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Preparation of $UC_{0.07-0.10}N_{0.90-0.93}$ spheres for TRISO coated fuel particles

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ABSTRACT

The US Department of Energy is considering a new nuclear fuel that would be less susceptible to ruptures during a loss-of-coolant accident. The fuel would consist of tristructural isotropic coated particles with dense uranium nitride (UN) kernels with diameters of 650 or 800 μm. The objectives of this effort are to make uranium oxide microspheres with adequately dispersed carbon nanoparticles and to convert these microspheres into UN spheres, which could be then sintered into kernels. Recent improvements to the internal gelation process were successfully applied to the production of uranium gel spheres with different concentrations of carbon black. After the spheres were washed and dried, a simple two-step heat profile was used to produce porous microspheres with a chemical composition of $UC_{0.07-0.10}N_{0.90-0.93}$. The first step involved heating the microspheres to 2023 K in a vacuum, and in the second step, the microspheres were held at 1873 K for 6 h in flowing nitrogen.

1. Introduction

A new nuclear fuel, tristructural isotropic (TRISO) coated particles with dense uranium nitride (UN) kernels, has been proposed that would be less likely to fail during a loss-of-coolant accident. Since UN has a very high fuel density, it has been studied extensively for use in space power reactors and liquid–metal-fast-breeder reactors. Initially, UN was synthesized from uranium metal by the hydride–dehydride–nitride process [1,2]. The carbothermic-reduction and nitriding process, which starts with a mixture of uranium oxide and carbon, was developed for UN production [3,4]. This process has been used with powders [5], pellets [6,7], and microspheres from gelation processes [8–13]. The use of uranium powders and pellets can be problematic due to insufficiently homogeneous mixing of uranium oxide and carbon and due to relatively high level of metal impurities. The internal gelation process, which can produce kernels with low levels of metal contaminants, can homogeneously incorporate fine insoluble particles such as carbon throughout the gel spheres. At the end of the carbothermic-reduction and nitriding process, the UN microspheres still have a relatively low density, so they are suitable for direct pressing. A sintering step is needed to generate dense UN fuel kernels, which must be near theoretical density, for the TRISO coated fuel particles.

The internal gelation process has been used at Oak Ridge National Laboratory (ORNL) to make large quantities of uranium kernels with and without carbon [14–16]. Two studies on UN microspheres [9,11] demonstrated similar pathways for the conversion of uranium dioxide (UO_2) microspheres with enough carbon to make $UC_{1-x}N_x$ with x approaching unity. Both studies ultimately recommended conversion to the UO_2–C microspheres to UO_2–uranium carbide (UC) and then conversion of the latter into UN. Other studies [17,18] detailed the T-x-pN2 characteristics of the UC–UN solid solution and the intermediate compositions encountered in converting UC to nearly pure UN. These efforts [9,11,17,18] illustrated that conversion temperatures and pressures below 1723 K and 101.3 kPa, respectively, can lead to nearly pure UN formation via reaction (1).

$$UO_2(s) + 2C(s) + 0.5N_2(g) \rightarrow UN(s) + 2CO(g) \quad (1)$$

At temperatures above 1723 K, it was recognized that the inclusion of hydrogen in the reactant gas results in formation of HCN [11], which affects the conversion, as shown in reaction (2).

$$UC_{1-x}N_x(s) + 0.5(1-x)H_2(g) + (1-x)N_2(g) \rightarrow UN(s) + (1-x)HCN(g) \quad (2)$$

However, no quantitative details were provided concerning any effects of the use of hydrogen. In Fig. 1 of an earlier paper [11], the carbon monoxide (CO) profile was analyzed to demonstrate that by the time the reaction temperature of 2023 K was attained in flowing argon, the sample had indeed been converted to a UO_2 + UC mixture before N_2 was introduced to complete the conversion to UN at 2023 K.

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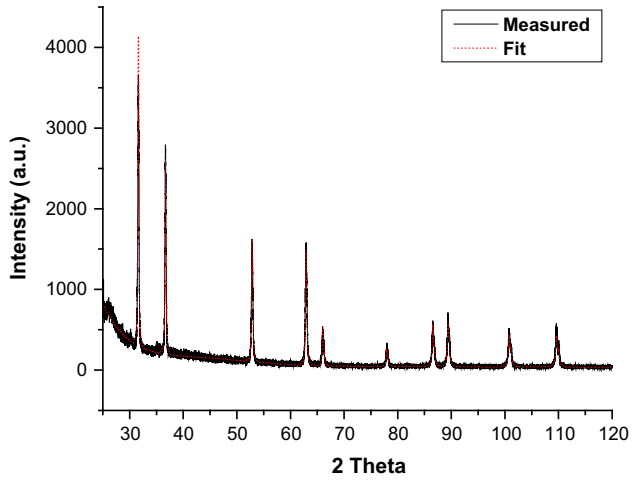

Fig. 1. Powder XRD pattern of $UC_{0.07-0.10}N_{0.90-0.93}$ sample. Note the following for the peak positions for a few of the chemical phases that are related to this UN synthesis route. UN (space group Fm-3 m): 31.7°, 36.8°, 53.0°, 63.1°, and 66.3°. UO_2 (space group: Fm-3 m): 28.3°, 32.8°, 47.1°, 55.8°, and 58.8°. UC (space group: Fm-3 m): 31.3°, 36.3°, 52.2°, 62.1°, and 65.2°. UC_2 (space group: I4/mmm): 29.5°, 29.9°, 36.2°, 47.6°, and 52.1°.

A modification of this process was investigated in this effort. In the first step, vacuum was used to first convert UO_2 and carbon to form the mixture of UO_2, UC, and uranium dicarbide (UC_2) at temperatures up to 2023 K. This step efficiently removed 2/3 of the CO produced by the overall process. The second step is the conversion to UN, which can be described by the reactions (3) and (4). This step only took 6 h at 1873 K.

$$UO_2(s) + 2UC(s) + 1.5N_2(g) \rightarrow 3UN(s) + 2CO(g) \quad (3)$$

$$UO_2(s) + UC_2(s) + N_2(g) \rightarrow 2UN(s) + 2CO(g) \quad (4)$$

With one exception [19], very little development work on UN microspheres has occurred in the past two decades. During the interim, key reactants such as carbon blacks and the dispersing agents have changed, and several process improvements for the incorporation of fine particles into gel uranium spheres have occurred. Therefore, new reactants and recent process improvements were applied to the production of uranium spheres with carbon. In addition, the new two-step heating profile to form UN microspheres was successfully demonstrated. Optimization of the UN conversion process and sintering to 650 or 800 μm will be presented in detail in a subsequent paper, which also discusses faster reaction kinetics due to the use of well dispersed carbon nanopowders.

2. Experimental

2.1. General

The preparation of uranium gel spheres with adequately dispersed carbon black involved several steps. First, stock solutions of acid-deficient uranyl nitrate (ADUN) and urea–carbon-dispersing agent-hexamethylenetetramine (HMTA) were prepared. Inductively coupled plasma mass spectrometry has shown that the ADUN is relatively free of metal contaminants as compared to traditional powder techniques. The contaminants include nickel, aluminum, calcium, copper, chromium, iron, and magnesium. While the average nickel contamination was 20 μg/g, the average for the other metal impurities was less than 10 μg/g. Urea is a metal complexing agent, while the carbon dispersing agent is Tamol™ SN. Next, the solutions were chilled to 273 K, combined, mixed, and placed into internal gelation equipment. The chilled feed solution was passed through a vibrating nozzle to form uniform droplets, which were then transferred into flowing silicone oil at 335–337 K to cause gelation to occur. After the microspheres were collected, they were aged in the hot silicone oil for 15 min. They were washed with trichloroethylene (TCE) and then with 1.5 M ammonium hydroxide (NH_4OH). It must be noted that water and 0.5 M NH_4OH could not be used without significant carbon losses to the wash solutions. Since the C/U molar ratio should be precisely controlled, solutions that would leach carbon from the gel spheres were not used. The diameters of several of the washed microspheres were measured using a Motic B5 Professional Series digital microscope. After microspheres were air-dried, 2–3% of the microspheres was rejected as nonspherical, and the remaining microspheres were sieved. The air-dried microspheres were then calcined and converted to UN.

2.2. Preparation of stock solutions and the feed solution

The standard concentrations for the HMTA (Fisher Scientific) and urea (Fisher Scientific) in the stock solution were 3.2 and 3.2 M, respectively. After a stock solution was prepared and filtered, the density was measured to be 1.14 kg/m^3, and the pH ranged from 8 to 9. The pH of the 50 g of Cabot Mogul L carbon black in 1 L of water is between 4 and 11, while the pH of a 2% solution of Tamol™ SN is 9.4. Since the pH values of Mogul L and Tamol™ SN were much closer to the pH of the HMTA-urea stock solution than the pH of 1.05 for the ADUN stock solution, the Tamol™ SN and then the Mogul L were added to the HMTA-urea solution in the same manner as earlier studies [15,16].

The Mogul L carbon black was passed through a 170 mesh sieve so all particles with a diameter greater than 90 μm were excluded. According to Cabot Corporation, Mogul L Carbon Black has a surface area of 138 m^2/g and a particle size of 24 nm when it is adequately dispersed. After 0.6 g of Tamol™ SN was dissolved into 113.2 g of the HMTA/urea solution, then 7.3–7.9 g of Mogul L was added to the solution, and the C/U molar ratios ranged from 2.5 to 2.7, which was used in a comparable study [10]. A few experiments had C/U molar ratios between 2.1 and 2.2, which was common in most of the earlier internal gelation studies [8,9,11–13]. Carbon was then dispersed for 5 min at room temperature using a Hielscher UP200S ultrasonic probe. The HMTA/urea and ADUN solutions were chilled at 273 K for a minimum of 30 min. The 159.3 g of ADUN solution had a uranium concentration of 2.87 M and a nitrate to uranium molar ratio of 1.8. The chilled solutions were combined into the feed solution, and 2.2 g of water was added to the feed solution to aid in the transfer. The chilled feed was mixed by hand and transferred into the internal gelation system.

2.3. Experimental apparatus and conditions for the internal gelation

The experimental system, which is described in detail elsewhere [20], is a modification of an apparatus used to produce uranium and plutonium microspheres in a glove box. The use of carbon black required a few minor modifications. First, a chilled glass feed vessel was used to observe and ensure that the feed solution was adequately mixed throughout the experiments. The transfer line was located close to the bottom of the glass vessel. Second, the glass feed vessel was placed on a magnetic stir plate so a magnetic stir bar could be used to keep the carbon black suspended. Third, the position of the Micropump gear pump was changed so the broth solution was pumped from the feed vessel through the gear pump to the accelerometer of the vibrator. Due to potential gelation of the feed in the gear pump head, the head was also cooled to 273–278 K.

The conditions were selected to generate sintered kernels that were 650 or 800 μm in diameter. The system had previously been

used to generate microspheres that became 350 or 500 μm UO_2 fuel kernels. For the 650 μm kernels, a 17 or 18 gage electropolished stainless steel needle with a beveled tip (Cadence) was used. For the 17 gage needle, the frequency of the accelerometer (Labworks Inc.) was set at 57 Hz, and the feed pump was set to deliver 24.45 cm^3/min. These settings produced droplets with an average diameter of 2280 μm. For the 800 μm kernels, a 16 gage needle was used. The accelerometer frequency was 38 Hz, and the feed pump delivered 29.3 cm^3/min. The average diameter of droplets from these settings was 2825 μm. The frequency and the flow rate of the feed were varied as needed. The droplets were directed into a veil of hot silicone oil at the top of the gel-forming column. The droplets then flowed down the column. The exact gel time was not measured because the droplets and the gel spheres were both black.

The temperature of the gelation column was potentially another key variable. In nearly all of the experiments in this study, the temperature of the gelation column was maintained from 335 to 337 K. This temperature range promotes the formation of the largest uranium crystals, which increases the porosity of the microspheres and aids in the release of gases during the subsequent heat treatment. An earlier effort [21] demonstrated that the uranium crystal size drops dramatically when the gel temperature is increased from 333 to 343 K. Then, the uranium crystal size increases as the gelation temperature is increased from 343 to 353 to 363 K. In our experiments, the transition for the crystal size occurred between 337 and 339 K, as demonstrated by the bulk density of the air dried microspheres. The gelation temperature as well as the size of the droplets plays significant roles in the time required for gelation. As expected, the lower gelation temperature and larger droplets increased the time for gelation. In order to generate the most spherical microspheres, the gelation should occur before the microspheres leave the gelation column. With our experimental system, the gelation time was quick enough even at gelation temperatures of 335 to 337 K.

2.4. Aging, washing, and drying

During an experiment, the microspheres were collected in the wire-mesh basket. At the end of a test, the basket was lowered into the reservoir with the hot silicone oil for 15 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 washed four successive times with TCE to remove the residual silicone oil. Using the same equipment that was used in the TCE washes, 1.5 M NH_4OH was used to remove the ammonia nitrate, urea, and unreacted HMTA from the microspheres. Each NH_4OH wash lasted for a minimum of 15 min. The electrical conductivity of the spent NH_4OH solution was measured using an YSI 3100 conductivity instrument. The washings continued until the electrical conductivity of the spent washes was approximately 1.4 m℧. Then, the diameters of 20–30 washed gel spheres were measured using the Motic digital microscope. Finally, the microspheres were transferred to a stainless steel pan and spread out into a single layer. The excess NH_4OH was decanted, and the microspheres were permitted to dry for a minimum of 48 h.

2.5. Subsequent analysis and heat treatment

The relative crush strength of individual microsphere was estimated using a custom-designed apparatus. The apparatus consists of a Mettler PM1200 balance and a stainless steel rod, which is attached to a micrometer. A microsphere was placed on the balance, and the rod was slowly lowered on top of the microsphere. As long as the particle did not break, the weight registered by the balance would gradually increase up to 1.200 kg. Once the microsphere broke apart, the weight shown on the balance would drop, and the highest observed weight was recorded. This weight and the crush strength of the microsphere should be directly related. Select samples were ground to produce a polished plane at the base of the hemisphere to permit the distribution of the carbon in the air-dried and heat-treated microspheres to be examined. These samples were analyzed using X-ray diffraction (XRD) and a scanning electron microscope (SEM). The SEM images of air-dried and calcined microspheres did clearly indicate that the carbon was adequately dispersed throughout the microspheres and that no changes to the dispersing agent and carbon black were needed.

Several furnaces were used to convert the $UO_3{\cdot}2H_2O$ microspheres into UN microspheres. A Harrop Model ST-736 differential thermal analyzer/thermogravimetric analyzer (DTA/TGA) was used to determine suitable ramp rates and dwell times for the microspheres up to 1473 K in purified argon. A Thermal Technology Astro Graphite Furnace was used to heat the microspheres up to 2023 K in a vacuum. This process reduced the UO_3 to UO_2, and then the UO_2 was reacted with the carbon to form a mixture of UO_2, UC, and UC_2. The conversion process to UN was performed in a custom-built furnace with a graphite element.

3. Results and discussion

3.1. Impacts of carbon and gelation temperature

At ORNL, uranium microspheres, which were prepared at gelation temperatures above 343 K, were prone to cracking as the spheres were air dried. This cracking was most likely due to increases in hydrostatic pressures within the small diameter pores or capillaries in the gel spheres. As the water was evaporated, the diameter of the pores would shrink at a faster rate and led to increases in pressure, which would lead to crack formation and fragmentation. The successful use of carbon in this study and earlier work [10] at gelation temperatures higher than 343 K clearly demonstrated that carbon particles prevent the catastrophic increase of pressure within the microspheres.

Regardless of the gelation temperature, less than 5% of the air-dried microspheres at the various C/U ratios failed when 1.200 kg was applied to individual microspheres. However, the calcination process significantly weakened the microspheres. This observation was confirmed during the handling of the air-dried and calcined microspheres. A noticeable amount of powder and fragments was produced when the calcined microspheres were passed through sieves. In sharp contrast, the air-dried microspheres never produced any powder. Typically, the weight required to break the calcined microspheres with an initial C/U molar ratio of 2.65 was between 150 and 700 g/particle with an average of 500 g/particle. For comparison, the average weight to break the calcined UO_2 microspheres without carbon was 960 g/particle, and 20% of the calcined UO_2 microspheres broke below 400 g/particle. Two modifications to the gelation process were tested in an effort to improve the strength of the calcined microspheres. First, the C/U molar ratio in the gel spheres was reduced to 2.15, and the microspheres were then reduced at 873 K in 4% hydrogen. The lower C/U molar ratio did not noticeably change the crush strengths of the calcined microspheres with carbon. Second, the gelation temperature was increased to 355 K, which significantly reduced the uranium crystal size. For these calcined microspheres from a higher gelation temperature, the average weight to break the microspheres was 490 g/particle, which was comparable to all of the other calcined microspheres with carbon. Therefore, changes to the carbon concentration and gelation temperature did not significantly impact the crush strengths of the calcined microspheres, and it precluded the use of a fluidized bed during the heat treatments.

3.2. Calcination and the formation of the uranium oxide/uranium carbides

The presence of carbon nanoparticles during calcination is potentially problematic. Both hydrogen and adequately dispersed carbon in a microsphere are effective in the reduction of uranium trioxide (UO_3) to uranium dioxide (UO_2) in the 773 to 873 K range. In fact, the rate of the weight change due to carbon reduction was much greater than that due to hydrogen reduction [15]. The earlier internal gelation studies on the preparation of UN microspheres [9–13] have indicated that the carbon to uranium (C/U) molar ratio is a critical variable. If both carbon and hydrogen are used for the uranium reduction, then the C/U molar ratio during calcination becomes an unknown. For this study and an earlier effort [10], an additional 0.5 mol of carbon per mole of uranium was added to the feed so only carbon could be used to reduce UO_3 to UO_2 and form carbon dioxide.

Previously, the following heating profile [14] was successfully developed for uranium oxide microspheres without carbon. The ramp rate for the uranium spheres was 200 K/h with a 2 h dwell at 353 K, a 3 h dwell at 423 K, and a 5 h dwell at 873 K. The presence of carbon should facilitate the release of gases without cracking so a significant reduction in the dwell time and a small increase in the ramp rate were expected. In the DTA/TGA, the air-dried microspheres that were prepared by gelation at 335–337 K were heated to 873 K at 200 K/h in argon. After the microspheres reached 873 K, it took 50 min for the sample to reach a constant weight. When the microspheres from the 355 K gelation experiment were subjected to the same experimental parameters, the sample took 85 min to achieve a constant weight. For comparison, the air-dried uranium microspheres with no carbon that had been gelled at 335 K took 80 min to achieve a constant weight. In all cases, the calcined microspheres lost slightly more than 20 wt.%, remained intact, and displayed no obvious surface defects. Therefore, a gelation temperature of 335 to 337 K with adequate control is recommended since it is considerably easier for the gases to escape the microspheres during the initial heat treatment.

In the DTA/TGA, an air-dried sample in argon was heated to 873 K at 200 K/h and then to 1473 K at 600 K/h with no surface imperfections. Next, a larger sample of air-dried microspheres was heated to 873 K at 200 K/h and then to 2023 K at 600 K/h in a vacuum in the Astro furnace. As soon as the microspheres had reached 2023 K, the microspheres were cooled back to room temperature under vacuum. After this heat treatment, the weight to break the UO_2–UC/UC_2 microspheres increased to 1170 g/sphere. The density of these particles was 9.90 kg/m^3, and the average diameter was 900 μm. The target diameter for the sintered UN kernel was 800 μm. The XRD results indicated that the microsphere composition was 36.4 wt.% UO_2, 47.9 wt.% UC, and 15.7 wt.% UC_2. For this composition, the theoretical density of a fully dense kernel would be approximately 12.29 kg/m^3.

3.3. Conversion to UN

A sample that was previously heated to 2023 K in the Astro furnace was placed in the custom-built, high-temperature furnace. The microspheres were heated to 1873 K in less than 60 min while nitrogen was passed through the furnace at a rate of 200 cm^3/min. After 6 h at 1873 K, a significant fraction of the microspheres had broken. It is currently postulated that the cracking problem may be due to the much faster conversion at the surface of the microsphere as compared to conversion in the center of the sphere. Therefore, a lower conversion temperature is under investigation. The XRD results show that the chemical composition of the microspheres was $UC_{0.07-0.10}N_{0.90-0.93}$. The UC_xN_{1-x} lattice parameter varies linearly with respect to x. This linear variation of the lattice parameter for a solid–solution is called the Vegard's Law. Since carbothermic reduction was used to synthesize the UN sample, our converted microspheres are actually UC_xN_{1-s}. A linear plot was used to estimate x using the refined lattice parameters for UN. As shown in Fig. 1, UO_2, UC, and UC_2 phases were not detected in this sample. This resultsuggests that the conversion of the UO_2 and UC/UC_2 microspheres into UN and UCN spheres was nearly complete. The SEM was used to examine the microstructure of the $UC_{0.07-0.10}N_{0.90-0.93}$ sample. The particle density was estimated to be 8.30 kg/m^3 using geometrical information of the UN particles. As seen in Fig. 2, the microspheres contained a significant amount of pores. The presence of the pores is the most likely reason for the brittleness and low density of the spheres and for the relatively fast conversion rate. The UN conversion did not significantly change the diameter of the kernels as expected. Prior to any attempt to optimize the initial carbon content or heat profile, this result clearly demonstrates the success of the new two-step heat profile, which avoids the formation of hydrogen cyanide, a major safety concern.

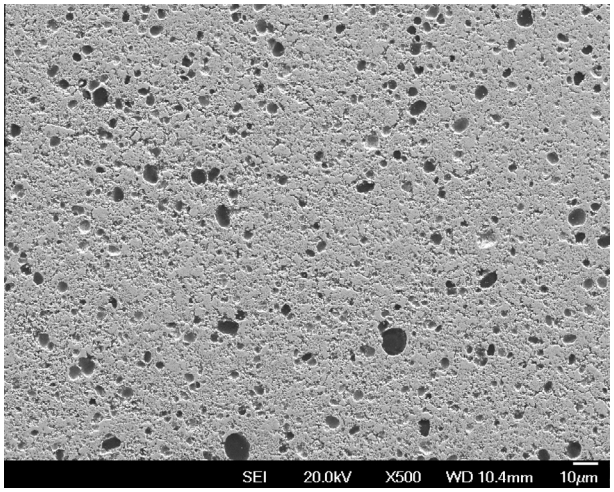

Fig. 2. A secondary electron mode SEM image of a cross section of a $UC_{0.07-0.10}N_{0.90-0.93}$ particle.

4. Conclusions

Recent process improvements to the internal gelation process were applied to the production of uranium spheres with carbon. The SEM results indicated that the Mogul L carbon black was adequately dispersed in the microspheres using the Tamol™ SN and the ultrasonic probe even at C/U molar ratios of 2.7. It should be noted that the use of well dispersed carbon nanopowders led to faster reaction kinetics. The carbon loss during the final wash step was prevented through the use of 1.5 M NH_4OH. The addition of carbon into the microspheres facilitated the release of gases as the air-dried microspheres were heated to 873 K. Unlike the uranium microspheres without carbon, no dwell time at 873 K was needed for the microspheres with higher carbon concentrations. When the uranium microspheres with carbon that were gelled at 335–337 K and 355 K were heated to 873 K and held at 873 K, the microspheres from 355 K experiment took 70% longer to complete the UO_3 reduction. Therefore, a gelation temperature of 335 to 337 K with adequate control is recommended since it is considerably easier for the gases to escape the microspheres during the initial heat treatment. In addition, the use of carbon for the UO_3 reduction is preferred because the C/U molar ratio does not become an unknown during the reduction. A simple two-step heat profile was developed for the UN conversion process, and microspheres with a chemical composition of $UC_{0.07-0.10}N_{0.90-0.93}$ were produced.

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