and fuel elements for the SER, only one set of dies was used for the extrusion of 32 slugs.

![Figure 12. Extruded Rod](image)

D. MACHINING OF FUEL RODS

The lengths of extruded rods were cut into 10-5/8-in. long slugs and into specimen wafers; a specimen was taken from between each slug and from each end of the rod. Centers were cut in each end of the slugs, and their diameters were machined to remove residual lubricant and to clean the surface. These operations completed the scope of work included in this report; subsequent operations of hydriding, cladding, and testing are covered in other reports.¹,²,³
IV. QUALITY CONTROL

A. SAMPLING

Each lot of uranium and zirconium received was sampled for spectrographic analysis to insure that impurity specification maxima were not exceeded. The satisfactory performance of a compact nuclear reactor depends upon uniform distribution of fuel material throughout a matrix. Therefore, sample wafers were taken at each end of each fuel rod. These samples were used for the various analyses discussed below, and then were stored for permanent reference.

During the preliminary stages in the production of SER fuel rods, an extruded ingot was carefully sampled and analyzed to determine that the alloy was homogeneous. It was concluded that dispersion was uniform, at least to the extent of analytical accuracy. Typical results are shown in Table III.

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<td>R-3</td>
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</tr>
<tr>
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<tr>
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<td>R-9</td>
<td>6.9</td>
</tr>
<tr>
<td>R-10</td>
<td>7.1</td>
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</table>

B. METALLURGICAL

Specimens of cast and extruded materials were mounted and polished for microscopic examination. Photomicrographs are shown in Figure 11a, b, and c.
Metallographic examination of the structure of the cast alloy ingot (See Figure 11a) shows a typical coarse Widmanstatten pattern of alpha zirconium plates. Heat treatment of this material, at 1480°F for 2 hr, followed by a 16-hr furnace cool, produced little noticeable change in microstructure and slight grain refinement (Figure 11b), a fact which is borne out by the negligible hardness change cited earlier.

The microstructure of the extruded material (Figure 11c) shows the consequent elongation and fragmentation of the initial cast structure, typical of a material having been subjected to such a reduction in area (14 to 1). Upon examination at higher magnifications, the darker strings of fragmented material showed no evidence of hard, brittle phases, indicating insignificant amounts of intermetallics, such as oxides, etc.

This alloy appears to be a clean material, free of the inclusions normally found in metals not purified by vacuum melting.

C. PHYSICAL

Physical testing of raw fuel rods included hardness, density, and soundness determinations. Each test is discussed in the following sections.

1. Hardness

Hardness of both the cast and extruded alloy was determined by means of a Kendall Model CT4 Hardness Tester. Most of the hardness values, on both cast and extruded material, were in the range of 92 to 96 Rockwell B. In the case of extruded rods, hardness increased from the outside to the center. Generally, hardness was used as an index to the impurity content of the alloy, particularly oxygen and nitrogen, and also as a guide to its extrudability.

2. Density

Density of extruded and machined components was obtained by the Archimedes principle, using a small amount of wetting agent in the water. Measured densities ranged from 6.800 to 6.820. A theoretical density of 6.803 was calculated for the 7 wt % alloy on a simple mixture basis.
3. Soundness

The physical integrity or soundness of the SER fuel rods was determined by two methods: ultrasonic testing, and Zyglo. The Zyglo procedure was used primarily for screening, since it detects only surface flaws and must rely upon visual interpretation; whereas the ultrasonic method is fully automated and detects all flaws, both external and internal, to within micron widths. This ultrasonic testing instrument, Figure 13, was purchased from Automation Instruments Company in Pasadena, California. Figure 14 shows a typical ultrasonic scan photograph, depicting an internal flaw in a rod.

D. ANALYTICAL

All available analytical methods were used to determine the composition of SER materials. This was done to evaluate each method on its own merits, and to establish satisfactorily reliable control procedures for the SER and future similar applications. Each method will be discussed briefly. A comparison of values obtained is shown in Table IV.

TABLE IV

<table>
<thead>
<tr>
<th>Sample</th>
<th>X-ray Spectrograph</th>
<th>Wet Chemical</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Solid Samples (wt % U)</td>
<td>Solutions (wt % U)</td>
</tr>
<tr>
<td>A</td>
<td>6.83</td>
<td>6.86</td>
</tr>
<tr>
<td>B</td>
<td>6.90</td>
<td>7.02</td>
</tr>
<tr>
<td>C</td>
<td>6.95</td>
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</table>

1. Chemical

Uranium was determined volumetrically by oxidizing the +4 valent form to the +6 valent form, using a ceric sulfate solution which had been standardized
Figure 13. Ultrasonic Tester

Figure 14. Ultrasonic Scan Photograph
with NBS U$_3$O$_8$. Corrections were made for interfering ions, such as iron. This method was useful only for the determination of total uranium.

Oxygen, nitrogen, and hydrogen were determined in an NRC Model G12 Vacuum Fusion Gas Analyzer. Chips of the alloy were dropped into molten platinum at 1950°C, and the liberated gases were measured. Hydrogen was oxidized to water, which was absorbed in magnesium perchlorate; oxygen was converted to carbon dioxide and removed by freezing; and nitrogen was measured as the remaining noncondensable gas. Hydrogen in the hydrided materials was determined in the gas analyzer equipment by thermal dissociation and measurement of the liberated gas.

Zirconium was not determined directly by any method, but was identified only as the base or major constituent.

2. X-ray Fluorescent

A Norelco X-ray Spectrograph, operating at 50 kv and 40 ma, and utilizing a tungsten target X-ray tube, also was used to analyze for total uranium. The X-ray spectra were dispersed by means of a lithium fluoride analyzing crystal, and the intensities of the Lα$_1$ line of uranium and the second order of the Kβ$_1$ line of zirconium were measured in all cases. The ratio of the intensity of uranium to the intensity of zirconium of standard samples was plotted against the percent of uranium to establish a working curve for the determination of the amount of uranium present in the unknown samples.

Both solid samples and solutions were analyzed by the X-ray spectrographic method. Solid samples were prepared for analysis by smoothing a surface; solution samples were made by dissolving solid material in nitric and hydrofluoric acids and diluting to a known volume. Standard solutions, made from NBS U$_3$O$_8$ and reactor grade zirconium metal, were used for calibration. Strontium nitrate, added to the above solution, provided an internal standard solution in which the ratio of the intensity of uranium to that of strontium was determined and plotted against percent uranium.

The methods described are quick and accurate to within about 0.03 wt % U.
Figure 15. Gamma Pulse Spectrometry Apparatus - For Rods
3. Spectrographic

Impurities in raw materials were determined by means of a Baird Three Meter Model GXI Spectrograph, using a 15,000 lpi grating. A Baird spectro-source was used for dc arc excitation. Small sample chips or turnings were oxidized in platinum containers in a muffle furnace at low temperatures, and the resultant oxides mixed with carriers for excitation.

Standard plates were prepared from synthetic samples containing known quantities of impurities; these plates were used for visual comparison with the spectra of the materials being analyzed.

4. Gamma Pulse Spectrometry

Gamma pulse spectrometric techniques were used for the nondestructive determination of $^{235}\text{U}$ in sample wafers and for full-length, clad fuel elements. A slightly modified apparatus, as described by Judd and Leboeuf in KAPL-1925, was used. This method consisted essentially of counting either a 186- or 256-kev gamma ray which is emitted in $^{235}\text{U}$ decay. A sodium iodide (thallium activated) crystal was used for detection. The light quanta produced in the crystal by gamma radiation absorption were then sensed, amplified, and translated onto a recording chart and/or to a scaler in conjunction with a printer. This apparatus is shown in Figure 15.

In the case of the sample wafers, a total count of the 256-kev gamma ray, for a relatively short time, and the counts per gram were compared to show the variation in $^{235}\text{U}$ content from sample points at each end of the fuel elements. Little variation was found.

In the case of the fuel rods, the entire surface of each rod was exposed to the detector, through a 1/2 in. by 1/4 in. window. The rod was moved past the window with a rotating motion. Thus, a cumulative count of the 186-kev gamma ray was obtained for each rod, so as to define its homogeneity as well as its total $^{235}\text{U}$ content.

Although the results of these measurements were not expressed as percent of $^{235}\text{U}$, they served to show that all of the elements were homogeneous in fuel content and that each element contained the same quantity of fuel, to within ±0.1 wt %.
V. SUMMARY AND CONCLUSIONS

Basic equipment and procedures were used to produce massive alloy ingots of 7 wt % uranium (both natural and highly enriched) and 93 wt % zirconium. These ingots were then heat treated at 1480°F and machined into slugs for extrusion into rods. Low temperatures (850°F) and a high reduction ratio (14:1) were used to give rods of highly worked structure. The extruded rods were machined into raw fuel slugs for hydriding.

An elaborate procedure of sampling and analysis was used to insure uniformity of composition and maximum quality of product. Particular emphasis was placed upon X-ray fluorescence analysis for total uranium, and upon gamma pulse spectrometry analysis for $^{235}$U.

A sufficient number of raw slugs was produced to develop the required techniques for the hydriding and cladding of 61 finished fuel rods meeting the exacting specifications for insertion into the SER core.

Sufficient experience has been gained, and suitable techniques developed, during the fabrication of the core elements for the SER, that a minimum amount of material and time will be required for the production of arc melted alloys for subsequent SNAP 2 reactors.
REFERENCES


2. S. Elchyshyn, "Improved Welding Techniques for SER Fuel Element Assembly Closures," NAA-SR-4909 (to be published)

