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Sphere-Pac Evaluation for Transmutation

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CONTENTS

ACRONYMS AND ABBREVIATED FORMS

AFCI	Advanced Fuel Cycle Initiative
EBR	Experimental Breeder Reactor
EFTTRA	Experimental Feasibility of Targets for Transmutation
ETR	Engineering Test Reactor
FCCI	fuel-clad chemical interaction
FCMI	fuel-clad mechanical interaction
FIMA	fissions of initial metal atoms
FUJI	fuel irradiations for JNC and PSI
JNC	Japan Nuclear Cycle Development Institute
LWR	light-water-reactor
MOX	mixed oxide
NRG	Nuclear Research and Consultancy Group
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Research Reactor
PIE	post-irradiation examination
PSI	Paul Scherrer Institute
Sphere-pac	spherical compaction
TREAT	Transient Reactor Test Facility
Vi-pac	vibratory compaction

ABSTRACT

The U.S. Department of Energy Advanced Fuel Cycle Initiative (AFCI) is sponsoring a project at Oak Ridge National Laboratory with the objective of conducting the research and development necessary to evaluate the use of sphere-pac transmutation fuel. Sphere-pac fuels were studied extensively in the 1960s and 1970s. More recently, this fuel form is being studied internationally as a potential plutonium-burning fuel. For transmutation fuel, sphere-pac fuels have potential advantages over traditional pellet-type fuels.

This report provides a review of development efforts related to the preparation of sphere-pac fuels and their irradiation tests. Based on the results of these tests, comparisons with pellet-type fuels are summarized, the advantages and disadvantages of using sphere-pac fuels are highlighted, and sphere-pac options for the AFCI are recommended. The Oak Ridge National Laboratory development activities are also outlined.

1. INTRODUCTION

Sphere-pac (and vi-pac) fuels, which have potential advantages as a transmutation fuel form, were evaluated in the United States and worldwide from ~1960 until 1990 for the purpose of plutonium-utilization in both thermal and fast reactors.[1-4] For minor-actinide-bearing fuels, the sphere-pac form is likely to accept the large helium-release from ^{241}Am 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. Sphere-pac (and vi-pac) fuel is being explored as a plutonium-burning fuel by the European Community, the Russian Federation, and Japan.[5–9]

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.

This report provides a review of development efforts related to the preparation of sphere-pac fuels and their irradiation tests. Based on the results of these tests, comparisons with pellet-type fuels are summarized, the advantages and disadvantages of using sphere-pac fuels are highlighted, and sphere-pac options for the AFCI are recommended. The ORNL development activities are also outlined.

2. OVERVIEW OF SPHERE-PAC TECHNOLOGY

Spherical compaction (sphere-pac) and vibratory compaction (vi-pac) are two methods for loading fuel pins with particle fuels. In the case of sphere-pac, the fuel is in the form of small spheres—often in two or three different size fractions. Vi-pac fuel consists of shards or debris that are produced by crushing and milling pellets. Both techniques use vibration to increase the density of the fuel during loading.

Peddicord et al. provide a review of the development of sphere-pac fuels up to 1986,[1] while refs. 2–4 provide a more comprehensive summary of the methods used to produce spherical fuel particles, rod-loading techniques, quality assurance methods, and irradiation experience. All of these authors agree 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.[10] Furthermore, no dust is produced. 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 it is still an open question as to whether or not this advantage is maintained during rapid power transients.[1]

Methods that have been typically used to produce spheres include water extraction gelation, external chemical gelation, and internal chemical gelation. Each of these methods is summarized in ref. 1. A recent ORNL report[11] describes the internal gelation method in detail. To make smaller, dense fractions, resin-loading methods have been used at ORNL, in particular for the production of ^{244}Cm targets and fuels (e.g., refs. 12–14). Such methods could also be used for the production of minor actinide spheres and have a special advantage because of the simplicity of the process for remote operations.

The smear density of typical light-water-reactor (LWR) pellets is 91–95%. To approach such densities with sphere-pac fuels, multiple fraction loadings are used. For optimum binary packing, the diameters of the two sphere fractions need to differ by at least a factor of 7. (ref. 2) Blending of spheres at smaller diameter ratios results in difficult blending and some segregation.[4] Additionally, a mixture of 70 vol % coarse and 30 vol % fine spheres is needed.[2,15,16] 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%. There are three options for loading rods with a ternary mixture: (1) blending all three sizes during loading; (2) sequential infiltration of the three sizes; and (3) a combination of the first two, in which the two larger sizes are preblended, followed by infiltration of the smallest size. Infiltration is the most common method used in laboratory work with short tubes. Blending during loading is potentially faster than infiltration and was successfully demonstrated at ORNL on long fuel rods to give high packing densities. ORNL infiltration tests with plutonium-bearing rods yielded smear densities ranging from 73.2 to 86.1% (ref. 2). For ternary packing, ref. 2 reports that the ratio of the tube diameter to the largest particle diameter, as well as the ratio of the successive diameters, must be about 10 to achieve optimum packing.

To perform infiltration, the larger spheres must be held down to prevent levitation of the bed by the smaller spheres. Multidirectional and variable-frequency vibration is sometimes used to facilitate rod loading.[1] After loading, pins are inspected by methods such as gamma densitometry to verify the uniform density of the loading.

3. IRRADIATION EXPERIENCE WITH SPHERE-PAC FUELS

Irradiation programs have often compared the performance of pellet and sphere-pac fuels. Three types of irradiations have been performed: LWR fuel in thermal reactors, fast fuel in thermal reactors, and fast fuel in fast reactors.[1] Table 1 provides a selected summary of past experiments with sphere-pac fuels. As available, the characteristics of the fuels that were irradiated (e.g., sphere size, smear density, fuel type) and the irradiation characteristics (e.g., linear power, temperatures) are provided. The majority of the irradiation experiments were performed in a thermal spectrum (even for the testing of fast fuels), with a limited number of experiments performed in a fast spectrum. Note that for fast fuels that are tested in thermal reactors, the heat source depression because of self-shielding of the thermal flux has to be considered when interpreting the results from these experiments. This depression results in lower centerline temperatures than would be seen for a fast spectrum.

3.1 ORNL SPHERE-PAC DEVELOPMENT EFFORTS

During the 1960s and early 1970s, an extensive program was undertaken at ORNL to develop oxide fuels for the Fast Breeder Reactor program.[4] As part of this effort, the use of sphere-pac fuels was investigated and compared with the use of conventional pellet-type fuels. After the completion of this program, an assessment was performed regarding the use of sphere-pac technology for the preparation of fast breeder reactor oxide and carbide fuels.[3] A similar report was prepared for thermal reactor fuels.[2] These reports provide a thorough review of the development program including methods to prepare spheres, rod-loading techniques, characterization of loaded rods, and test-fuel irradiation and performance. Also addressed were the considerations for remote fabrication and the scale-up of fuel fabrication operations. In general, these reports found that the sphere-pac fuels performed as well as or better than pellet fuels.

The irradiation tests performed by ORNL are described in refs. 2–4. The following discussion summarizes the major results from those tests. The irradiation program consisted of noninstrumented and instrumented thermal flux tests, transient tests, and fast flux tests. Fuels consisted of $(U,Pu)O_2$, and typically both sphere-pac and pellet fuels were irradiated for comparison. The results of these irradiations were described in terms of the thermal performance, fuel restructuring (i.e., sintering of the spheres into a more pellet-like form), actinide and fission product redistribution, FCMI, and FCCI.

Table 1. Summary of selected reported tests of sphere-pac fuels

	Sphere sizes (μm)							Fuel pin dimensions (mm)				Burnup						
Flux	Fraction 1	Fraction 2	Fraction 3	Smear density (%)	Fuel	Enrichment (% Pu) except as indicated	Cladding	Length	ID	OD	q' (W/cm)	(MWd/t)	FIMA	$T_{ctrline}$ (ºC)	T_{clad} (ºC)	Gas release (%)	Comments	Reference
Thermal	707–840	70–100		80	$(U,Pu)O_2$	4.6	Zircaloy-2	71.5	14.12	15.06	450–460			2,100				1
Thermal	800–1,110	125–200		72.7–76.6	$(U,Pu)O_2$	9.3–16.5	316 SS[a]	165	12	13	630–694			2,900			Irradiations 2 min to 10 h. Single infiltration method	2, 3
Thermal	1,000–1,250	<90[b]		77.3–77.8	UO_2	0	316 SS[a]	120–390	14.29	15.1	515–650	1,800–3,100			130–230		Single infiltration method	2, 3
Thermal	290–500	37–61		79.5–81.5	$(U,Pu)O_2$	2–18	Zircaloy-2 and 316 SS	200–890	6.4–11.36	7–12.76	410–920	500–15,000		~2,200	250–500	52–60	Cladding attack of zircaloy clad seen for some high-power-level samples. Attributed to hydriding and oxidation from impurities in the fuel	8
Thermal (transient test)				80–81	$(U,Pu)O_2$	20	304 SS	203	5.6	6.5	5,085–5,840 (peak)			1,800 (calculated)			Irradiated in TREAT	4
Thermal	300–600	<44		84	UO_2, $(U,PU)O_2$	15–20	304 and 316 SS	191	5.1–5.9	5.8–6.4	480–690		5–13.8		360–530		Irradiated in ETR. Active length of fuel column = 79 mm. Some rods at higher linear power density (1470 to 1640 W/cm) failed after reaching temperatures of 900 to 1000 ºC.	4
Thermal	1,000	100	10	84–89	UO_2, $(U,Pu)O_2$		Zircaloy-4	48.7	9.3	10.76	400–700	4,000–25,000				21	Sequential infiltration method, Pu incorporated into Fraction 1 only	5, 6
Thermal	630–1,000	33–100		81–82	$(Th,U)O_2$	(4.45% U, 90% enriched ^{235}U)	Zircaloy-4	276	9.31–10.53	10.75–11.77	560–600	44,700–50,300				41–51	Irradiated at KFA Jülich	9
Thermal and epithermal	700	60		77–80	(U,Pu)C	~15	316L SS	75–500	5.83–8	6.6–9	330–1,070		0.1–5.7		320–725	0.24–40		7
Fast	700	60		77–80	(U,Pu)C	~15	316L SS	75–500	5.83–8	6.6–9	640		6.4		640		Six of 15 pins failed because of local clad carburization	7
Fast	420–595	<25		79.71–86.10	$(U,Pu)O_2$	20 (93% enriched ^{235}U)	304 and 316 SS		5.5	6.4	400–525		6–12		570–600	80	Single infiltration method. EBR-II, encapsulated pins	4
Fast					$(^{235}U,Pu)O_2$	20	316 SS	1020	5.6	6.4	<460	100,000					EBR-II, unencapsulated pins. Fuel region 343 mm long	4

[a] Actually reported as material 1.4988, which is similar to 316 stainless steel.
[b] Nonspherical.

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The thermal conductance of a fuel pin consists of two components: fuel thermal conductivity and fuel-cladding gap conductance. The ORNL tests found the gap conductance of sphere-pac fuel at linear heat rates up to 16 kW/ft (525 W/cm) and burnups up to 6% fissions of initial metal atoms (FIMA) to be about twice that of pellet-fueled pins. In the case of the instrumented tests for irradiations in the Oak Ridge Research Reactor (ORR), the gap conductance in the pellet fuel was measured as 0.273 $W/(cm^2 \bullet {}^{o}C)$, while it was 1.93 W/cm^2C for the sphere-pac fuel. These higher conductances result in lower fuel-surface temperatures.

The ORNL tests showed that the primary mode of restructuring at high temperatures is a vaporization-condensation mechanism. The rate of restructuring was found to increase rapidly with increasing temperature and oxygen potential. Most restructuring occurs early in the life in the fuel. For fast flux irradiations, it was found that for rods that reach restructuring temperatures, the same structure as a pellet fuel rod develops—namely a central axial void is found with radial cracks emanating from the void. These cracks terminate at the outer edge of the restructured region, which is surrounded by a non-restructured annulus. The nonrestructured annulus was found to provide two benefits. (1) Lower stress is exerted on the clad (because spheres are able to move vertically), as compared with pellet fuel rods. (2) Reduced fission product transport to the cladding occurs because cracks in the fuel do not extend to the cladding. Therefore, no particular location exhibits a concentration of fission products. These two benefits should act to "improve fuel element performance and decrease cladding failures."[3] The fuel restructuring in the fast flux irradiations was found to be similar to that in the thermal irradiations.

For the sphere-pac fuels, some preferential movement of uranium down the temperature gradient in columnar grain-growth regions occurred. This resulted in regions that were depleted in uranium and therefore concentrated in plutonium (near the central void). Consequently, the fission heat generation rate increased in that area. For the fission products, there was a "distinct accumulation of molybdenum, cesium, and tellurium . . ." in the fuel adjacent to the cladding.[4]

FCMI can result from two phenomena: differential thermal expansion between the fuel and cladding and fuel volume changes resulting from fission product accumulation. For the ORNL tests, which had smear densities <85% and burnup < 50 GWd/t, no significant mechanical deformation was seen. In one case, a small amount of plastic deformation was seen for one of the transient tests conducted at the Transient Reactor Test Facility (TREAT). This deformation likely resulted from the high differential thermal expansion during a rapid power excursion. The results from the ORNL tests showed that sphere-pac fuels can reach high burnup with essentially no mechanical interaction between the fuel and the cladding.

FCCI has been observed in pellet fuels, with oxidation being the primary reaction. For the sphere-pac tests, the only significant chemical interaction seen was for some high-burnup thermal irradiations. But even for this test, the interaction was one-half that for a similarly irradiated pellet pin. The ORNL investigators reported that the evidence to date supports improved performance of sphere-pac fuels (as compared with pellet fuels) with respect to FCCI. It is suggested that the improved performance may result from better gap conductivity (and hence, lower clad temperatures), lower cladding stresses, and lower potential for localized chemical attack.[4]

Because, in many of the experiments, both pellet fuels and sphere-pac fuels were irradiated, comparisons were made between the results for the two technologies. For an Experimental Breeder Reactor–II (EBR-II) experiment that consisted of pellet, sphere-pac and vi-pac fuels all irradiated together in the same subassembly, after 4% burnup, a nondestructive examination showed no major differences among the rods. Both steady-state and transient tests showed that "sphere-pac oxide fuel should behave similarly to pellet oxide under fast breeder reactor operating conditions."[3] Tests comparing sphere-pac and pellet fuel in the ORR showed FCCI for the pellet fuel but none for the sphere-pac fuel. Similar results were seen for an irradiation in the Engineering Test Reactor (ETR) in Idaho.[3]

3.2 OTHER IRRADIATION EXPERIMENTS

As indicated in Table 1, a number of irradiation experiments have been performed on sphere-pac fuels in addition to the ORNL work. The major insights gained from each of these experiments are summarized in the following paragraphs.

Cervellati et al. irradiated $(U,Pu)O_2$ spheres in both a thermal and fast flux; however, only post-irradiation examination results from the thermal experiments were reported.[17] Cladding attack was seen for the higher-power-level samples (reaching up to 920 W/cm). This attack was attributed to hydriding and oxidation resulting from impurities initially present in the fuel. Cervellati et al. concluded that better control is needed over the fuel product specification to prevent the cladding attack.

Calza-Bini et al. performed a series of irradiation tests on $(U,Pu)O_2$ spheres.[18] Short-term tests (2 to 800 min) at linear powers up to 1000 W/cm were used to study the restructuring of the sphere-pac fuel. Post-irradiation examination (PIE) showed five distinct zones in the fuel from the center outward: (1) molten, (2) gross void, (3) densified, (4) modified, and (5) as-fabricated. The authors found that most of the microstructure changes occurred within the first 2 min of the irradiation.

Sens and Majoor performed thermal irradiations of pins with either spheres of $(U,Pu)O_2$ or UO_2.[19] They found low mechanical interaction between the fuel and cladding. Sens and Majoor measured the strain produced in sphere-pac and pellet rods after irradiation. The strain in the pellet rod was almost twice that of the sphere-pac rod. Strain relaxation occurred much more quickly in the sphere-pac rod. Therefore, they expect that the behavior of sphere-pac fuel will be better than that of pellets during transients, because of "reduced strain concentration processes."

Stratton irradiated (U,Pu)C spheres under fast, thermal, and epithermal flux conditions.[20] For the thermal irradiations, which had linear heat rates up to 1070 W/cm, 6 of 15 test pins failed because of local clad carburization.

Lahr et al. summarized the results of thermal irradiations of pins containing either $(U,Pu)O_2$ or UO_2 spheres.[21,22] For the tests with $(U,Pu)O_2$, short irradiations were performed. For pins with 76% smear density and pin power ~ 700 W/cm, they found that "substantial restructuring of the fuel takes place within 2 minutes." As part of this restructuring, a central channel was formed. Lahr et al. made a number of general observations about the sphere-pac fuel, which indicated certain advantages. "Uniform and relatively low mechanical interaction between the cladding and the fuel" was noted. Compared with pellet fuel, "axial dilations of length are small and more or less elastic." Ovality formation is reduced. Heat transfer between the fuel and the cladding is better for sphere-pac fuel than for pellet fuel. The authors noted that "after initial restructuring, the kernel [sphere-pac] fuel has an effectively higher thermal conductivity."

3.3 SUMMARY OF OBSERVATIONS FROM SPHERE-PAC IRRADIATION EXPERIMENTS

Peddicord et al. provided a comparative summary of sphere-pac fuels and pellet fuels for the types of effects that were observed during irradiation tests.[1] These effects are discussed in the following paragraphs and include restructuring, FCMI, migration of fuel constituents and fission products, and thermal response.

During irradiation, sphere-pac fuel can undergo significant restructuring in the center of the fuel pin, with an unrestructured annulus remaining at the fuel-clad interface, and sometimes the appearance of a void in the center of the pin. The presence of the unrestructured annulus acts to relieve stress. Additionally radial cracks do not reach the clad (because of the unrestructured region), which results in a more tortuous path for corrosive fission products to the clad. Because a "short-circuit" path does not exist in the fuel, concentrations of fission products at the clad tend to be more uniform in sphere-pac fuels. In general, with respect to restructuring, sphere-pac fuel behaves very similarly to pellet fuels with grain growth and the formation of a central core. However, an annular unrestructured zone remains.

With respect to thermal response, sphere-pac temperatures can be higher or lower than those for pellet pins, depending on the initial fuel-clad gap and operating history. The lower thermal conductivity of the unrestructured sphere-pac fuel may be somewhat offset by the lack of a fuel-clad gap. The thermal response in a sphere-pac pin is a "strongly damped system."[1] At higher temperatures, restructuring occurs, which increases conductivity and thereby lowers the temperature. The amount of fission gas release is dependent on the temperature of the fuel.

A final point of comparison was with respect to the potential for loss of fuel from a breached pin. For failed pins resulting from carburization,[20] no fuel was lost.[1] Hence, to prevent loss, sufficient sintering and necking of the spheres at the outer region of the fuel must have occurred. Out-of-core beginning-of-life simulations of a cladding breach showed that some fuel could be lost before initial startup. However, no loss would occur immediately after startup.[1] It should also be noted that a thermodynamic analysis performed in the AFCI program determined that the volatility of americium carbide would likely be very high. Thus, this compound is not a favorable fuel form for americium-containing fuel.[23]

Peddicord et al.[1] reviewed a number of models that have been developed regarding sphere-pac fuel performance. Modeling of the effective thermal conductivity evolved in an attempt to account for restructuring and heat transfer through the noncontact (gas) regions of the bed. Both theoretical and semiempirical methods have been used. Monte Carlo models have been developed to simulate the packing of beds. This has led to the derivation of a "coordination number," which is the average number of contacts for a sphere. Such numbers have been incorporated into thermal conductivity models. The mechanical response of the sphere-pac bed has been incorporated into restructuring models, which, in turn, affects the thermal conductivity.

Another important property for the thermal behavior of the sphere-pac fuel is the thermal conductance at the fuel-clad interface. As Peddicord et al. indicate, "estimates of sphere-pac gap conductance range from 60% to 300% of typical pellet pin values."[1] Based on experimental evidence, no open gap formed during steady-state or transient tests. Intimate contact occurs between the clad and the spheres. Four major approaches have been used to model the thermal conductance: (1) use estimated values that depend on whether the bed is two fraction or three fraction, (2) calculation based on empirical models, (3) use of a two-region model with a "temperature jump" immediately at the wall, and (4) assumption that a temperature jump due to gap conductance is negligible because of the good contact between the fuel and clad.

Models for fission gas release have been based on pellet-fuel models. In the restructured region, the behavior of sphere-pac fuel is essentially like that of a pellet. In the unrestructured region, temperatures are sufficiently low that little diffusion of gas atoms occurs. Retained gases result in swelling, and pellet models have also been applied to sphere-pac fuels with respect to swelling. Note that swelling in sphere-pac fuel can actually enhance restructuring.

Codes that have been developed to model the overall performance of sphere-pac fuels include COBRA-3SP, GAPCON-THERMAL-2, and SPECKLE.[1] Members of the modeling group at the Paul Scherrer Institute (PSI) have developed several models for sphere-pac fuels: SPHERE, PINTEMP, and SPACON.[24]

3.4 RECENT IRRADIATION EXPERIMENTS

More recent efforts to evaluate sphere-pac fuels are being performed by the FUJI (Fuel irradiation for JNC and PSI) project, which represents a collaboration of Japan Nuclear Cycle Development Institute (JNC), Nuclear Research and Consultancy Group (NRG), and PSI.[5–7] The FUJI project is investigating the performance of mixed-oxide (MOX) fuel pellets, as well as vi-pac, and sphere-pac forms, with respect to their use in fast reactors using stainless steel cladding. The authors recognize that particle fuels are good candidates for the fabrication of low-decontaminated fuels and minor actinide transmutation fuels because of remote handling requirements for these materials.

The FUJI project will study the early-in-life restructuring of the particle fuels by their irradiation in the pool-side facility of the High Flux Reactor at Petten. Four different irradiations will be performed[7]: (1) initial sintering test (0 to full power in 36 h); (2) restructuring test I (startup and then 48 h at steady state), (3) restructuring test II (startup and then 96 h at steady state); and (4) power-to-melt test (startup, followed by 48 h at steady state and then an increase in power until fuel center melting occurs). The maximum linear power for the first three tests is planned to be ≤550 W/cm, while that for the power-to-melt test is <900 W/cm.

For the sphere-pac fuel pins, filling tests were performed first with metal spheres and the MOX pins were then prepared.[6] MOX microspheres were prepared by the internal gelatin technique as described by Pouchon et al.[5] The MOX microspheres were nominally 20% Pu and 80% U. In addition, some neptunium-containing MOX microspheres, designated “Np-MOX,” were also prepared. The composition of the Np-MOX is nominally 20% Pu, 5% Np, and 75% U. Two filling methods were investigated[6]: infiltration and simultaneous. For infiltration loading, a coarse fraction (710–800 μm) is first loaded into the pin, followed by the infiltration of a fine fraction (63–75 μm). A smear density of about 82% was achieved. For simultaneous loading, the coarse fraction and a larger fine fraction (180–212 μm) are loaded at the same time. Simultaneous loading resulted in smear densities of about 73%.

Three of the four planned short-time irradiations have been completed, with PIE of the third test ongoing. Results from these tests are not yet available, and papers will be prepared as the work is completed.[25] The FUJI experiments should increase the understanding of the early restructuring behavior of sphere-pac fuels.

4. DISCUSSION AND RECOMMENDATIONS

The extensive experimental program that has been conducted for sphere-pac fuels has shown their advantages over pellet fuel forms in terms of the potential for improved fuel performance. Additional performance benefits are expected for transmutation applications, especially with respect to the ease of remote fabrication of minor actinide transmutation targets. Sphere-pac fuels have been shown to behave much like pellet fuels after restructuring. However, the former maintain an advantage with respect to FCMI and FCCI because of their ability to relieve stress in the nonrestructured zone and the lack of channeling pathways for fission products to accumulate at particular clad locations (thus limiting the development of corrosive concentrations at the clad). Such behavior is expected for both thermal and fast fuels.

While not as mature as pellet fuels, a significant effort has been undertaken to model the behavior of sphere-pac fuels during irradiation. These models have attempted to account for the thermal conductivity; gap conductance; and, importantly, restructuring of the fuel after irradiation. Consideration of these models and their underlying experimental work can lead to useful estimates of the thermal behavior of AFCI sphere-pac fuels.

For this AFCI program, the interest is in the development of a transmutation fuel for minor actinides. One characteristic of such fuels is the evolution of large amounts of helium during an irradiation, which results in pressures much larger than those typically encountered in LWR fuels. One of the recent transmutation tests for ^{241}Am has measured the production of helium, and this test can be used to project such production in a sphere-pac transmutation pin.[26] Note that the smear density of a sphere-pac fuel is less than that of a pellet fuel, and, at high burnup, an unrestructured zone remains well distributed along the fuel column. This zone may help accommodate the relatively large helium production in transmutation fuels.

The current development activities at ORNL for sphere-pac fuels center initially on reestablishment of the capability to remotely reload sphere-pac fuel pins. Follow-on work will be to load test pins with minor actinides (probably making up some portion of the fine-sphere fraction), irradiating these pins, and then performing PIE. These tests can then directly address the performance of the transmutation fuels, which is expected to be satisfactory based on the prior testing results for both thermal and fast sphere-pac fuels.

The recommended parameters for the initial irradiation investigation of a sphere-pac transmutation fuel are listed in Table 2. Such a test would be performed as part of the LWR-2 experiments and would thus be a thermal-spectrum irradiation. In summary, the fuel would consist of a two-fraction oxide bed. The coarse fraction would contain the uranium (and perhaps plutonium) fissile driver, while the fine

Table 2. Recommended (target) values of parameters for initial sphere-pac investigation[a]

Parameter	Value	Comments
Fuel form	MO_2 microspheres[b]	
Cladding material	Zircaloy-?	*Stainless steel and advanced claddings can be evaluated by analysis and tested after LWR-2*
Fuel smear density	82–86%	
No. of size fractions	2	*Use of three size fractions is considered only if demanded by further analysis (for loading or conductivity)*
Ratio of sphere diameters	7–10	
Irradiation spectrum	Thermal for 1st test	*Consideration of fast spectrum options to be done by analysis*
Limiting ramp rate		*Requires further analysis— defined by Advanced Test Reactor operating characteristics and pin design*
Large size fraction		
Radiochemical fabrication class	Semiremote	*Plant application similar to MOX with Pu/Np. If ^{235}U is used (no Pu or Np), standards for low-enriched-uranium fuel apply*
Diameter (μm)	500–800	
MO_2 volume fraction	70%	
Density	>95% theoretical density	
Fissile constituent	^{235}U or ^{239}Pu	*Plutonium-239 can be used at increased cost to the program*
Fissile level	5–10 atom %	
Nonfissile constituents		
Fertile	^{238}U	*This is the most widely tested, readily fabricated, and thoroughly modeled option*
Nonfertile option	Zr	*Use of Zr will require additional development work*
Transuranics (TRU)	^{237}Np	*Recently separated Np is assumed to minimize dose*
TRU level	4:1 Pu:Np	*As in previous tests, Pu:Np ratio of 4:1 is likely*
Small size fraction		
Radiochemical fabrication class	Fully remote	*Plant operation and maintenance is performed in hot cells*
Diameter (μm)	50–80	
MO_2 volume fraction	30%	
Density	>50% theoretical density	
Fissile constituent	^{235}U for LWR-2	*Plutonium-239 can be used at increased cost to the program*
Nonfissile constituents		
Fertile	^{238}U	*This is the most widely tested option and the most readily fabricated and modeled*
Nonfertile option	Zr	*Use of Zr will require additional development work*
TRU	^{241}Am	
TRU level	2–10%	*Levels expected to be bounded by EFFTRA test (high) and anticipated LWR series with pellets*

[a]Initial irradiation is assumed to be LWR-2: ATR thermal spectrum using the LWR-1 irradiation vehicle.
[b]M = U, Pu, Zr, Am, Np

fraction would contain the americium oxide, with a balance of uranium oxide (to achieve the desired loading). It is expected that the coarse fraction would be prepared by internal gelation, while the fine fraction would be prepared by resin loading. Zircaloy would be used as the cladding.

Based on the extensive irradiation programs that have been conducted, oxides have been shown to be a suitable form for sphere-pac fuels in terms of their thermal behavior, as well as FCMI and FCCI. In studies that have compared sphere-pac and pellet performance, the sphere-pac fuel in general has performed as well as or better than pellet fuels.

Cladding materials studied have included various zircaloy and stainless steel compositions (see Table 1). For one thermal zircaloy-clad experiment, cladding attack was seen at high powers. However, this attack was attributed to lack of tight control over the fuel specification, which resulted in impurities in the fuel.[17] Tests performed at KFA Jülich with zircaloy-4 cladding to burnups of 44,700 to 50,300 MWd/t showed no evidence of either FCMI or FCCI.[2]

Some concerns have been expressed about the performance of zircaloy-clad sphere-pac fuel during power transients. Knudsen et al. performed transient tests that compared pellet and vi-pac UO_2 fuel clad in zircaloy-2.[27] The vi-pac fuel consisted of sintered granules ranging from 45 to 4000 µm, with a smear density of 84%. Prior to the ramp tests, the rods were irradiated to 20,600 MWd/t. Ramp rates used in the tests were 50–60 W/cm•min. For the pellet fuel pins, the linear power was ramped from 330 to 470 W/cm and then to 720 W/cm, which resulted in immediate failure of the pin. For the vi-pac pin, the linear power was ramped from 390 to 640 W/cm, which resulted in failure after 7 min. Knudsen et al. concluded that "the performance of the vi-pac pin at the high-burnup stage was not essentially different from that of the pellet pin."

More recently, Kjaer-Pederson and Woods have performed ramp tests that compared pellet, annular pellet, and (three-fraction) sphere-pac particle fuels.[28] The pellet rods were not prepressurized, while some of the annular pellet rods and all of the sphere-pac rods were prepressurized (to 4.5 atm). The fuel was clad in zircaloy-2. Prior to the ramp tests, the fuels had been irradiated to burnups of 18,000–22,000 MWd/t or 28,000–32,000 MWd/t, depending on the number of irradiation cycles. Ramp rates were much more severe than those used in the Knudsen test, with values of either 600 or 1200 W/cm•min being used. The pellet and annular rods survived the various ramp tests. It was found that the sphere-pac rods "failed consistently when ramped to 520 W/cm or higher in a fast single-step ramp." These rods survived fast single-step ramps to 460 W/cm. In addition, sphere-pac rods that were first ramped to 460 W/cm and then, after a holding time, ramped to linear powers as high as 600 W/cm also survived. The failure of the sphere-pac rods was "attributed to high hoop stress in the cladding due to the absence of a fuel-to-cladding-gap." It should be noted that the sphere-pac rods had the lowest fission gas release of the three fuel types.

The failure of some sphere-pac rods during severe ramping conditions is cause for concern. However, the factors that play a role in such failure need further exploration. Does the number of fractions used, and hence the smear density, matter? Use of two-size-fraction spheres will result in a lower smear density. The many tests performed with two size fractions have consistently shown the ability of a sphere-pac fuel to relieve stress. Additionally, can some preconditioning of the fuel eliminate the potential for failure under severe transients? Once again, note that the rods that were fast ramped to 460 W/cm, held at that linear power, and then ramped to the higher power did not fail. The fuel, especially early in life, may be preconditioned to limit the stresses experienced in extreme transients. Of course, answers to these questions can be addressed only by further experimental testing of these ideas.

The concentration of ^{241}Am to be used in the target pins is estimated to be up to 10 wt %. This value would be consistent with that used in analyses for separations flow sheets and could also be compared with the EFTTRA-T4 (Experimental Feasibility of Targets for Transmutation) experiment in which 10-12 wt % ^{241}Am was irradiated. In the case of EFFTRA-T4, the targets were prepared by infiltrating $MgAl_2O_4$ pellets with americium nitrate solution, followed by calcination under Ar/H_2 at 700°C and then sintering at 1650°C (ref. 26).

Finally, the literature regarding development and evaluation of sphere-pac fuels can be divided into several major areas: thermal conductivity, fabrication, quality assurance, preparation of spheres by resin loading, testing, and transmutation. The bibliography included as an appendix to this report provides a list of references in each of these areas that were consulted in the preparation of this report.

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