

High-Performance Uranium-Metal Fuels for Savannah River Reactors

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Abstract

The development of high-performance uranium-metal fuel elements for Savannah River reactors required a major metallurgical effort to achieve satisfactory irradiation behavior. Fuel element designs were progressively modified from the original small-diameter slug through a hollow slug of similar dimensions to the final large-diameter tubes that provided the increased heat transfer surface needed for high reactor powers. Concurrent adaptation of cladding techniques led to the development of hot-press bonding and later to hot-die-size bonding of the aluminum can onto the nickel-plated uranium core to replace the original aluminum-silicon-dip bonding process.

Concurrent modifications in processing the uranium cores were necessary for satisfactory reactor performances of the fuel elements. An intermediate rate cooling process, specifically oil quenching, for beta host treatment of the alpha phase cores was developed to prevent introducing (a) crystallographic texture due to fast cooling from the beta phase, which resulted in anisotropic shape changes during irradiation, and (b) a large grain size from slow cooling, which produced surface roughening. Low solubility alloying elements, such as silicon and aluminum, were added to the uranium to control the gross cavitation swelling encountered during irradiation at elevated temperatures. The resulting uranium fuel elements proved capable of sustaining the reactor exposures required for many years production of nuclear materials at the Savannah River Plant.

Introduction

Uranium-metal fuel elements used in the heavy-water-moderated-and-cooled Savannah River Plant (SRP) reactors evolved over the initial 20 years of reactor operation through a series of changes in design, composition, and fabrication processes. These changes were dictated by production requirements, cost reduction, and fuel-element performance. This paper outlines the principal technological features of this evolution, emphasizing the metallurgical development required to produce a high-performance fuel element.

Fuel Design and Fabrication

The fuel elements employed originally in the SRP reactors on startup in the early 1950s were 1-inch-diameter x 8-inch-long solid unalloyed uranium slugs machined from rolled rods and clad in aluminum cans for corrosion protection

from the coolant water. To improve heat-transfer capability, this design was changed in the late 1950s to a hollow slug of similar external dimensions. The slug was clad with aluminum on both outer and inner surfaces. Further improvement in heat transfer was realized in the early 1960s by introducing nested assemblies of large-diameter (2 to 4 inches) tubular elements with extruded uranium cores and aluminum cladding. Typical fuel configurations are shown in Figure 1.

The fuel elements were fabricated from uranium cores produced at the Fernald Plant, National Lead Company of Ohio, by shaping cast billets at high-alpha-phase temperatures and heat treating at beta-phase temperatures.¹ The heat-treated cores were clad in aluminum cans, which were metallurgically bonded to the core to ensure good heat transfer during operation and to limit core corrosion if the cladding were to be penetrated, as at a closure defect.

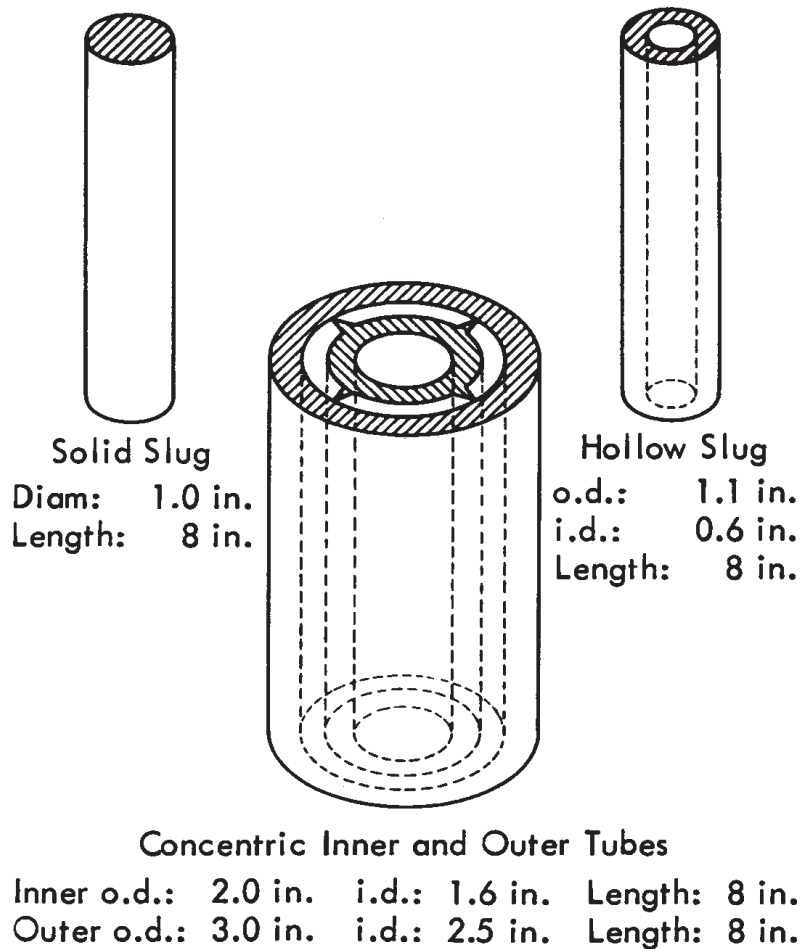


Figure 1. Typical configurations for SRP uranium fuel elements

Three techniques, Al-Si dip canning, hot pressing, or hot-die sizing, were employed to bond the aluminum cans to the uranium cores.

The solid slugs and some of the hollow slugs were canned by the Al-Si dip process, in which the heated slug was inserted into the aluminum can under the surface of a molten Al-Si bath, and an aluminum endcap was added. This assembly was then quenched in water. Bonding resulted from the thin Al-Si braze layer. Final closure was by tungsten-inert-gas (TIG) welding the can to the endcap.

Before canning, the uranium slugs were given a beta-phase heat treatment in a molten bronze bath, followed by quenching in a molten tin bath. This heat treatment was originally in-

cluded as a preliminary step in the canning operation, but in later Al-Si dip-canning processes the treatment was performed during slug fabrication.

Most of the hollow slugs and all of the large-diameter tubular fuel elements were canned by hot-pressing or hot-die-sizing techniques. Both processes required a previously beta-treated core, electroplated with a 0.0001-to-0.0005-inch nickel layer, to serve as a bond between the uranium and the aluminum.

In the hot-pressing process, the electroplated uranium core was placed in a preformed aluminum can, an aluminum endplug was inserted, and the can was pressed mechanically at elevated temperatures to achieve bonding.

The hot-pressing operation produced bonding by interdiffusion at the uranium-nickel and the nickel-aluminum interfaces to form well-defined intermetallic compounds. Final closure was by welding.

The hot-die-sizing process was developed specifically for canning large-diameter tubular elements. In this process, the nickel-plated uranium core was loaded into the aluminum can, an aluminum endcap was inserted, and the preheated assembly was forced through a die to size the can onto the core. The die-sizing operation reduced substantially the thickness of the can wall and formed a diffusion bond between the uranium and aluminum components and the intermediate nickel layer. The ends of the element were bonded by a hot-pressing operation. Final closure was by welding.

Bond strengths of the hot-pressed and hot-die-sized elements were about twice that of the Al-Si-bonded elements. Bond strengths were used as indicators of good heat transfer for the elements. Special advantages of the hot-die-sizing process were the high rate of throughput of canned elements, small capital investment, and low cost of operation.

Fuel Element Structure and Irradiation Behavior

Tailoring the metallurgical structure and properties of the uranium core was necessary to achieve satisfactory fuel performance under the increasingly severe irradiation conditions imposed by the optimized fuel designs. In SRP reactors, the fuel elements were stacked in vertical columns within aluminum-alloy process tubes, through which the relatively low-temperature coolant water (D_2O) flowed. Coolant-channel spacing was maintained by longitudinal ribs on the process tube and fuel-cladding surfaces. The coolant-channel spacing limited the amount of distortion of a fuel element that could be tolerated during irradiation without disruption of reactor operation.

The most important sources of dimensional change during irradiation of a uranium fuel element were anisotropic growth and related surface roughening and swelling. Anisotropic growth—a change in shape with no large change in volume—and surface roughening were direct manifestations of the basic dimensional instability of the orthorhombic uranium crystal under irradiation. Polycrystalline specimens with crystallographic texture would undergo dimensional changes during irradiation analogous to those of the alpha-uranium single crystal (i.e., lengthening in predominantly b -axis directions, shortening in a -axis directions, with essentially no change in c -axis directions). Random textures were required to avoid anisotropic growth. Moreover, a specimen with large grain (or grain-domain) size would suffer a surface roughening during irradiation because of the same instability in the individual grains; therefore, a small final grain size was required.

Swelling, a volume increase of the fuel element during irradiation, occurred as the result of agglomeration of either irradiation-generated lattice vacancies into cavities and voids at intermediate temperatures or fission gases into bubbles at higher temperatures. The swelling at intermediate temperatures could be greatly enhanced by stress from the interaction of individual grains undergoing anisotropic growth during irradiation. Agglomeration of fission gases constituted a less severe problem in SRP reactors because operating temperatures were below the gas-swelling range.

Anisotropic Growth Control

Satisfactory resistance of the SRP fuel elements to anisotropic growth was attained by adopting heat-treatment processes that provided a randomly oriented, small-grained uranium structure. Experience prior to startup of the SRP reactors had led to the development of beta-phase heat treatments to avoid, on the one hand, the anisotropic growth of uranium caused by texture produced by forming in the alpha phase and, on the other hand, the excessive surface roughening caused by the large

grain size of cast or gamma-formed metal. The beta treatment, incorporated initially as a preliminary bronze-tin-dip in the Al-Si canning process, yielded a structure with acceptable dimensional stability for early SRP operation.

The introduction of hot-press bonding a few years later, however, required a prior beta treatment, which was initially carried out in molten salt with water quenching. Application of this process to hollow slugs led to the incidence of anisotropic growth caused by texture produced in the fuel core during fast cooling from beta-phase temperatures. The hollow slugs, heat treated using a water-quench process and canned by either Al-Si-dip or hot-pressing procedures, increased in both inside and outside diameters, as well as length, and decreased in wall thickness, by a much greater extent than slugs heat treated by the previous bronze-tin-quench procedure.

The as-worked texture of the metal formed in the alpha phase was usually eliminated during beta treatment. Conditions of the beta treatment, particularly the means of cooling from beta-phase temperatures, had to be controlled, however, so as not to reintroduce the texture (McDonell and Sturken 1966). Fast cooling, as produced by water quenching, was found to produce a pronounced texture, with the *a* axis of the uranium crystal oriented predominantly in the direction of heat flow during cooling, and the *b* axis oriented predominantly in other directions. Thus, a hollow slug quenched from beta temperatures exhibited predominantly *a*-axis textures in radial (wall thickness) directions and *b*-axis textures in longitudinal and circumferential directions. Such a texture accounted for the anisotropic growth of the water-quenched hollow slugs during irradiation. Slow cooling, as in air, produced a large grain size in the metal, causing surface roughening during irradiation. The slow cooling could also produce a texture in the fuel, though resultant anisotropic growth was obscured by the surface roughening.

Intermediate-cooling-rate treatments were found necessary to obtain an adequately small

grain size without excessive texture. Irradiation tests, conducted in conjunction with texture and metallographic studies, correlated the behavior of hollow slugs and tubular fuel elements with several candidate, intermediate-cooling-rate treatments. Of these, the oil-quench treatment was generally the best. It yielded a moderately small-grained microstructure with relatively little texture and resulted in near-optimum stability during irradiation.

Swelling Control

Fuel-core splitting, which caused slug-type elements failure, led to considerable cooperative efforts among Hanford, Savannah River Plant, Mallinckrodt, and National Lead before SRP startup to reduce levels of impurities that might embrittle the metal. This effort was successful, and this good-quality unalloyed uranium was in use as fuel in SRP reactors when preliminary irradiation tests of the large-diameter fuel tubes revealed a new and unexpected form of dimensional instability (Angerman and Caskey 1964). This instability was caused by formation of large (100 mm) internal cavities in the fuel at intermediate temperatures (see Figure 2). Tubular fuel with large cross sections of metal at intermediate temperatures swelled 10% or more. This swelling produced principally wall-thickness increases that caused detrimental coolant-flow changes and increased the potential for cladding failure. Below the intermediate temperature range, only minor volume increases (<1%, attributed to formation of solid fission products) were observed; above this temperature range, large brittle cracks, but no cavities, were seen.

The intermediate temperature swelling, termed "cavitation swelling," differed considerably from the hitherto recognized swelling of fuel materials caused by the formation of fission-gas bubbles at high temperatures. Fission-gas bubbles in SRP fuel were shown to contribute only a small volume increase (<1%). Formation of the large cavities was therefore attributed to internal interactions between individual grains undergoing anisotropic growth in the metal.

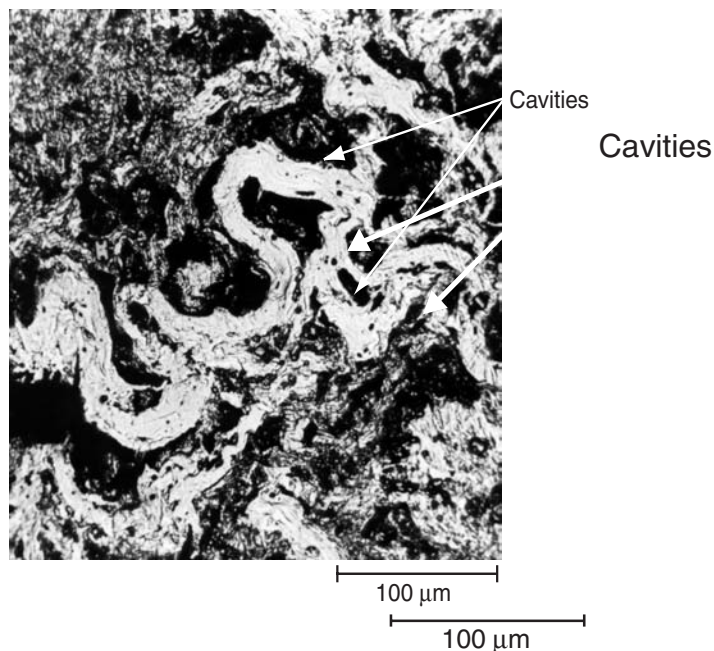


Figure 2. Large cavities formed in swollen regions of uranium fuel elements

Cavitation swelling produced significant volume changes only after critical exposure thresholds were exceeded (see Figure 3). The exposure threshold for unalloyed uranium decreased with increasing temperature in the intermediate temperature range, but at higher temperatures, apparently increased again, in accordance with the decreased susceptibility of the metal to anisotropic growth.

Observations that swelling behavior varied markedly among fuel elements derived from different billet castings suggested that relatively minor variations in impurity levels affected cavity formation. Impurity silicon in the range 10 to 65 ppm appeared especially significant. Irradiation tests of such variables demonstrated that minor additions of iron and silicon (100 to 150 ppm) markedly improved fuel behavior by extending exposure thresholds for cavitation swelling (see Figure 3). These results agreed with Harwell reports of the good behavior of uranium fuel containing small iron and aluminum additions. SRP fuel specifications were accordingly modified to provide for additions

of 125 to 200 ppm of iron and 75 to 150 ppm of silicon, and these values were later increased to 125 to 225 ppm of iron and 125 to 225 ppm of silicon.

Additional irradiation tests over the period, undertaken in conjunction with microstructural and mechanical testing studies, served to define the swelling behavior and its relation to the structure and properties of a large number of Fe-, Si-, Al-, Cr-, and Mo-containing alloys, for exposures up to 5000 MWd/t at temperatures up to 500°C (see Figure 4) (McDonnell, et al. 1969). These results were later extended to greater than 10,000 MWd/t at temperatures up to 400°C. Testing specimens at high exposures was generally undertaken in NaK-containing stainless-steel capsules, where detrimental dimensional changes could be accommodated without interference with reactor operation. These tests were followed by irradiation of satisfactorily performing compositions as full-size tubular fuel to verify expected performance characteristics.

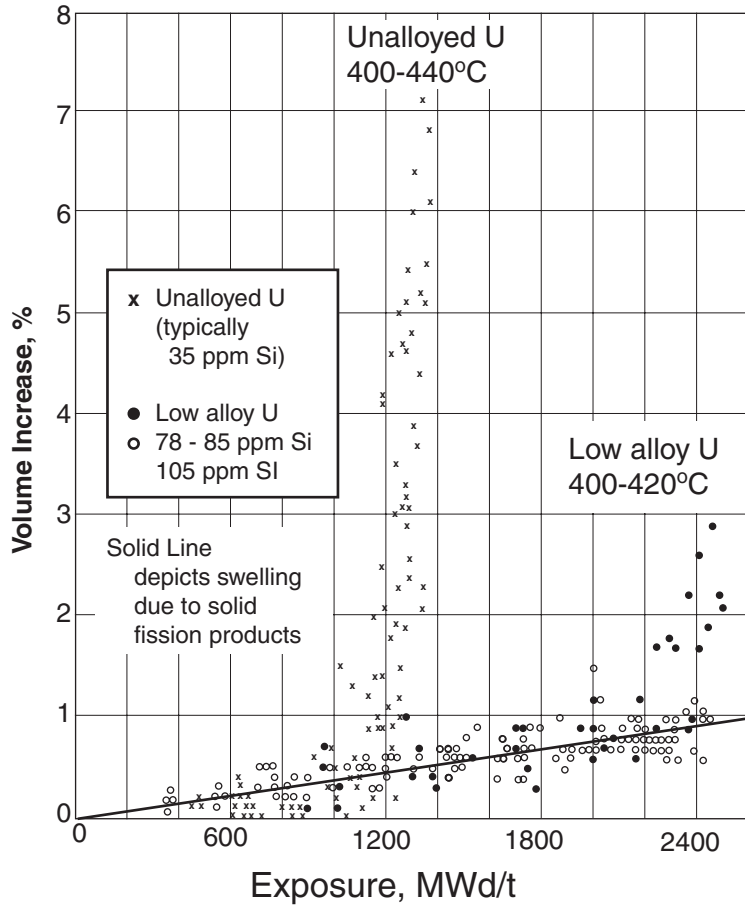


Figure 3. Swelling of unalloyed uranium and low silicon-containing alloy fuel elements during irradiation

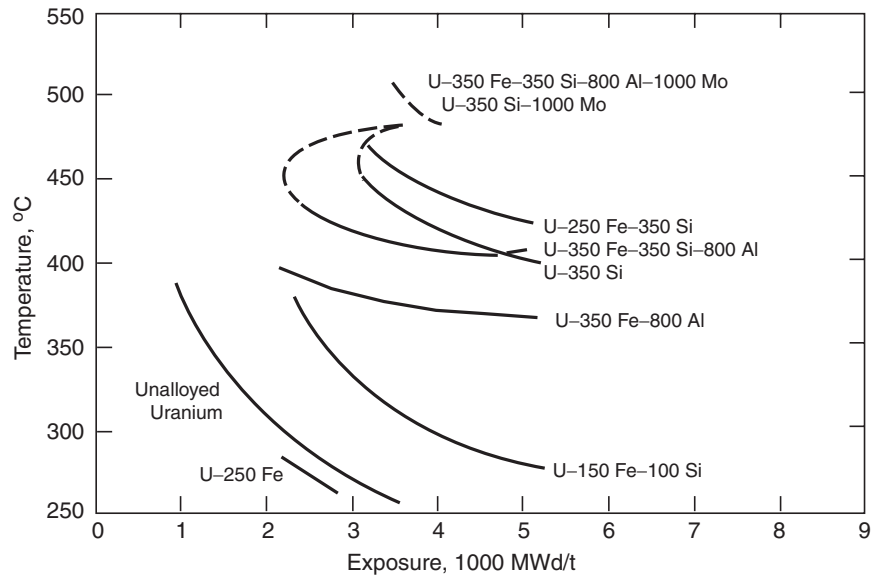


Figure 4. Temperature-exposure thresholds for swelling of uranium fuel (alloying concentrations in parts per million)

The principal features that emerged from these investigations were as follows:

- Unalloyed uranium swelled at relatively low temperatures and exposures, depending in some measure on impurity (especially silicon) concentrations in the metal.
- Alloying additions of low-solubility elements increased swelling resistance and allowed higher temperatures and/or longer exposures during irradiation before swelling began. Silicon concentrations of 250 to 350 ppm alone, or in conjunction with other alloying elements, especially molybdenum, were most effective for exposures up to 5000 MWd/t. Aluminum and larger silicon additions were less effective, and incremental iron or chromium additions produced no discernible benefit. At exposures over 10,000 MWd/t, alloys containing 800 ppm aluminum in conjunction with other alloying elements were more swelling-resistant than those containing 250 to 350 ppm silicon.
- Irradiation produced gross distortions of grain structure and alloy-phase distributions that minimized the effects of the initial structures. The grain distortions resulted from interactions of individual grains undergoing anisotropic growth during irradiation. The cavities formed as a consequence appeared more closely related to the characteristics of the substructure that emerged during irradiation than to the original grain structure, which was essentially destroyed. The alloy content rather than the grain structure appeared to be the primary controlling factor in swelling resistance. Alloy phases initially present in the structure, such as UAl_2 , were dispersed and possibly dissolved during irradiation, and the relative swelling resistance of the alloys appeared to be established by the characteristics of the submicroscopic dispersions of alloy phases thus formed.

The above results served for many years to define limiting conditions for using metallic uranium fuel in SRP reactors without detrimental swelling. Tubular elements irradiated to high

exposures occasionally exhibited gross core cracks, but such cracks did not penetrate the aluminum cladding unless swelling thresholds were exceeded.

Extensive studies of the fundamentals of swelling of uranium and uranium alloys pursued at the Savannah River Laboratory and at Hanford indicated that cavitation swelling, closely related to anisotropic growth in uranium, probably represented the first manifestation of the later generally recognized problem of void formation induced by fast-particle irradiation of many materials (McDonell 1973).

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End Notes

1. Metallic uranium exhibits three crystallographic structures depending on temperature (Foote 1956). Up to 668°C, the metal has an orthorhombic crystal structure (alpha phase); from 668-774°C, a tetragonal crystal structure (beta phase); and above 774°C to the melting point 1132°C, a cubic crystal structure (gamma phase). The orthorhombic alpha phase is highly anisotropic exhibiting in single crystals marked differences in properties and irradiation growth in different crystallographic directions. Polycrystalline uranium specimens share the anisotropic properties and behavior of the single crystal, to the extent the crystals are preferentially oriented in the specimens. Uranium specimens with preferred crystallographic orientations are said to be "textured".

Biographies

William R. McDonell

B.S., Chemistry, University of Michigan, 1947;
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Participated in the development of uranium fuel elements for Savannah River reactors, product forms for Pu +238, Cm -244, and Cf -252 radioisotopic heat and radiation sources, technology and costs of disposal of high-level, transuranium, and low-level radioactive wastes. Principal research interests in effects of radiation on metals, ceramics, and glasses. Recent work devoted to long-range planning for disposition of spent reactor fuels and radioactive materials.

George R. Caskey, Jr.

B.S., Metallurgical Engineering, University of Illinois, 1950; M.S., Metallurgy, MIT, 1952; Ph.D., Materials Science, MIT 1969.

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