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The addition of silicon carbide to surrogate nuclear fuel kernels made by the internal gelation process

R.D. Hunt [a,*], J.D. Hunn [a], J.F. Birdwell [a], T.B. Lindemer [b], J.L. Collins [b]

[a] Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6223, United States
[b] Harbach Engineering and Solutions, Dayton, OH 45458, United States

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ABSTRACT

The US Department of Energy plans to use the internal gelation process to make tristructural isotropic (TRISO)-coated transuranic (TRU) fuel particles. The focus of this work is to develop TRU fuel kernels with high crush strengths, good ellipticity, and adequately dispersed silicon carbide (SiC). The submicron SiC particles in the TRU kernels are to serve as getters for excess oxygen and to potentially sequester palladium, rhodium, and ruthenium, which could damage the coatings during irradiation. Zirconium oxide microspheres stabilized with yttrium were used as surrogates because zirconium and TRU microspheres from the internal gelation process are amorphous and encounter similar processing problems. The hardness of SiC required modifications to the experimental system that was used to make uranium carbide kernels. Suitable processing conditions and equipment changes were identified so that the SiC could be homogeneously dispersed in gel spheres for subsequent calcination into strong spherical kernels.

1. Introduction

The Deep-Burn Development Project of the US Department of Energy plans to use the internal gelation process in the production of spheroid transuranic (TRU) fuel particles, which will consist primarily of plutonium oxide with small fractions of americium, curium, and/or neptunium oxides. In addition, the TRU kernels may also contain an oxygen getter such as silicon carbide (SiC) or zirconium carbide (ZrC), which can react to form silicon dioxide (SiO_2) or zirconium dioxide with carbon (C). The SiC may fix palladium (Pd), rhodium (Rh), and ruthenium (Ru) as well. Another alternative under consideration is to directly coat the TRU kernels with a layer of SiC, which would serve as an oxygen getter. Other researchers at Oak Ridge National Laboratory are conducting similar exploratory studies on the application of a SiC or ZrC coating directly on the TRU kernels. In our tests on oxygen getters in the kernels, SiC was chosen rather than ZrC because the density of the latter ($6700\,kg/m^3$) is more than twice that of SiC ($3200\,kg/m^3$) thus making it easier to suspend the SiC particles in the feed solution for the internal gelation process. After the internal or external addition of SiC, the kernels will be used in the preparation of tristructural isotropic (TRISO)-coated fuel particles.

The present effort explored the use of the internal gelation process, which is an advanced production process for nuclear fuels [1]. This process combines an acidic metal solution with a weak organic base solution such as hexamethylenetetramine (HMTA). One of the solutions will also contain a metal complexing agent such as urea. After the solutions are chilled and combined, a stream of the cold mixture is transferred into a heated stream of water-immiscible liquid such as silicone oil. The feed stream becomes droplets as it enters the heated solution. As heat is transferred to the aqueous droplets, a precipitation reaction occurs, and the droplets are converted into gel spheres. After the gel spheres are collected and aged, they are washed to remove the water-immiscible liquid using trichloroethylene (TCE). Urea, HMTA, and ammonium nitrate (NH_4NO_3) are then removed using ammonium hydroxide washes. The gel spheres are then air dried. For some applications, no additional processing steps are needed while other spheres may require additional washing steps, aging, calcination, and sintering.

Previously, zirconia (ZrO_2), which was stabilized with approximately 8 mol% yttria (Y_2O_3), was used as a surrogate for the TRUs. Without the Y_2O_3, the ZrO_2 microspheres would crack at approximately 1000 °C when the crystal structure of ZrO_2 changes from monoclinic to tetragonal and its volume increases by approximately 9%. A minimum of 7 mol% Y_2O_3 is needed to provide a stable cubic crystal solid solution phase [2]. The plutonium (Pu) content of the proposed TRU kernel will be approximately 90 mol%; therefore, Pu chemistry and its gelation behavior should determine the nature of the gel spheres. Because they form only amorphous gel spheres, hydrous plutonium oxide and hydrous zirconium oxide [3–5] have shown similar cracking problems during calcination. Oak Ridge National Laboratory (ORNL) researchers encountered extensive cracking when they tried to calcine

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[*] Corresponding author. Tel.: +1 865 574 5481; fax: +1 865 574 6872.
E-mail address: huntrd@ornl.gov (R.D. Hunt).

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plutonium dioxide (PuO_2) kernels from the water extraction process [6] and the internal gelation process. The new wash techniques to prevent cracking of the zirconium/yttrium (Zr—Y) oxide gel spheres [7] should solve the cracking problem encountered with the Pu spheres.

In this internal gelation study, SiC powder was added to a Zr—Y or a HMTA/urea solution. A key advantage of the internal gelation process is the ability to homogeneously incorporate fine insoluble particles into the gel microspheres. For example, this process has been used at ORNL to prepare large quantities of dense, highly spherical uranium (U) fuel kernels with and without carbon [1,8,9]. The use of carbon powder required the use of a dispersing agent, Tamol SN, and an ultrasonic probe to minimize agglomeration of the carbon in the solutions. With the U microspheres, potential cracking problems can result from the decomposition of organics and NH_4NO_3. The crystal size of hydrolyzed uranium species, which is responsible for the formation gel network, must be adequately controlled to prevent cracking [10]. Larger crystals increase porosity and provide interconnecting pathways that enable the decomposing gases to escape to the surface of the microspheres and lessen the possibility of cracking. TRU microspheres are expected to encounter cracking problems unless nearly all of the organics and NH_4NO_3 are removed. Because the SiC did not appear to agglomerate as extensively as carbon and because any dispersing agent must be removed prior to calcination, the study focused on the incorporation of SiC into the gel spheres without the aid of a dispersing agent. Silicon carbide is extremely hard and is used as a grinding material in many applications; therefore, several modifications to the internal gelation system were required. These modifications and suitable processing conditions are discussed in this paper.

Since the SiC powder can lead to a plugged needle and other problems with internal gelation process, it is desirable to keep the amount of SiC to a minimum. Three factors affect the amount of SiC added per 1 mol of Pu in the TRU fuel. The initial O/Pu of approximately 1.7 is set by the $SiC + O_2 = SiO_2 + C$ reaction. The chemical potential of oxygen is nearly identical for both and maintains the carbon monoxide (CO) pressure below 1.01×10^5 Pa up to 1800 K [11]. During hydrogen reduction of the O/Pu to 1.7, it is possible that some SiC may be oxidized via the reaction $PuO_2 + 0.1\,SiC = PuO_{1.7} + 0.1\,SiO_2 + 0.1\,CO$. If the SiC provided all of the O/Pu reduction, 0.1 mol of SiC would be required. A second factor is gettering of oxygen released by fission. Gettering is desired to prevent CO pressures approaching 5.07×10^7 Pa at full burnup, which pressure would be added to the maximum of 2.43×10^7 Pa of Kr plus Xe. The design burnup of the Pu is approximately 60%. The oxygen release is calculated to be approximately 0.6 per fission for $PuO_{1.7}$ [11] so $0.5 \times 0.6 \times 0.6$ or 0.18 mol of SiC is required to getter this oxygen. A third factor affecting SiC content is possible sequestering of Pd, Ru, and Rh in the kernel. Interactions between SiC and fission products in the TRISO-coated uranium fuel particles have been studied extensively [12], and this work indicated that Pd may be retained in the TRU kernel as the 1335 compound ($PuPd_3Si_3C_5$) and thus prevented attack by the palladium on the SiC layer. The Rh and Ru may also be sequestered in a similar manner. The approximate sequester reaction is $PuO_2 + 3(Pd, Rh, \text{or } Ru) + 4SiC + C \rightarrow 1335 + SiO_2$. The moles of Pd, Rh, and Ru produced per mole of fissioned plutonium are 0.11, 0.07, and 0.18, respectively for a total of 0.36 mol. At 60% burnup, the amount of SiC needed per mole of plutonium would be $0.6 \times 1.33 \times 0.36$ or 0.29 mol. The sum of these three effects results in a total SiC requirement of 0.57 mol per mol of Pu.

The molar volume of SiC is about half that of PuO_2, and thus the final kernel would have about 22 volume percent SiC. This amount of SiC would have very little effect of the final diameter of the Pu kernel. For example, a 300 μm Pu kernel contains the same amount of Pu as a 321 μm Pu kernel with sufficient SiC for the gettering and sequestration.

2. Experimental

2.1. Preparation of stock solutions and the feed solution

Zirconyl nitrate hydrate (Aldrich Chemical Company) was dissolved in deionized water. Concentrated nitric acid (HNO_3) was used to dissolve yttrium metal (Morton Thiokol Inc.). When these solutions were combined, the zirconium, yttrium, and nitric acid concentrations were 1.056, 0.094, and 0.719 M, respectively, and the solution density was 1182 kg/m^3. The standard concentrations for the HMTA (Fisher Scientific) and urea (Fisher Scientific) in the other stock solution were both 3.2 M, and its density was 1140 kg/m^3. The SiC powder (<100 nm) was initially dry sieved with a 125 μm stainless steel sieve to remove any large agglomerated particles, which were more likely to plug the nozzle. Various amounts of SiC powder (Sigma Aldrich) were added to the Zr—Y solution with a pH of nearly 0 or to the HMTA/urea solution with a pH slightly above 10. A previous study [13] had shown that the maximum dispersion of SiC powder without a dispersing agent occurred at a pH of 10. However, the addition of the SiC powder to the HMTA/urea produced more foaming during the sample preparation than did the SiC addition to the acidic metal solution. The temperature of each solution was lowered and maintained near 273 K until the droplets were formed. The chilled solution with SiC was treated for 5 min period(s) with a Hielscher UP-200S Ultrasonic Processor to break up the SiC agglomerations.

In a typical experiment, 2–2.5 g of sieved SiC powder was added to 49.16 g of the HMTA/urea stock solution or to 118.2 g of the Zr—Y stock solution. The HMTA/urea solution at 273 K was slowly added in stages to the Zr—Y solution at 273 K to form the broth. After the solution was hand mixed, it was passed through a sieving sheet with 106 μm openings. Because of a noticeable difference in the amount of foaming with the different sample preparations, a larger amount of solids was removed from the feed that was prepared with the SiC addition to the HMTA/urea solution than from the feed that was prepared with the SiC addition to the acidic metal solution. The exact amount of SiC in the filtered feed solutions was not measured. The filtered feed solution was then transferred to a jacketed Pyrex broth pot that was mounted on a Cimarec 1 magnetic stirrer. The temperature of the broth solution was maintained near 273 K. A 1 in. "Starburst" stir bar was used to provide smooth uniform mixing of the SiC particles during each test.

2.2. Experimental apparatus and conditions

The experimental system was used previously to incorporate monosodium titanate (MST) powder into hydrous titanium oxide microspheres [14] and carbon powder into hydrous uranium oxide microspheres [8]. A gear pump was used to transfer feed solutions loaded with MST or carbon from the feed pot to the nozzle. However, the hardness of the SiC required several modifications to the experimental system. Typically, the gears in a gear pump are made of a softer material such as graphite or a polymer resin, and the SiC powder would rapidly erode the gears and render the pump ineffective. In the present apparatus, an air pressure system, rather than a gear pump, was used to transfer the feed with SiC from the broth pot to the nozzle. The vibrating system consisted of an ET-132-2 electrodynamic shaker with an SC-121 sine servo controller system (Labworks, Inc.) and a low-impedance accelerometer (Kistler Instrument Corporation). As shown in Fig. 1, the location of the shaker was moved from the transfer line to the nozzle in an effort to reduce the potential for plugging. While the use

Fig. 1. Modified internal gelation system for use with SiC.

of a vibrating nozzle is common [15], this modification required a new alignment system that could easily support the weight of the shaker. Third, the diameter and length of the transfer line between the broth pot and the shaker were reduced.

An air control system was developed to provide a constant pressure to the broth pot during a run. The system included a gas cylinder with a Medco low-pressure regulator (0–2.07 × 10^5 Pa) and a Bellofram Type 41-2 precision pneumatic regulator (0–6.89 × 10^4 Pa). The precision regulator was needed to provide sufficient control to make fairly monodispersed microspheres. The pressure was measured with an Ashcroft pressure gauge (0–1.03 × 10^5 Pa) on the inlet side of the Type 41-2 regulator and an Ashcroft pressure gauge (0–3.45 × 10^4 Pa) on the outlet side, which is followed by a Swagelok pressure relief valve set to relieve at 2.07 × 10^4 Pa. During each experiment, the supply pressure to the Type 41-2 regulator was set at 5.52 × 10^4 Pa. A pressure of 6.3 × 10^3 Pa to the broth pot was needed to deliver the broth through the 21 gage needle at a flow rate of 11.2 cm^3/min or at nozzle jet velocity of 0.97 m/s.

A stainless steel stand for the shaker was custom-designed for future use in a glove box. As shown in Fig. 1, the nozzle assembly was connected to the permanent magnetic shaker. The nozzle assembly consisted of a specially designed stainless steel needle from Cadence Science. The tip of the blunt-end needle was tapered with an orifice that is circular, burr- and crack-free, and mildly electropolished. The needle is attached to a male Luer-lock connector with a modified glass-tube fitting.

A 0.5 m long Tygon tube with an inner diameter of 0.318 cm was used as a feed line from the bottom of the broth pot to the nozzle. Because the nozzle is attached directly to the shaker, the tube must be lightweight and flexible so that it has little impact on the nozzle vibration. The relatively small inner diameter of the tube increased the feed velocity through the line and helped minimize the possibility of SiC settling in the transfer line.

The experimental conditions were selected to generate sintered kernels on the order of 300 μm in diameter. The small target diameter is needed to reduce self-shielding in the TRU kernels. The frequency of the shaker (Labworks, Inc.) was set at 221 Hz, and the feed rate was 11.5 cm^3/min. For the best results, the 21 gauge needle was positioned at a 35° angle to the veil of hot silicone oil. The jet stream of droplets was directed toward the bottom of the veil from 2 to 5 mm above the top of the gel-forming column. The goal was to provide a steady stream of droplets into the forming column with minimal turbulence. Laminar flow helps to minimize coalescing of the droplets before they are gelled. The gelation time for the broth droplets in the hot silicone oil in the gel-forming column was approximately 8 s at 365 K. The chemistry of this internal gelation process has been described elsewhere [16].

2.3. Aging, washing, and drying

During each experiment, the microspheres were collected in a stainless steel wire-mesh basket. At the end of each test, the basket was lowered into the reservoir with the hot silicone oil for 20 min. After the silicone oil was drained from the gel spheres, the basket was placed in a beaker with a magnetic stir bar and the microspheres were then washed with four successive batches of TCE. Each TCE (J.T. Baker) wash lasted for a minimum of 15 min. The same equipment used with TCE to remove the residual silicone oil was also used with 0.5 M NH_4OH to remove the NH_4NO_3, urea, and unreacted HMTA from the microspheres. Each NH_4OH wash lasted for a minimum of 30 min. The electrical conductivity of the spent NH_4OH solution was measured using an YSI model 3100 conductivity instrument. The washings continued until the electrical conductivity of the spent washes was below 1000 μΩ.

Next, several washes with deionized water were performed until the electrical conductivity in the spent water became constant. The conductivity of the deionized water was 0.7 μΩ. The gel spheres were then washed 5 times with Dowanol PM ($CH_3OCH_2CH(OH)CH_3$) for a minimum of 30 min each. The microspheres treated with Dowanol PM (Aldrich Chemical Company) were placed in a small beaker and covered with deionized water. The microspheres were then placed in a Teflon container with thick walls, and more water was added to the Teflon container to ensure that the microspheres remained covered with water during the heat treatment. After the Teflon container was sealed, it was placed in a programmable oven (Barnstead-Thermolyne). All pressurized water treatments in this study were done at 473 K for 3 h. Then, the treated microspheres were washed with deionized water until the conductivity became constant. Because the Dowanol PM should leave the gel spheres faster than water, the microspheres were treated once more with this solvent.

After the final wash with Dowanol PM, the microspheres were permitted to dry under ambient conditions for a minimum of 24 h. The microspheres were rolled across a stainless steel pan positioned with a slight slope to permit the identification and removal of any nonspherical microspheres. A slit was cut along one of the pan walls so that the defective microspheres could be pushed into a second pan.

2.4. Heat treatment and subsequent analysis

Small samples of the microspheres with the different SiC loadings were placed in a Harrop model ST-736 differential thermal analyzer (DTA)/thermogravimetric analyzer (TGA). The standard ramp rate was 100 K/h until 663 K and then 200 K/h until 1438 K under flowing 4% hydrogen which is needed to reduce oxygen reactions with SiC.

The partially sintered microspheres were then inspected using the digital microscope to determine the relative amount of cracking. The diameters of several intact microspheres were measured. Then, the crush strengths of individual microspheres were measured using a custom-designed apparatus that could apply up to 1.200 kg to an individual microsphere. Finally, select samples were ground to produce a polished plane at the base of the hemisphere to permit the distribution of SiC in the kernels to be examined.

3. Results and discussion

At the conclusion of each successful heating to 1438 K, the partially sintered kernels were examined for ellipticity, particle size distribution, and crush strength. Previously, the addition of carbon black to hydrous uranium oxide spheres (up to 1.3 mol of carbon per mole of uranium) had little or no impact on the ellipticity of the microspheres [8]. Because the mass and particle sizes of the carbon black used in the earlier work are comparable to those for the SiC in this study, no significant problems with ellipticity were expected. In these SiC tests, the nonspherical kernels were separated from the spherical kernels by rolling the particles down a slight incline. After suitable process parameters for the internal gelation process were identified and used, this rolling process typically eliminated <7 wt.% of the SiC kernels. Then, the ellipticity of the remaining kernels was evaluated. The average kernel ellipticty was 1.014, and only 0.3% of the kernels had ellipticities >1.06. For comparison, the current United States standard for spherical fuel particles requires that 10% or less of the kernels have ellipticities >1.06. When the recommended process parameters were implemented, <0.2% of these kernels had irregular surfaces during the kernel inspection. In contrast, nearly 5% of the kernels from our initial SiC tests had very irregular surfaces that would pass the rolling test. Fortunately, nearly all of these irregular kernels had diameters, which were twice as large as those of the typical, spherical kernels. Therefore, these large, irregular kernels could easily be removed with additional sieving after the sintering process.

The particle size distribution with Zr—Y—SiC kernels created via the pneumatic system was expected to be wider than the distribution of the Zr—Y kernels made using a gear pump with no SiC [7]. The gear pump provided a more uniform flow of the broth to the nozzle. After the Zr—Y spheres were heated to 1438 K in oxygen, the average diameter of these partially sintered kernels was 288 μm, with a standard deviation of 11 μm. The kernels were between 271 and 315 μm in diameter. The average diameter of the Zr—Y—SiC kernels was 293 μm, with a standard deviation of 22 μm. The Zr—Y—SiC kernel sizes were between 249 and 332 μm. Nearly 50% of the SiC kernels were between 282 and 304 μm while 20% of kernels were outside of the 271–315 μm range achieved for the Zr—Y kernels without SiC. Therefore, use of the pneumatic system doubled the width of our particle size distribution. This distribution may be noticeably reduced with additional operating experience. During the formation of the gel spheres with SiC, a strobe light for enhanced observation and small pressure changes with a precision regulator were utilized in an effort to maintain a constant droplet size. However, changes in the solution height in the broth pot and the viscosity of the broth apparently led to the wider particle size range. If the kernels with SiC are converted into TRISO-coated particles, then the coating operations must accept a wider particle size range or the production rate from the internal gelation procedure will drop by a maximum of 20%.

In our previous study [7], Zr—Y kernels with no cracking and crush strengths >1.200 kg per sphere were prepared using a combination of water washes, pressurized water washes, and Dowanol PM washes. It is plausible that the water washes are more effective at the removal of water soluble compounds and the Dowanol PM washes target organics. Even though SiC is a very hard compound, the presence of SiC should not improve the strength of the SiC-loaded kernels. With 5 min of ultrasonic dispersion of the 0.002 kg of SiC in the Zr—Y solution and our recommended wash procedure [7], 35% of the partially sintered kernels failed before a crush strength of 1.200 kg per sphere. In an effort to improve the crush strengths, the ultrasonic dispersion of the SiC was increased from a single 5 min period to two 5 min periods and the Dowanol PM washes were performed before and after the pressurized water treatment. This additional SiC dispersion time reduced the failure rate at 1.200 kg per sphere to 4%, and the average crush strength increased to nearly 1.200 kg per sphere. In addition, no irregularly shaped kernels were detected during the visual inspections. A cross section of a polished Zr—Y kernel loaded with SiC is shown in Figs. 2 and 3; the darker regions contain SiC while the lighter regions consist of Zr and Y. The SiC content in the polished kernels was approximately 0.44 mol of SiC per mole of metal. The amount of SiC in the kernels was confirmed using X-ray diffraction, and no evidence of SiO_2 as found as expected. The largest SiC particles appeared to be 2 μm in diameter, and the SiC was evenly distributed throughout the Zr—Y kernel. Similar, quality Zr—Y kernels with SiC were also produced when SiC was dispersed in the HMTA/urea solution using the ultrasonic probe for two 5 min periods.

When the SiC concentration was increased to approximately 0.55 mol of SiC per mole of metal, the addition of the SiC to the HMTA/urea or the acidic metal solution became important to the quality of the Zr—Y kernels regardless of the subsequent wash procedure. When the SiC was added to the HMTA/urea solution, the resulting kernels using both wash procedures had a failure rate of 4% before 1.200 kg per sphere; the average crush strength was 1.200 kg per sphere. In sharp contrast, when SiC was added to the acidic metal solution, the resulting kernels from both wash procedures had a failure rate of 52–56% before 1.200 kg per sphere, and the average crush strengths ranged from 1.070 to 1.100 kg per sphere. The initial solution for the SiC powder addition could play an important role in the final performance of the Zr—Y kernels for

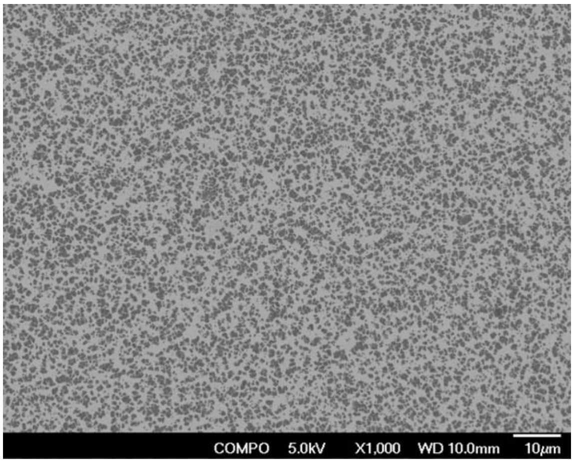

Fig. 2. Backscattered electron image of a polished cross section of a Zr—Y—SiC kernel.

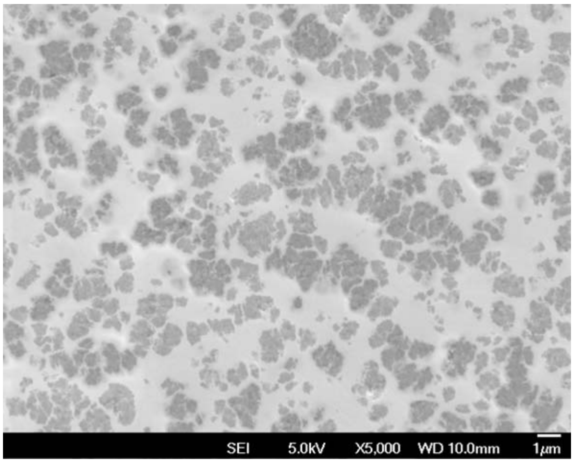

Fig. 3. Secondary electron image of a polished cross section of a Zr—Y—SiC kernel.

two reasons. First, based on the pH of the solutions, the SiC powder should be better dispersed in the HMTA/urea solution than in the acidic metal solution. A better initial dispersion should result in a better dispersion in the feed. Second, as a result of the foaming problem after the SiC addition to the HMTA/urea solution and the filtration of the feed solution, the SiC concentration in the kernels from the SiC addition to the HMTA/urea solution may be lower than expected. Increases in the SiC concentration can be expected to eventually lead to higher failure rates for the kernels.

4. Conclusions

One option under consideration for TRUs in spent nuclear fuel is to use the TRUs in the production of TRISO-coated fuel particles. This TRU fuel would then be used in a high-temperature gas-cooled reactor until a burnup of 60% is achieved. Under this scenario, the internal pressure for each particle could easily exceed 5.07×10^7 Pa at 1400 K. Therefore, it has been proposed to use SiC or ZrC as an oxygen getter and a potential sequestering agent for palladium, rhodium, and ruthenium. The maximum SiC requirement for gettering (initial plutonium reduction and reactions during fission) and sequestering is 0.57 mol of SiC per mole of initial plutonium. When Zr—Y is used as a surrogate for the TRUs, this level of SiC powder has been successfully added to gel spheres that were prepared using the internal gelation process. If the SiC powder is added to the HMTA/urea solution and adequately dispersed using an ultrasonic probe, then strong Zr—Y kernels loaded with SiC can be prepared without the use of a dispersing agent. In the tests with 0.44 mol of SiC per mole of metal, stronger kernels were produced when most of the Dowanol PM washes were performed before pressurized water treatment. Because of the hardness of SiC, a pneumatic system instead of a gear pump was used to deliver the broth as droplets to the gelation column. The particle size distribution of the Zr—Y—SiC kernels from the pneumatic system was double that of the Zr—Y kernels produced using the gear pump system.

Since SiC powder could be adequately dispersed in Zr—Y kernels with limited problems, preparations are underway to incorporate SiC powder into Pu microspheres and 90% Pu/10% Np microspheres. If these TRU kernels with SiC can be fabricated successfully, then small fractions of Am and Cm (<10%) may also be added to Pu microspheres.

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