Fuel Fabrication Development for the Surrogate Sphere-Pac Rodlet

July 2005

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Nuclear Science and Technology Division

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ACRONYMS AND ABBEVIATED FORMS

AFCI	Advanced Fuel Cycle Initiative
AGR	Advanced Gas Reactor
ATR	Advanced Test Reactor
FCCI	fuel-clad chemical interaction
FCMI	fuel-clad mechanical interaction
INL	Idaho National Laboratory
LWR	light-water reactor
ORNL	Oak Ridge National Laboratory

1. INTRODUCTION

Sphere-pac fuel consists of a blend of spheres of two or three different size fractions contained in a fuel rod.¹⁻⁵ The smear density of the sphere-pac fuel column can be adjusted to the values obtained for light-water reactor (LWR) pellets (91–95%) by using three size fractions, and to values typical of the fast-reactor oxide fuel column (~85%) by using two size fractions. For optimum binary packing, the diameters of the two sphere fractions must differ by at least a factor of 7 (ref. 3). Blending of spheres with smaller-diameter ratios results in difficult blending, nonuniform loading, and lower packing fractions.⁵ A mixture of about 70 vol % coarse spheres and 30 vol % fine spheres is needed to obtain high packing fractions.^{2,6,7} The limiting smear density for binary packing is 86%, with about 82% achieved in practice. Ternary packing provides greater smear densities, with theoretical values ranging from 93 to 95%.

Sphere-pac technology was developed in the 1960–1990 period for thermal and fast spectrum reactors of nearly all types (U-Th and U-Pu fuel cycles, oxide and carbide fuels), but development of this technology was most strongly motivated by the need for remote fabrication in the thorium fuel cycle. The application to LWR fuels as part of the DOE Fuel Performance Improvement Program did not result in commercial deployment for a number of reasons, but the relatively low production cost of existing UO₂ pellet fuel is probably the most important factor. In the case of transmutation fuels, however, sphere-pac technology has the potential to be a lower-cost alternative while also offering great flexibility in tailoring the fuel elements to match the exact requirements of any particular reactor core at any given time in the cycle. In fact, the blend of spheres can be adjusted to offer a different composition for each fuel pin or group of pins in a given fuel element. Moreover, it can even provide a vertical gradient of composition in a single fuel pin. For minor-actinide-bearing fuels, the sphere-pac form is likely to accept the large helium release from ²⁴¹Am transmutation with less difficulty than pellet forms and is especially well suited to remote fabrication as a dustless fuel form that requires a minimum number of mechanical operations. The sphere-pac (and vi-pac) fuel forms are being explored for use as a plutonium-burning fuel by the European Community, the Russian Federation, and Japan.⁸⁻¹²

Sphere-pac technology supports flexibility in the design and fabrication of fuels. For example, the blend composition can be any combination of fissile, fertile, transmutation, and inert components. Since the blend of spheres can be used to fill any geometric form, nonconventional fuel geometries (e.g., annular fuels rods, or annular pellets with the central region filled with spheres)^{13,14} are readily fabricated using sphere-pac loading methods.

A project, sponsored by the U.S. Department of Energy Advanced Fuel Cycle Initiative (AFCI), has been initiated at Oak Ridge National Laboratory (ORNL) with the objective of conducting the research and development necessary to evaluate sphere-pac fuel for transmutation in thermal and fast-spectrum reactors. This AFCI work is unique in that it targets minor actinide transmutation and explores the use of a resin-loading technology for the fabrication of the remote-handled minor actinide fraction.

While there are extensive data on sphere-pac fuel performance for both thermal-spectrum and fast-spectrum reactors, there are few data with respect to their use as a transmutation fuel. The sphere-pac fuels developed will be tested as part of the AFCI LWR-2 irradiations.

This report provides a review of development efforts related to the fabrication of a sphere-pac rodlet containing surrogate fuel materials. The eventual goal of this activity is to develop a robust process that can be used to fabricate fuels or targets containing americium. The report also provides a review of the materials, methods, and techniques to be used in the fabrication of the surrogate fuel rodlet that will also be used in the actual LWR-2 irradiation specimen.

2. OVERVIEW OF SPHERE-PAC TECHNOLOGY

Vibratory consolidation of spherical granules (sphere-pac) or irregular granules (vi-pac) is the primary method for loading fuel pins with particle-bed fuels. Sphere-pac particles are produced by carefully regulated wet-chemical methods, whereas vi-pac particles are produced by crushing or milling. Both techniques use vibration to increase the density of the fuel during loading and multiple sizes to achieve a high packing fraction.

Peddicord et al. provide a review of the development of sphere-pac fuels up to 1986.² Other sources provide a more comprehensive summary of the methods used to produce spherical fuel particles, rod-loading techniques, quality assurance methods, and irradiation experience.³⁻⁵ All of these authors indicate that the use of sphere-pac fuels offers several advantages. The wet-chemical production methods for spheres require fewer mechanical steps than does pellet production,¹⁵ and no dust is produced. In addition, the fuel production and fabrication methods can be adapted to remote operations—an important consideration for use with minor actinide transmutation fuels. Irradiation test data show less fuel-clad mechanical interaction (FCMI) as compared with pellet fuel, which results in reduced strain and probability of pin failure by stress corrosion cracking. There is also evidence of reduced fuel-clad chemical interaction (FCCI). However, Peddicord et al. indicate that there is still an open question as to whether this advantage is maintained during rapid power transients at the beginning of fuel irradiation.²

Methods that have been used to produce all types and sizes of spheres include water extraction gelation, external chemical gelation, and internal chemical gelation.² A recent ORNL report¹⁵ describes

the internal gelation method in detail. To make the smaller-size particles, resin-loading methods have been used at ORNL for the production of ^{244}Cm targets¹⁶ and ^{233}U fuels.¹⁷⁻¹⁸ Resin-loading methods are proposed for production of the minor actinide spheres because of the simplicity of the process for remote operations and the very low level of waste generation.

Two techniques have been proven effective for loading particles in the fuel column: (a) infiltration of small spheres into a bed of large spheres with vibratory energy, and (b) simultaneous blending of the various particle size fractions during a single vibration-assisted loading operation. The blending option was developed in order to load long fuel pins in the fastest time for plant-scale applications. The infiltration method is commonly used in the laboratory and gives good results if certain guidelines are followed. For both types of fuel-pin loading, the inner diameter of the tube should be at least ten times the diameter of the largest particle diameter. In order to achieve good infiltration, the larger spheres must be held in place to prevent levitation of the bed by the smaller spheres. Multidirectional and variable frequency vibration is sometimes used to facilitate rod loading.² After loading, pins are inspected by nondestructive techniques (i.e., X-ray and gamma densitometry, autoradiography) to verify the uniformity of the fuel column.¹⁹

3. FUEL PIN DESCRIPTION

The fuel pins to be tested are based on the design used for the Fissile Material Disposition Program mixed oxide (MOX) and AFCI LWR-1 irradiations. A typical pin assembly, pin components, and pin and capsule assembly are shown in Figs. 1–3, respectively. The fuel pins will be fabricated with Zircaloy-IV, but the surrogate fuel pins will be fabricated from stainless steel in an equivalent pin geometry. A standard fabrication package for the surrogate fuel rod will be used to document material certifications and inspections required for the eventual LWR-2 pins.

The LWR-series test-fuel pins are nominally 8.09 in. long and have an outside diameter of 0.3810 in. Typical parameters of the LWR-series pin are provided in Table 1, along with the particular parameters for the surrogate rodlet and the LWR-2 rodlet. The fuel column will consist of two size fractions of spheres and will be about 5 in. long. The coarse fraction will consist of 500- to 800- μm spheres, while the fine fraction will consist of 50- to 80- μm spheres. The composition of the microspheres is described in the next section. A hafnia insulating pellet (identical to that used in LWR-1) is used on each end of the fuel column to hold it in place. The top of the column is held down by a spring, which also provides for a plenum region above the column.

Fig. 1. Sphere-pac pin assembly.

Fig. 2. Sphere-pac pin components.

Table 1. Sphere-pac pin parameters^a

	Material Parameters	
	Surrogate rodlet	LWR-2 rodlet
Cladding	316 stainless steel	Zircaloy-IV
Large spheres	HfO ₂	ZrO ₂ , UO ₂
Small spheres	Rare-earth oxide (Yb ₂ O ₃)	Am oxide and filler (UO ₂ , Zr)

^aCommon pin parameters: cladding O.D., 0.3810 in.; cladding I.D., 0.3295 in.; bond material, helium; fuel type, oxide; fuel smear density, 82%; fuel height, 5 in.

The fuel pin will be loaded with microspheres using specially designed equipment and procedures to control the loading. Documentation of the microsphere properties (size, density, composition, impurities), fuel column properties (smear density/uniformity), and pin-loading operations will be provided as part of the fabrication documents for both the surrogate rodlet and the LWR-2 rodlet. After loading, the cap will be welded into the pin, and the welded assembly will be helium leak checked. The pin will then be radiographed and inspected prior to shipment. The surrogate rodlet will be used to test interferences and troubleshoot other aspects of acceptance of the sphere-pac fuel form for irradiation in the Advanced Test Reactor (ATR). The actual LWR-2 rodlet will be received and inspected at Idaho National Laboratory (INL), and the pins will be enclosed in capsules provided by INL. The capsules are also based on the LWR-1 design. The capsule/pin assembly (Fig. 3) will then be loaded into a basket in preparation for insertion into the ATR. The basket will be fabricated by INL.

The nonfuel components of the LWR-2 rodlet are essentially identical to those used in the LWR-1 irradiation. Welding procedures for the LWR-1 rodlets were developed at Los Alamos National Laboratory in collaboration with ORNL experts. These same welding techniques will be deployed in existing facilities at ORNL designed for inert-atmosphere welding of transuranic (TRU)-containing packages for the actual TRU rodlets. Stainless steel welding procedures and capabilities to support the surrogate rod fabrication currently exist at ORNL.

The primary difference in this LWR-2 irradiation is the novel sphere-pac fuel form. Both the spheres and the fuel column will be characterized. The large spheres for the surrogate rodlet will be a commercial material (HfO₂). The impurities of this material will not be measured because they play no role in the function of this material as a surrogate. However, impurity information provided by the manufacturer will be used to evaluate the material's suitability for use in future irradiations. Because the purpose of surrogate rod fabrication is to prepare for fabrication of the actual fuel rodlet, the sphere characterization measurements listed in Table 2 will be performed for both the surrogate rodlet and the LWR-2 rodlet.

Table 2. Sphere characterization measurements

Property	Method for surrogate	Method for LWR-2	Status at ORNL
Impurities	Not applicable	ICP-AES ^a	Existing
Phase Content	Not applicable	X-ray diffraction	Existing
Size	Precision screening	Precision screening	Existing
Density, bulk	Tap density	Tap density	Existing
Density, skeletal	Helium pycnometry	Helium pycnometry ^b	Existing

^a ICP-AES – Inductively Coupled Plasma Atomic Emission Spectrometer.

^b Apparatus to be deployed in a glove box for the americium-containing material.

For those properties we do not propose to measure, the surrogate rod will serve as an exemplar for the actual LWR-2 measurement. Much of the sphere-characterization procedures have been developed in conjunction with the ongoing Advanced Gas Reactor (AGR) Program for analysis of the kernels in coated- particle fuel.

Testing for uniformity of material loaded into the fuel column will also be necessary. In the past this measure of uniformity was determined by gamma densitometry.¹⁹ We will use a more modern X-ray system that currently exists at ORNL to perform this type of density profiling. The ORNL X-ray system is shown in Fig. 4, and a sample X-ray image of an LWR rod obtained during preliminary testing is shown in Fig. 5. The surrogate rodlet will be used to begin the optimization of this equipment for density profiling of the fuel column.

Fig. 4. X-ray imaging system for density profiling in the sphere-pac fuel column.

Fig. 5. Sample X-ray image of LWR rod containing hafnia microspheres.

4. TEST MATRIX

The surrogate rodlet is intended to serve as a stand-in for all three types of sphere-pac pins proposed for irradiation: (1) an americium target, which consists of ZrO_2 and AmO_2 spheres; (2) a UO_2/AmO_2 fuel pin; and (3) a $\text{UO}_2/\text{AmO}_2/\text{Zr}$ fuel pin. The last option includes an addition of small metallic zirconium particles and is motivated by recent work at the Paul Scherrer Institute that indicates much better performance can be obtained with this “cermet” variant.²⁰

The compositions proposed for LWR-2 irradiation tests are shown in Table 3. All sphere-pac compositions are estimated based on the following assumptions: smear density, 82%; coarse fraction, 70 vol %; fine fraction, 30 vol %; coarse spheres, 95% theoretical density; and fine spheres, 60% theoretical density. The final LWR-2 compositions may need to be changed based on the analysis of their expected performance in the ATR. However, it is expected that the values given in Table 3 are reasonable estimates.

Table 3. Estimated compositions of sphere-pac pins

Sphere composition		Uranium enrichment (wt %)	Americium (wt %)	Mass (g)				Irradiation time (years)
Coarse	Fine			UO ₂	AmO ₂	ZrO ₂	Zr	
ZrO ₂	ZrO ₂ and AmO ₂	--	10	--	3.1	24.5	--	2
UO ₂	UO ₂ and AmO ₂	5	5	48.2	2.9	--	--	1
UO ₂	UO ₂ and AmO ₂	5	10	45.4	5.8	--	--	1
UO ₂	Zr and AmO ₂	5	10	39.6	5.5	--	3.6	2

5. COARSE SPHERES

The surrogate rodlet will contain commercially available HfO₂ microspheres in the 500- to 800- μ m size range (Brace GmbH). The coarse fraction for LWR-2 will consist of 500- to 800- μ m spheres of two different compositions: UO₂ (~5% ²³⁵U) and ZrO₂. Fabrication of UO₂ for the LWR-2 irradiation specimen will be carried out by procedures previously developed by ORNL and used for making depleted uranium kernels for coating studies in the AGR Program.¹⁵

6. PRODUCTION OF THE FINE SPHERES

Dense packing of a binary mixture of spheres is obtained only if the diameters of the two sphere fractions differ by at least a factor of 7 (ref. 4). A diameter ratio of 10 was adopted in this study to ensure that good packing will be obtained. The coarse fraction will consist of 500- to 800- μ m spheres produced by internal gelation, and the fine fraction will consist of 50- to 80- μ m spheres. For the smaller-sized spheres, a microsphere production method that offers advantages over the internal gelation process may prove beneficial. A resin-loading method has been successfully used for many years at ORNL for the preparation of actinide targets for heavy-element production.¹⁶ This method was chosen because of its simplicity and ability to produce the smaller-size fraction with almost no waste. The method consists of (a) ion-exchange sorption of the metal species (either americium or uranium) onto the resin, (b) heat treatment in oxygen (calcination) to produce a metal-oxide residue, and (d) densification of the metal oxide by sintering in a controlled atmosphere (usually 4% H₂). Because the initial resin particles are spherical, the residual particles after sintering are also spherical. The final density and size of the

particles depend on the initial size of the resin, the amount of metal sorbed on the resin, and the calcining and sintering conditions. It is usually not possible to produce fully dense spheres by this method, but densities somewhat above 50% of the theoretical limit can be made.

For our present project, three resins have been evaluated. Two are strong-acid cation resins containing sulfonate groups—DOWEX 50W-X8 resin and the macroporous equivalent Bio-Rad AG-MP-50. The third is a weak-acid cation resin, BioRex 70, based on carboxylate groups. The three are being evaluated with respect to potential impurities as well as the characteristics (e.g., size, density, porosity, crush strength) of the product spheres.

The strong-acid cation resins are known to provide relatively high loadings from acidic solutions but require a three-step calcination, produce corrosive off-gas products, and may suffer from sulfur contamination of the final oxide product. In this document we report on the weak-acid anion resins because they are not subject to this complication associated with sulfur.

The weak-acid resins that contain carboxylate groups can be easily calcined in just one step in air and produce a very pure oxide product. The potential problem is that they load only at near neutrality ($\text{pH} > 5$), and actinides will normally precipitate under these conditions. However, a test showed that successful loading of Yb^{3+} (stand-in for Am^{3+} and Cm^{3+}) on a weak-acid resin at $\text{pH} = 5$ could be achieved using an acetic acid/acetate buffer. During that test, ytterbium did not precipitate in the buffer solution and remained in solution. Under these conditions, very high loading was achieved. The behavior of americium and curium is expected to be the same, but a test will be performed to validate the process.

A batch of weak-acid resin (Bio-Rex 70 50–100 mesh) in the sodium form was procured. The resin particles were sieved into four size fractions, namely, $<150\text{ }\mu\text{m}$, $150\text{--}212\text{ }\mu\text{m}$, $212\text{--}300\text{ }\mu\text{m}$, and $300\text{--}330\text{ }\mu\text{m}$. Each fraction was converted to the acid form using 0.8 M HNO_3 . The resins were then pre-equilibrated in an acetate buffer at $\text{pH} 5.0$. Ytterbium oxide was dissolved in nitric acid and the resulting solution buffered with acetic acid and sodium acetate to $\text{pH} 5.0$. Complexation by acetate of the Yb^{3+} species prevented the precipitation and allowed for the resin loading. All four size fractions were loaded with ytterbium to breakthrough, dried, and calcined. Figure 6 shows the particle size distribution as determined by sieving of the ytterbium oxide particles obtained after calcination of the resin fractions. Calcination was performed in an air atmosphere under a temperature profile that held the particles for 2 h at 1250°C . The 300- to $330\text{-}\mu\text{m}$ resin fraction produced oxide particles of the desired size ($\sim 80\text{ }\mu\text{m}$). The particles will be characterized (microscope imaging, porosity, and surface area) and a larger batch produced in order to prepare the surrogate rodlet.

Fig. 6. Particle size distribution of the Yb_2O_3 produced by ion-exchange loading on Bio-Rex 70 resin and subsequent calcination.

7. PIN LOADING

The quartz “fuel rod” and glass beads shown in Fig. 7 were used to test the equipment and to provide visual evidence of the uniformity of the rod loading. A 9-mm-ID quartz tube was loaded with two sizes of glass spheres (80- and 800- μm diameter) by vibratory infiltration of the smaller-size fraction. A commercial accelerometer, shown in Fig. 8, was used to provide a selectable and stable vibration motion. The volume of the tube designated for sphere-pac loading was 10 cm^3 . The smear density was reproducible and measured to be about 83%—close to the theoretical limit.

A special test was also done to confirm uniform loading. A rod was segmented into 2.54-cm lengths and used to measure the uniformity of the loading. This uniformity test was conducted as follows: (a) the rod was segmented before loading, (b) the rod was reassembled as an integral tube joined by an adhesive, (c) the rod was loaded with spheres, and (d) the rod was carefully sectioned from top to bottom in such a manner as to preserve the underlying loading information. The results of this profiling method are provided in Table 4.

Table 4. Axial uniformity of sphere-pac in segmented-pin trial

2.54-cm pin-segment no.	Mass in segment (g)	
	80 μm	800 μm
1	0.76	2.14
2		
3	0.75	2.11
4	0.73	2.19
5	0.77	2.19

Fig. 7. Quartz “fuel rod” and glass beads used to test and optimize loading conditions.

Fig. 8. Accelerometer used to impart a reproducible vibration motion to load the pin.

The initial fuel-column loading experiments were conducted with a manual sphere-feeding system. A more automated system is needed to achieve a reproducible fuel column. An improved sphere-feeding system, shown in Fig. 9, was assembled using a low-speed variable motor, a flexible shaft, and a modified glass feeder. A follower rod, shown in Fig. 10, was fabricated that contains a mesh in the bottom. After loading the coarse fraction of spheres, this rod will be inserted into the top of the fuel pin. The fine fraction of spheres is introduced through the top of the follower rod. The rod holds the coarse fraction in place while allowing the fine fraction to infiltrate. Additionally, the follower rod can be used as an index gage for measurement of fuel height.

Documentation of the microsphere properties (size, density, composition, impurities), fuel column properties (smear density/uniformity), and pin-loading operations will be provided as part of the fabrication documents, as described in the preceding sections. After loading, the cap will be welded into the pin, and the welded assembly will be helium leak checked. The pin will then be radiographed and inspected prior to shipment.

8. SUMMARY

This report provides a review of development efforts related to the fabrication of a surrogate sphere-pac rodlet. The surrogate sphere-pac pin will be: (a) fabricated from 316 stainless steel, (b) filled with 500- to 800- μm HfO_2 spheres, (c) infiltrated with 50- to 80- μm Yb_2O_3 spheres, and (d) assembled and welded into a prototypical irradiation rodlet. The large spheres will be obtained from a commercial source, while the small spheres will be prepared at ORNL using a resin-loading/calcination process. All of the fabrication infrastructure has been established for the surrogate rodlet, and nearly all of the characterization capability exists at ORNL. A method for testing the uniformity of the fuel column with an existing X-ray instrument at ORNL will be pursued as part of the surrogate rodlet fabrication.

Fig. 9. Pin-loading equipment.

Fig. 10. Follower rod (lower left) along with fuel pin and screw feeder.

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