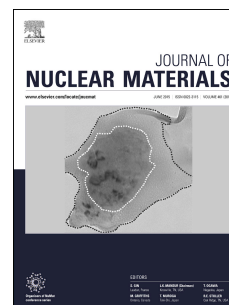


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## **Evaluation of various carbon blacks and dispersing agents for use in the preparation of uranium microspheres with carbon<sup>1</sup>**

R.D. Hunt\*, J.A. Johnson, J.L. Collins, J.W. McMurray

Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee, USA 37831

T.J. Reif, D.R. Brown

X-energy, LLC, Greenbelt, Maryland, USA 20770

### **Abstract**

A comparison study on carbon blacks and dispersing agents was performed to determine their impacts on the final properties of uranium fuel kernels with carbon. The main target compositions in this internal gelation study were 10 and 20 mole % uranium dicarbide ( $\text{UC}_2$ ), which is  $\text{UC}_{1.86}$ , with the balance uranium dioxide. After heat treatment at 1900 K in flowing carbon monoxide in argon for 12 h, the density of the kernels produced using a X-energy proprietary carbon suspension, which is commercially available, ranged from 96% to 100% of theoretical density (TD), with full conversion of UC to  $\text{UC}_2$  at both carbon concentrations. However, higher carbon concentrations such as a 2.5 mole ratio of carbon to uranium in the feed solutions failed to produce gel spheres with the proprietary carbon suspension. The kernels using our former baseline of Mogul L carbon black and Tamol SN were 90–92% of TD with full conversion of UC to  $\text{UC}_2$  at a variety of carbon levels. Raven 5000 carbon black and Tamol SN were used to produce 10 mole %  $\text{UC}_2$  kernels with 95% of TD. However, an increase in the Raven 5000 concentration led to a kernel density below 90% of TD. Raven 3500 carbon black and Tamol SN were used to make very dense kernels without complete conversion to  $\text{UC}_2$ . The selection of the carbon black and dispersing agent is highly dependent on the desired final properties of the target kernels.

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\* Tel.: +1-865 574 5481; fax: +1-865 574 6872

E-mail address: [huntrd@ornl.gov](mailto:huntrd@ornl.gov)

**Keywords:** uranium microspheres, carbon black, dispersing agent, internal gelation

## 1. Introduction

In the early 1980s, laboratory-scale quantities of uranium dioxide ( $\text{UO}_2$ ) and uranium dicarbide ( $\text{UC}_2$ ) kernels were produced at Oak Ridge National Laboratory (ORNL) with Black Pearl L [Cabot Corporation (Cabot)] as the carbon source and Marasperse CB (Marathon) as the dispersing agent [1, 2]. The vast majority of these gel microspheres from the internal gelation process were plagued with large carbon agglomerations that made the conversion to  $\text{UO}_2$  and  $\text{UC}_2$  kernels much more difficult, requiring higher conversion temperatures and longer conversion times. Subsequent research at ORNL on acid-deficient uranyl nitrate (ADUN) resulted in a much better understanding of key process variables in the internal gelation process [3], which led to the production of kilogram quantities of  $\text{UO}_2$  kernels with near theoretical density (TD), uniform sizes, and smooth surfaces in the early 2000s [4, 5]. Shortly afterward, small samples of uranium microspheres with Black Pearl L or Monarch 4750 (Cabot) were prepared and analyzed using optical images [6]. The carbon blacks were dispersed using Tamol SN (Rohm and Haas) and either mechanical mixing or an ultrasonic probe. For each sample, high-density kernels of uranium oxycarbide (UCO) were achieved using a sintering temperature of 1953 K, dwell time of up to 6 h, and purified argon. UCO kernels contain  $\text{UO}_2$  and a mixture

of uranium carbide (UC) and  $UC_2$ . However, attempts to make high density kernels of  $UO_2$  and  $UC_2$  using the sintering conditions (1,823 K for 4 h in 1% carbon monoxide) from the 1980s effort [1, 2] failed. The results from this effort [6] suggested that further work on carbon dispersion in uranium microspheres was needed. In the same time frame, an evaluation of water-dispersible carbon blacks for the fabrication of UCO microspheres identified a carbon black solution, sample g (Cabot), as the most promising candidate [7]. Sample g was successfully used to make UCO kernels with manufacturing scale equipment [8, 9]. The carbon dispersion conditions were optimized for Raven 1000, Raven 1040, Raven M [Columbian Chemicals Company, Inc. (Columbian)] and Black Pearl L carbon blacks with Tamol SN, Tergitol XD (Dow), and Borch Gen 12 (Borchers) dispersing agents [10]. Research groups from around the world use a wide variety of carbon blacks, dispersing agents, and carbon-to-uranium molar ratios. These factors, along with others such as gelation temperature and uranium density, must be systematically studied and their effects on the process, product yields, and properties understood. The purpose of this work is to contribute to these earlier efforts with a comparison study of carbon blacks and dispersing agents in their roles in the preparation of UCO kernels.

Currently, researchers from ORNL and X-energy, LLC, are developing a pebble fuel composed of  $UO_2$  and  $UC_2$ , which is really  $UC_{1.86}$  in this study. The target  $UC_2$  concentrations in this study were 10 and 20 mole %. A carbon-to-uranium molar ratio of 2.5 was also evaluated as part of an ongoing uranium nitride (UN) study, which started with uranium oxide microspheres with carbon [11]. These microspheres were converted to UCO microspheres and then into UN kernels. As part of this effort, a comparison study on currently available carbon blacks and dispersing agents based on the current recommendations of various suppliers was conducted. The results of this comparison study are presented below.

## 2. Experimental methods

### 2.1 Preparation of stock solutions

The standard concentrations for the hexamethylenetetramine (HMTA; Fisher Scientific) and urea (Fisher Scientific) in the stock solution were 3.2 M each. After a stock solution was prepared and filtered, the density was measured to be 1,140 kg/m<sup>3</sup> and the pH ranged from 8 to 9. The ADUN stock solutions in these runs had a density of 1,870 kg/m<sup>3</sup> with a pH of 1.3 and a uranium concentration of 2.85 M. Only concentrated nitric acid (Fisher Scientific), distilled water, and triuranium octoxide (U<sub>3</sub>O<sub>8</sub>) were used in the preparation of the ADUN. The acid concentration was reduced using U<sub>3</sub>O<sub>8</sub> additions. The pH and uranium concentration [11] were used to estimate that the molar ratio of nitrate (NO<sub>3</sub><sup>-</sup>) to uranium was about 1.65.

### 2.2 Carbon blacks and dispersing agents

Several carbon blacks and dispersing agents were compared in this study. The carbon blacks included Raven 3500, Raven 5000 (Columbian), Mogul L (Cabot), and a modified carbon black suspension (X-energy proprietary) that is commercially available. The particle sizes of the Raven 3500 and 5000 are 13 and 8 µm, respectively, while the primary particle size for Mogul L is much smaller at 24 nm. Before use, the solid carbon blacks were first passed through a 120- or 170-mesh sieve to exclude all particles with diameters greater than 125 or 90 µm, respectively. However, this sieving step could have been eliminated for the Raven 3500 and Raven 5000. The dispersing agents used with these carbon blacks included Tamol SN, Disperbyk 2013 (Byk USA), and Solsperse WV400 (Lubrizol). The Disperbyk 2013, which is a

structured copolymer with pigment affinic groups, has a low solubility in the HMTA-urea solution at room temperature. To achieve the manufacturer's recommended concentration of Disperbyk 2013, the HMTA-urea solution was heated to 308 K. Tamol SN is a sodium salt of sulphonated naphthalene formaldehyde condensate while Solsperse WV400 is a 40 % active high molecular weight hyperdispersant. For the UC<sub>2</sub> target concentration of 10 mole %, each of the 3 solid carbon black samples were tested separately with each of the 3 dispersing agents. For the UC<sub>2</sub> target concentration of 20 mole %, each of the 3 solid carbon black samples were tested separately with Tamol SN. The X-energy carbon suspension was evaluated at 3 different carbon concentrations.

### 2.3 Carbon dispersion in HMTA-urea

The dispersing agent was typically added to 33.1 mL, or 33.7 g, of HMTA-urea, and the resulting solution was mixed by hand until the dispersing agent went into solution. The amount of each dispersing agent was varied to determine its dispersing agent requirement, which is the minimum amount of dispersing agent needed to produce a fluid dispersion at a specified carbon black loading in H<sub>2</sub>O [13]. Too little or too much dispersing agent relative to carbon black can lead to larger carbon aggregates and compromise the stability of the feed solution. In this study, the weights of Tamol SN for each gram of carbon black were 0.07, 0.08, and 0.09 g. Similarly, the weights of Disperbyk 2013 for each gram of carbon black were 0.15, 0.20, and 0.25 g. For Solsperse WV400 tests, 1.75 g of dispersing agent was used for each gram of the carbon black. Next, 0.9–1.4 g of sieved carbon black powder was added to the unheated HMTA-urea solution, which was then chilled to 273 K in an ice water bath. For this study, the targeted nonvolatile carbon-to-uranium molar ratio was either 0.89 or 1.28 for the 10 or 20 mole % UC<sub>2</sub>, respectively.

The manufacturers indicated that volatile organics were 5–10 wt % of the carbon blacks, so the amount of carbon black used in each experiment was adjusted to account for these volatiles. If the carbon black correction was not appropriate based on the chemical composition of the kernels, adjustments to the initial amount of carbon black could easily be made. The carbon black in the chilled solution was next dispersed 2 times for 5 min each using an ultrasonic probe (Hielscher Ultrasound Technology model UP200S). The dispersed carbon black sample was cooled in an ice water bath for a minimum of 30 min. The X-energy carbon black suspension had been dispersed by the manufacturer before delivery.

#### *2.4 Internal gelation system*

The internal gelation system used in these tests was a modification of another apparatus that was used to produce uranium and plutonium microspheres in a glove box. The system is described in detail elsewhere [4, 5] and located in a radioactive fume hood. The use of carbon black required a few minor system modifications that are now used with and without carbon black. First, the stainless steel pot was replaced with a chilled glass broth pot, which has an exit line at the bottom of the vessel. The use of glass permits visual confirmation of sample mixing and solution height. Second, the glass pot was placed on a magnetic stir plate so a stir bar could be used to keep the carbon black suspended during each 6–7 min gelation experiment. A Micropump gear pump with graphite gears was used to transfer the solution through the feed line at a rate of 10.3 mL/min. The feed line was passed between a metal support and an accelerometer, which was typically set at 143–146 Hz. At the end of the feed line, the solution was passed through a 20-gauge hypodermic needle with a beveled tip (Cadence, Inc.). The feed was sent to the top of a gelation column with hot silicone oil, which was 335–338 K. The gel

time for the droplets was approximately 4 s, and the residence time of the spheres in the gelation column was 10 s. The feed and system parameters were selected to produce dense  $\text{UO}_2$  and  $\text{UC}_2$  kernels with diameters in the 425  $\mu\text{m}$  range.

From an operational standpoint, the only problematic components were the Solspense WV400 and a high concentration of the X-energy carbon suspension. In the Solspense WV400 tests, a considerable amount of foam was generated in the broth pot, and about 8% of the feed was not delivered to the gelation column. For the X-energy carbon suspension, a feed solution failed to gel when the carbon-to-uranium molar ratio increased to 2.5, which is typical ratio for the UN effort [14]. The use of X-energy carbon suspension adds 1–2 mL water to the typical feed composition. However, this additional water is not a problem for the feed designed to make 10 and 20 mole %  $\text{UC}_2$ .

### *2.5 Aging, washing, and drying*

With one exception, the procedures for aging, washing, and drying did not change with the presence of carbon, and they are described elsewhere [4, 5]. The one exception is the concentration of the ammonium hydroxide ( $\text{NH}_4\text{OH}$ ) in the wash step. Without carbon in the uranium microspheres, 0.5 M  $\text{NH}_4\text{OH}$  is typically used. Based on past experience with uranium microspheres with carbon black, a minimum  $\text{NH}_4\text{OH}$  concentration of 1.5 M was needed to prevent the transfer of carbon particles into the wash solution during the first three washes. A trace amount of carbon black from the uranium microspheres with Tamol SN was normally observed in the fourth and final wash solution with  $\text{NH}_4\text{OH}$ . The uranium microspheres with Disperbyk 2013 and Cabot Mogul L transferred carbon to most of the  $\text{NH}_4\text{OH}$  wash solutions, and the carbon transfer increased significantly as the Disperbyk 2013 concentration increased. A



noticeable amount of carbon leaching was observed in most of the washes of the microspheres with Solsperse WV400. In sharp contrast, nearly all of the  $\text{NH}_4\text{OH}$  wash solutions for the uranium microspheres with Disperbyk 2013 and the Raven carbon blacks remained clear. The spent  $\text{NH}_4\text{OH}$  washes of the uranium microspheres with the X-energy carbon suspension were initially red. No carbon in the spent washes was observed until the final wash, which contained a trace amount of carbon. The effectiveness of the  $\text{NH}_4\text{OH}$  wash step was monitored by measuring the electrical conductivity of the spent  $\text{NH}_4\text{OH}$  solution with a YSI model 3100 conductivity instrument. The microspheres were washed with  $\text{NH}_4\text{OH}$  until the electrical conductivity of the spent  $\text{NH}_4\text{OH}$  was below 2 mS. From an operational standpoint, the only problematic combination was Cabot Mogul L and Disperbyk 2013 due to its  $\text{NH}_4\text{OH}$  wash behavior.

## *2.6 Conversion to $\text{UO}_2$ and $\text{UC}_2$ kernels and subsequent characterization*

Our initial kernel conversion process was based on an earlier conversion process [1, 2]. The process was designed to first sinter the material then back react the sintered product to produce the desired phases by carefully controlling the CO pressure with temperature. The first step is carbothermic reduction, whereby  $\text{UO}_2$  and free carbon react to a mixture of  $\text{UO}_2$  and UC. Next, the microspheres are sintered at about 1,900 K in an atmosphere with a CO pressure of 1 % or less that favors  $\text{UO}_2$ -UC equilibrium, producing nearly full TD  $\text{UO}_2$ -UC composite kernels. Next, the CO partial pressure was increased so the  $\text{UO}_2$ -UC composition was changed to  $\text{UO}_2$ - $\text{UC}_2$ , the so-called back reaction. The sample was held under these conditions long enough to complete the conversion from UC to  $\text{UC}_2$  in most cases.

For this comparison study, all of the microspheres were subjected to the same conversion and sintering processes. Very small samples on the order of 300 mg were placed in a 449 F1 Jupiter simultaneous thermal analyzer (Netzsch), which is a high-temperature analytical device used to perform the sintering activities. The reduction of uranium trioxide to  $\text{UO}_2$  was performed by excess carbon in the microspheres. In a typical experiment, the microspheres were held at a temperature of about 1,900 K for 12 h in flowing 4% carbon monoxide (CO) in argon. The actual temperatures and holding times were varied based on the initial results.

A shadow imaging technique was used to determine the ellipticity and average diameter of a statistically significant number of converted kernels. After the optical image was obtained using an MZ16 stereomicroscope (Leica), the kernels were counted and weighed using an XP504 balance (Mettler Toledo) with a readability of 0.1 mg. These results were then used to estimate the average density of the kernels. Finally, x-ray diffraction (XRD) was used to determine the success of the conversion to 10% or 20%  $\text{UC}_2$ . A powder diffractometer (Bruker, D2 Phaser) with a Cu k-alpha1 source ( $\lambda = 0.15418 \text{ nm}$ ) and an energy dispersive one-dimensional detector (LYNXEYE) were used. Measurements were taken from  $10^\circ$  to  $120^\circ$  in  $2\theta$  over 2 h. Weight fractions of  $\text{UO}_2$ , UC, and  $\text{UC}_2$  were calculated using a reitveld refinement on the measured XRD diffraction pattern.

When the kernel density for a given carbon black and dispersing agent combination was at 90% of TD or higher, a small sample of kernels was embedded as whole particles in an acrylic epoxy. The kernels were then ground to near the spherical center, and optical images were obtained.

### 3. Results and discussion

### 3.1 Final sintering and conversion conditions for the comparison studies

Before this effort, Mogul L and Tamol SN were our preferred carbon black and dispersing agent, respectively. This combination was used to establish our baseline conversion and sintering conditions. The sintering and conversion attempts based on the conditions from the early study [1, 2] produced kernels that did not fully convert to  $\text{UO}_2$  and  $\text{UC}_2$  and did not achieve our minimum target of 90% of TD. The sintering temperature was increased to about 1,900 K, and the time at the high temperature was increased to 12 h. For the microspheres prepared using Mogul L and Tamol SN, these changes led to fully converted 90–92%  $\text{UO}_2$  and 10%  $\text{UC}_2$  kernels (nominal or target compositions) with a density of 90% of TD as shown in Table 1. These same conditions were applied to other uranium and carbon microspheres, and no operational problems were encountered during kernel fabrication.

**Table 1.** Density and XRD results of sintered kernels using key combinations of carbon black and dispersing agent.

	Raven 5000 & Tamol SN	Raven 3500 & Tamol SN	X-energy carbon suspension
Target 10 % $\text{UC}_2$			
Density	95% of TD	96% of TD	97–100% of TD
XRD	88% $\text{UO}_2$	81% $\text{UO}_2$	90–92% $\text{UO}_2$
	0% UC	9% UC	0% UC
	12% $\text{UC}_2$	10% $\text{UC}_2$	8–10% $\text{UC}_2$
Target 20 % $\text{UC}_2$			
Density	<90% of TD	98% of TD	96–98% of TD
XRD	Not measured	77% $\text{UO}_2$	77–80% $\text{UO}_2$
		3% UC	0% UC
		20% $\text{UC}_2$	20–23% $\text{UC}_2$

### 3.2 Carbon black and dispersing agent potentially better than Mogul L carbon black and Tamol SN

The combination of Raven 3500 and Tamol SN produced sintered microspheres with a density of 96% of TD for the target  $UC_2$  concentration of 10 mole %. Unfortunately, the XRD results revealed that the kernels contained comparable amounts of UC and  $UC_2$ . When the carbon concentration was increased to produce 20 mole %  $UC_2$ , the density of the kernels increased slightly to 98% of TD, and the ratio of  $UC_2$  to UC increased significantly to 7 to 1. This promising result led to the production of microspheres with Raven 3500 and Tamol SN, and with a carbon-to-uranium molar ratio up to 2.5, which is typical in the production of UN kernels.

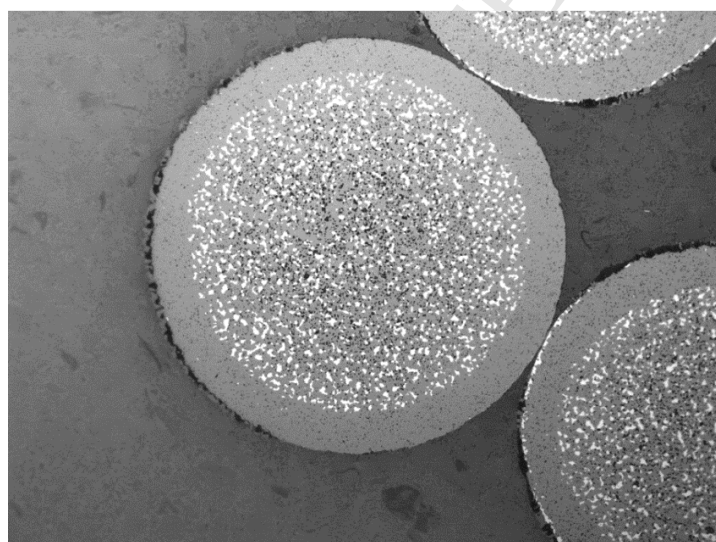
Sintered microspheres from the combination of Raven 5000 and Tamol SN had a density of 95% of TD for the target  $UC_2$  concentration of 10 mole %. Unlike the kernels with Raven 3500, the conversion to  $UC_2$  was complete. Unfortunately, when the carbon concentration was increased for the 20 mole %  $UC_2$  target, the density of the kernels dropped below 90% of TD.

The sintered kernels using the X-energy carbon suspension gave the best results for target  $UC_2$  compositions. For the 10 %  $UC_2$  kernels, the densities ranged from 97% to 100% of TD. The XRD results indicated complete conversion to  $UO_2$  and  $UC_2$ . These results meant the processing temperature and/or time could be significantly reduced. The optical image of the inside of the sintered kernel is shown in Figure 1. The outer ring of the kernels was carbon deficient, and it appeared even when no carbon was removed during the  $NH_4OH$  wash step. The outer ring appears at about 1600 K and is under investigation.

For the 20%  $UC_2$  kernels using the X-energy carbon suspension, the density dropped to 96% to 98% of TD, which still exceeded our product specifications. The other carbon blacks in this study were added as solids, and they did not significantly add to the volume of the combined

HMTA-urea and uranium solutions. However, the X-energy carbon suspension is added as a solution to the other solutions, which can significantly reduce the uranium concentration in the final feed solution. This feed dilution leads to more initial porosity in the air-dried microspheres. While a higher initial porosity can prevent flaking during the calcining process, a lower final density of the kernels is typical. In this study, the uranium and HMTA-urea stock solutions are very near saturation. If a denser kernel with 20%  $\text{UC}_2$  is needed, one possible alternative is to combine the HMTA-urea and carbon suspension and then evaporate some of the excess water from the combined solution.

When the amount of X-energy carbon suspension was increased so the carbon to uranium molar ratio was 2.5, the feed solution would not form a gel using our test conditions. When additional distilled water was used in place of the carbon suspension, the solution of uranyl nitrate, HMTA-urea, and water would still form a gel in a reasonable amount of time. Pathways to achieving appropriate uranium to HMTA-urea ratios for higher C/U sol-gel microspheres are the subject of an ongoing investigation.



**Figure 1.** Optical image of a sintered kernel with 92%  $\text{UO}_2$  and 8%  $\text{UC}_2$  from a batch made with the X-energy carbon suspension with a target of 10%  $\text{UC}_2$ .

#### 4. Conclusions

The selection of carbon black and dispersing agent can have significant impact on the operation of the internal gelation system and the density of the sintered kernel. Our traditional combination of Mogul L carbon black and Tamol SN performs reasonably well over a wide range of carbon-to-uranium mole ratios. However, the densities of the sintered kernels from this pair were generally 90–92% of TD for a wide range of carbon concentrations. Sintered kernels from the X-energy carbon suspension tests had final densities between 96% and 100% of TD when the  $UC_2$  concentration in the kernel was 20 mole % or below. However, as the carbon-to-uranium molar ratio increases, the advantage of the carbon suspension decreases, and an operational problem is encountered if the carbon-to-uranium mole ratio reaches 2.5. The sintered microspheres from the combination of Raven 5000 and Tamol SN had a density of 95% of TD for the target  $UC_2$  concentration of 10 mole %, and the conversion to  $UC_2$  was complete. Finally, the combination of Raven 3500 carbon black and Tamol SN is also a viable option if the conversion to  $UC_2$  is not a requirement, as in the preparation of UN kernels. Therefore, selection of the carbon black and dispersing agent is highly dependent on the desired final properties of the target kernels. In conclusion, for our target composition of 10 to 20 mole %  $UC_2$  kernels, the X-energy carbon suspension is clearly the best choice, and it is currently being used to make kilogram quantities of the kernels.

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