*Properties of Hafnium Oxide with Respect to
Its Use as a Simulant for $^{238}\text{PuO}_2$ in Safety Studies*

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**PROPERTIES OF HAFNIUM OXIDE WITH RESPECT TO
ITS USE AS A SIMULANT FOR $^{238}\text{PuO}_2$ IN SAFETY STUDIES**

by

Mary Ann Reimus and Michael G. Stout

ABSTRACT

The general-purpose heat source (GPHS) is a modular component of the radioisotope thermoelectric generators (RTGs) that provide power for National Aeronautics and Space Administration's (NASA's) deep-space missions. Because any space mission could experience a launch abort or return from orbit, the GPHS heat source has been designed to survive credible accident environments. Safety tests are conducted to expose simulant-fueled GPHSs to accident scenarios. Tests that require field testing of heat source and RTG are performed using GPHS capsules fueled with $^{238}\text{UO}_2$ (natural uranium depleted of ^{235}U). Although the levels of radioactivity associated with $^{238}\text{UO}_2$ are extremely low, it would be desirable to eliminate this potential dispersal during field experiments. We investigated one potential nonradioactive material, hafnium oxide (HfO_2), to replace $^{238}\text{UO}_2$ as mock material for $^{238}\text{PuO}_2$ in field test studies. In this report we discuss how closely the properties of hafnia match those of $^{238}\text{PuO}_2$ and whether or not it would be realistic to use HfO_2 as a simulant fuel.

1. INTRODUCTION

The general-purpose heat source (GPHS) is a modular component of the radioisotope thermoelectric generators (RTGs) that provide power for National Aeronautics and Space Administration's (NASA's) deep space missions. An RTG generates electric power by using the heat of ^{238}Pu α -decay to create a temperature differential across a thermoelectric array. The two RTGs used for the recent Cassini mission to Saturn were each fueled with 18 GPHS modules. Each GPHS module, Figure 1, contains four $^{238}\text{PuO}_2$ fuel pellets that provide a total thermal output of ~250 W. The fuel pellets

are individually encapsulated in a vented, DOP-26 iridium alloy shell. This alloy was developed by Oak Ridge National Laboratory,⁽¹⁾ with the alloy development complemented by axial testing and high-temperature compatibility studies conducted at Los Alamos National Laboratory.⁽²⁻³⁾ Two capsules are held in a Fineweave-Pierced Fabric[†] (FWPF) graphite impact shell (GIS), and two GISs are contained within an FWPF aeroshell module.

Because any space mission could experience a launch abort or return from orbit, the GPHS heat source has been designed to survive credible accident

GPHS Module

Figure 1. A schematic of an FWPF aeroshell containing two graphite impact shells.

[†] Fineweave-Pierced Fabric 3-D carbon/carbon composite, a product of TEXTRON Specialty Materials.

environments. Previous testing conducted in support of the Galileo mission to Jupiter, Ulysses (a solar-probe mission), and the Cassini mission documented the response of the GPHS heat source for a variety of accident scenarios including fragment-impact, aging, atmospheric reentry, and Earth-impact conditions.⁽⁴⁻¹²⁾ Tests that required field testing of heat-source and RTG components (such as solid-propellant fire, explosive overpressure, large- and thin-fragment interaction, and a launch pad impact) were performed using GPHS capsules fueled with $^{238}\text{UO}_2$ (natural uranium depleted of ^{235}U).⁽¹³⁻¹⁹⁾

The field tests of the heat source and RTG components simulate the environment, temperature, and mechanical insult severity predicted for the possible accident scenarios. These are difficult environments that can result in the rupture of the DOP-26 iridium alloy containment shell and the dispersal of $^{238}\text{UO}_2$. Because these experiments are in the field, it is nearly impossible to prevent the exposure of the environment to the $^{238}\text{UO}_2$. Although the levels of radioactivity associated with $^{238}\text{UO}_2$ are extremely low, it would be desirable to eliminate this potential dispersal during field experiments.

We have investigated one potential nonradioactive material, hafnium oxide (HfO_2), to replace $^{238}\text{UO}_2$ as mock material for $^{238}\text{PuO}_2$ in field test studies. In this report we will discuss how closely the properties of hafnia match those of $^{238}\text{PuO}_2$ and whether or not it would be realistic to use HfO_2 as a simulant fuel. There are four different material behaviors that we must be concerned with in selecting a mock fuel for $^{238}\text{PuO}_2$: thermal response, strength and density, impact behavior, and particle size

and shape distributions after impact. If these properties match those of plutonia or if the differences in the properties can be adjusted for in the design of the safety experiment, then our simulant is an acceptable substitute material.

All of our experimental safety analysis experiments are performed at elevated temperatures, near $1,000^\circ\text{C}$. Thus, the thermal properties of the simulant fuel are important. We must adjust the test temperatures, heating rates, and hold times at temperature to account for any differences in the thermal properties. In addition, we must allow for the fact that the $^{238}\text{PuO}_2$ is self heating but the simulant material will not be. Fortunately, if the heat capacity and thermal conductivity are known, thermal analyses can be conducted with computer models. Based on these simulations, the heating temperatures and times can be specified so that the experiment is a valid representation of the actual scenario.

In a safety experiment, the mechanical response of the fuel elements is critically important. We must assess how severe the deformation in the fuel cladding will be as a function of the overpressure and impact conditions and whether or not the fuel cladding will rupture. The performance of the fuel cladding is a function not only of the properties of the clad itself but of the fuel as well. A lower-density fuel will produce less deformation in the clad for a given impact condition. If the fuel is very weak, there will be enhanced clad deformation.

The mechanical response of both the fuel and the clad will be strain-rate sensitive. That is, we must assess the fuel's mechanical behavior under impacts as well as by means of quasi-static mechanical-properties

measurements in the laboratory. This assessment needs to be by means of controlled impacts at a specific temperature, velocity, and for a particular orientation, (for example “side on”). This will permit a comparison of the performance of the mock fuel to identical impacts of the $^{238}\text{PuO}_2$ actual fuel.

Finally, we must be concerned not only with the degree of deformation in the clad and whether it ruptured or not, but we must examine how the fuel breaks up when impacted. The size, morphology, and number distributions of fuel particles after impact will determine the extent to which contamination will spread in the event of a clad rupture. A mock fuel could behave identically in all other respects to the $^{238}\text{PuO}_2$ but be a poor simulant if its particle sizes and shapes differed dramatically from those of the actual fuel.

Our objective was to investigate these material behaviors and performance characteristics for hafnia and to compare the results to those of $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$, the current mock material. In the case of the thermal properties, the information is available in the literature. However, we needed to perform our own evaluation of the mechanical behavior, impact response, and fuel particle sizes after impact.

In order to confirm the viability of the use of hafnia as a fuel simulant, three impact tests of encapsulated hafnia were conducted and a comparison made between these impact results and those for plutonia and urania-fueled clads. Fueled-clad dimensional distortion and pellet fragmentation were compared.

2. THERMAL PROPERTIES OF HfO_2

2.1. Heat Capacity

The heat capacities²⁰ of four materials, $^{238}\text{PuO}_2$, $^{238}\text{UO}_2$, HfO_2 , and Y_2O_3 are shown as a function of temperature in Figure 2. The most rapid change or increase in heat capacity with temperature occurs below 700°C . Above 700°C , the change with increasing temperature is relatively slight. The data for $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$ are very close over almost the entire temperature range. The only real deviation is above $1,700^\circ\text{C}$ where the $^{238}\text{UO}_2$ data tail up, and the $^{238}\text{PuO}_2$ specific heat remains constant. These data are at the very end of the temperature range that was measured, and one might question their absolute reliability. The specific heat of HfO_2 is also shown in Figure 2 as a function of temperature. The specific heat of the hafnia follows very much the same trend with temperature as the plutonia and urania, but its absolute value is about 30% higher than the other two oxides.

Figure 2. Specific heat as a function of temperature for plutonia, urania, hafnia, and yttria.

The specific heat of the yttria is about 70% higher than the urania and plutonia, although it too follows the same trend as the other oxides.

2.2. Thermal Conductivity

Thermal conductivities for the four oxides, $^{238}\text{PuO}_2$, $^{238}\text{UO}_2$, HfO_2 , and Y_2O_3 , are shown in Figure 3. Again, one observes a very close match between the urania and plutonia. At a low temperature, 20°C, the thermal conductivity of the hafnia and also yttria are very different from $^{238}\text{PuO}_2$. The conductivity of hafnia is about 50% that of the $^{238}\text{PuO}_2$; yttria has a thermal conductivity twice that of $^{238}\text{PuO}_2$. As temperature increases, the thermal conductivities decrease and begin to converge. Above 700°C, the values for all four oxides are nearly the same. They remain equivalent up to about 1,700°C.

Figure 3. Thermal conductivity as a function of temperature for plutonia, urania, hafnia, and yttria.

3. STRENGTH CHARACTERISTICS OF HfO_2

3.1. Strength of $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$ as a Function of Temperature

The strength and ductility of $^{238}\text{PuO}_2$ were studied extensively by Petrovic et al.²¹ with a series of compression experiments. We complemented this work with a similar study,²² compression experiments on $^{238}\text{UO}_2$. Petrovic et al. found that there was very little ductility associated with the plutonia when reporting strains beyond the proportional limit of between 0.0018 and 0.0061. Accordingly, ultimate compressive strengths are listed in the Petrovic report. The experiments for the $^{238}\text{PuO}_2$ were done at temperatures of 1,000°, 1,200°, 1,300°, 1,400°, and 1,500°C. In the case of the $^{238}\text{UO}_2$, tests were also performed at these temperatures and at 900° and 1,100°C as well. The urania had greater ductility than the plutonia and thus both yield and ultimate strengths are found in the Stout, Ellis, and Pereyra²² report. The results from these two investigations are shown together in Figure 4. At 1,000°C, the yield and ultimate compressive strengths of $^{238}\text{UO}_2$ are very close to the strength of the $^{238}\text{PuO}_2$. One can see that the yield strength of the $^{238}\text{UO}_2$ is comparable, and its ultimate strength is only about 30 MPa greater than that of the $^{238}\text{PuO}_2$. As the temperature is increased, the strength of the urania and plutonia both decrease. The strength of the $^{238}\text{PuO}_2$ has decreased to only about 30 MPa at 1,400°C. For temperatures at and above 1,200°C, the ultimate strength of the plutonia falls consistently below that of the urania. One should note that at 1,400°C the average strength of the $^{238}\text{PuO}_2$ is about half that of the $^{238}\text{UO}_2$, although there is a great deal of

Figure 4. The ultimate compressive strength of $^{238}\text{PuO}_2$, Petrovic et al., compared to the strength of $^{238}\text{UO}_2$, Stout et al.

scatter in the data, and both strengths are very low.

3.2. Strength of HfO_2 as a Function of Temperature

3.2.1. Fabrication Processes

Two different processes were used to fabricate hafnia pellets, which can be best grouped into an isostatic press and sinter and a hot press.

A consistent history was used to fabricate all of the hot pressed pellets we worked with. The hafnia powder was loaded into graphite dies and prepressed to 50 psi. The pellets were then heated to between 1,550°C and 1,560°C at a rate of 25°C to 33°C/min. The target temperature was 1,550°C, but there was a slight overshoot in the furnace control. Pressure was applied evenly at a rate of 10 psi/min. over the heat-up period to a final

pressure of 500 psi. The conditions of 1550°C and 500 psi were held until the motion of the hot pressing piston ceased. This required 5 min.

At this point, the furnace power was shut off, and the pellet was allowed to cool under pressure. The final step in the process was to machine the pellets to final dimension and perform a bake-out at 250°C for 2 h. The heating and cooling rates for the bake-out were not critical. Various densities were reported for the pellets made by the hot-pressing process. We measured the densities of our hot-pressed compression specimens before testing. They averaged 87% of theoretical.

An isostatic pressing and ambient pressure sintering treatment was also used to fabricate hafnia pellets. The material was first cold isostatically pressed to 50 ksi and ramped to a sintering temperature at 250°C/h. The sample was sintered at 1,500°C for 8 h. The cool down was at 150°C/h. Following machining, the isostatically pressed material was also baked-out for 2 h at 250°C. These pellets were given an additional heat treatment of temperature cycling between 1,600°C and 1,800°C, which will be described in the following sections. After these treatments, we cored compression specimens and found their density to be 96% of theoretical.

3.2.2. Mechanical Properties of Cold-Pressed and Sintered Material—as Received

The cold, isostatically pressed and sintered material, with a density of 96%, was tested in compression in the as-received state at both 1,200°C and 1,300°C. At both temperatures, the material strength exceeded 300 MPa. We were not able to measure the exact strengths. In one case, we reached the load capacity of

the testing machine without the specimen fracturing, and in another experiment one of our test platens fractured leaving the specimen intact. The strength of the $^{238}\text{PuO}_2$ is much lower than 300 MPa at the same temperatures: 100 MPa at 1,200°C and 65 MPa at 1,300°C, respectively.

3.2.3. Mechanical Properties of Cold Isostatically Pressed and Sintered Material—Thermal Cycling

The hafnium/oxygen phase diagram²³ is shown in Figure 5. One can see that stoichiometric HfO_2 undergoes a solid/solid-phase transformation from the monoclinic to the tetragonal crystal structure at approximately 1,700°C. The change in crystal structure is accompanied by a

Figure 5. The hafnium-oxygen phase diagram. Note the solid/solid-phase transformation at $\approx 1,700^\circ\text{C}$, for HfO_2 .

material shape and volume change. For polycrystalline hafnia that passes through the phase transformation, this results in local stresses, on the length scale of the grain size. These stresses are potentially large enough to produce microfractures in the material, which would decrease its strength.

We have been able to take advantage of the solid/solid-phase transformation in the hafnia to tailor its strength. Individual compression specimens were thermally cycled between 1,650°C and 1,850°C, taking the material through its phase transformation. The compression specimens were ramped from 1,650°C to 1,850°C (two specimens were cycled between 1,600°C and 1,800°C) and back to 1,650°C in two minutes. This represents two passages through the phase transformation. The strength of the hafnia will be a function of the number of times it undergoes the phase transformation because each transit will produce additional microcracking. Measurements of the ultimate compressive strength were performed at 1,300°C and a strain rate of $\dot{\epsilon} = 2.5 \times 10^{-4} \text{ s}^{-1}$ as a function of the number of thermal cycles given the compression specimen. These results are plotted in Figure 6. One can see that the strength decreases dramatically with the number of thermal cycles, up to 40 cycles. Past 40 cycles, the strength increases. It is not clear why this increase in strength occurs. Our speculation is that there are two different mechanisms that affect the hafnia. Thermal cycling through the hafnia's phase transformation should introduce microcracking and produce a continued decrease in strength as the number of cycles increases. At the same time, we are well above the sintering temperature of the hafnia. Thus, the process of sintering and grain growth, a strengthening mechanism, is occurring concurrently. Evidently, as the

Figure 6. The compressive strength of hafnia at 1,300°C as a function of the number of thermal cycles. The compression experiments were done at a strain rate of $\dot{\epsilon} = 2.5 \times 10^{-4} \text{ s}^{-1}$.

time at temperature increases, the sintering process begins to dominate over the development of microcracks, and the strength of the material increases. This evidently occurs at about 40 cycles. The results were independent of the two cycling treatments used, and all of the data were consistent (independent of the 50°C difference in the mean temperature that we cycled around).

In the experiments described above, single, small compression specimens were individually thermally cycled. Because of the small sample size, temperatures were uniform throughout the specimen during the thermal cycling. When thermal cycling a full-size pellet, this was not the case. The temperature of the core of the pellet lagged the surface temperature. This was determined by mounting a thermocouple inside the specimen for comparison to one on the

specimen surface. Based on these temperature data we cycled our furnace temperature between 1,600° and 1,800°C in one-minute ramps and added one-minute hold times at the two temperature extremes. This resulted in a temperature cycle between 1,640°C and 1,745°C, measured by a thermocouple at the pellet core. It took two minutes to go from 1,640°C to 1,745°C and two minutes to return to the original temperature. We used one-hour-long ramps to heat the pellet from room temperature to the cycling temperature and then back to room temperature. Two large pellets were given thermal cycles in this manner; one pellet received 20 cycles, and one received 40 cycles.

We cored compression specimens and cylinders from each of these pellets. The specimens were nominally 6.2 mm in diameter and 6.2 mm long; each specimen was weighed before testing to ascertain density. Specimen densities were the same, $\approx 96\%$ of the theoretical density, independent of whether the material received 20 or 40 thermal cycles. Testing was performed at multiple temperatures: 800°C, 900°C, 1,000°C and 1,100°C and at a strain rate of $\dot{\epsilon} = 2.5 \times 10^{-4} \text{ s}^{-1}$. We typically tried to perform four experiments at each temperature, but in some cases we were limited to three tests because of the total number of specimens. The standard deviations in the strength were calculated for the multiple experiments at each given temperature. We have plotted the ± 1 standard deviation result in Figure 7 that shows ultimate compressive strength versus test temperature. One can see that there is a distinct increase of strength with temperature. This is exactly opposite the behavior of the plutonia and urania for which the strength decreases with increasing temperature.

Figure 7. Compressive strengths of cold-isopressed and thermally cycled hafnia as a function of temperature; ± 1 standard deviation is shown.

Because the trends in strength as a function of temperature are exactly the opposite, we will not be able to match the properties of hafnia and plutonia over a range of temperatures; rather, the properties will be the same only at a singular temperature where the trend lines intersect. In order to be able to estimate the properties of the hafnia at any particular temperature, we have fit a second-order polynomial through the ± 1 standard deviation values shown in Figure 7. This establishes an error band that we can use to estimate the strength for an arbitrary temperature. For example, at 975°C, the strengths of the hafnia that received 20 and 40 thermal cycles should be between 202 and 282 MPa, and 135 and 190 MPa, respectively. These estimates of ultimate compressive strength are shown in Figure 7. In Figure 8, we have plotted the error bands for the cold-

pressed and sintered material, which was thermally cycled, together with the strength data for plutonia. One can see that at low temperatures, $\leq 1,000^\circ\text{C}$ where the plutonia is reasonably strong, the properties of the two materials are relatively close. However, at higher temperatures, $>1,000^\circ\text{C}$, the hafnia is much too strong.

3.2.4. Mechanical Properties: Hot-Pressed Material

We found that the strength of the hot-pressed hafnia was much less than that of the cold-pressed and sintered material. These data are shown in Figure 9. These compression data were acquired from samples that had been hot pressed and then given 20 thermal cycles. The temperature cycling profile was identical to that discussed for the cold isostatically pressed pellets, a four-minute cycle between 1,640°C and 1,745°C at the pellet core. The strength of this material was only 65 MPa at 1,000°C. Its strength increased slightly with increasing

Figure 8. A comparison of the compressive strengths of the cold isopressed hafnia to $^{238}\text{PuO}_2$.

Figure 9. Strength of the hot-pressed hafnia. The hafnia had been thermally cycled and in one case sintered before testing. We show $^{238}\text{PuO}_2$ data for comparison.

temperature to about 100 MPa at 1,300°C. Such low strengths are suitable for very high temperature simulation of the behavior of $^{238}\text{PuO}_2$, $\approx 1,200^\circ\text{C}$, but the material is much too weak at the low temperatures of concern, for example, 975°C . We found that if we performed a short-time, low-temperature sinter on one of these hot pressed and thermally cycled pellets, we could increase its strength to the point that it might be an acceptable simulant for plutonia at 975°C . The sinter was performed by holding the compression specimens at $1,300^\circ\text{C}$ for 0.5 h before they were tested. After the $1,300^\circ\text{C}$ hold, the specimens were cooled to the appropriate test temperature, held for 5 min to allow the temperature to stabilize, and then tested. These results are also shown in Figure 9. One can see that the 30-min sinter at $1,300^\circ\text{C}$ increased the compressive

strength of the hafnia to about 125 MPa. The strength of the hafnia after this treatment was the same for temperatures between $1,000^\circ\text{C}$ and $1,300^\circ\text{C}$. We performed a similar sintering treatment for 2 h; however, the longer sintering time did not result in any additional strengthening. All of these experiments were performed at the same strain rate as was used to test the cold isostatically pressed and sintered samples, $\dot{\epsilon} = 2.5 \times 10^{-4}\text{s}^{-1}$.

3.2.5. Fractographs of the Hot-Pressed and Isostatically Pressed and Sintered Compression Cylinders

Low magnification optical micrographs have been taken of both the hot-pressed (plus 20 thermal cycles, then 30 min at $1,300^\circ\text{C}$) and cold isostatically pressed (plus 20 thermal cycles) hafnia that were tested at $1,000^\circ\text{C}$, $\dot{\epsilon} = 2.5 \times 10^{-4}\text{s}^{-1}$.

Representative pictures are shown in Figures 10a and 10b, micrographs of the hot and cold isostatically pressed material, respectively. One can see that both materials split into slivers and fine particles. There appears to be a range of particle sizes for both material conditions, and the shapes and aspect ratios of the particles are similar for the two conditions. The large particles have aspect ratios between approximately 3:1 and 7:1. We did not perform a quantitative size analysis on these particles, and from the photographs it is difficult to do more than estimate a weight fraction distribution. However, there appear to be fewer coarse particles in the case of the higher-strength

Figure 10a. Micrograph of the remnants of a compression test at 1,000°C. The hafnia was prepared by hot pressing.

Figure 10b. Micrograph of the remnants of a compression test at 1,000°C. Preparation was by cold isostatic pressing and sintering.

(cold-pressed) material. The elastic energy stored in the compression specimens drives the fragmentation. Thus, one might expect that the higher-strength material, with more elastic energy, would fracture into finer pieces.

4. IMPACT RESPONSE OF HfO_2

4.1 Impact Tests

Impact tests of bare GPHS capsules are conducted to determine the response of bare clads that have been stripped from their graphite components. The average terminal velocity of a GPHS module has been measured at 50.3 m/s, and typically 10% is added to this to produce test impact velocities of 54 to 55 m/s.⁽²⁴⁾ The temperature of the heat source in its launch configuration

is approximately 1,091°C; therefore, impact tests are often conducted at this temperature. Other impact test temperatures correspond to the predicted impact temperatures of heat sources under prompt reentry (919°C) and orbital-decay reentry (975°C) conditions.

The three hafnia impacts we conducted used a “different” hafnia material in each case. The hafnia pellet used, HF-1, for experiment BCI-HA1, clad ID SC0131, was hot pressed, given 20 thermal cycles, and then sintered at 1,300°C for 30 min. The materials HF-29 and HF-31, BCI-37 clad ID SC0144 and BCI-38 clad ID SC0145, respectively, were both cold isostatically pressed and sintered at 1,500°C. The HF-29 pellet was given forty thermal cycles and the pellet HF-31 twenty cycles. As we have seen, these treatments produced material that had very different strength characteristics.

4.2. Pretest Data

After the hafnia pellets were encapsulated in iridium-clad vent sets, the dimensions of the clads were recorded. The clads were then submitted for ultrasonic testing (UT) of the girth weld. Table I lists the clad vent sets and the dimensions of the hafnia simulant-fueled GPHSs used in this study. Table II lists the UT results.

4.3. Impact Testing

We selected a bare-clad, side-on impact condition for our three controlled hafnia experiments. This impact configuration allowed for a direct comparison to previous experiments using $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$ fuel performed with the same configuration. Such a configuration allows for the direct comparison between fuels because the experimental results are not complicated by the presence of multiple fueled clads or other components of the heat source, GIS or FWPF aeroshell. The impact temperature and velocity were picked to match the

TABLE I. Hafnia-Fueled Test Clads

Clad ID	Pellet ID	PICS ^a No.	Vent Cup Diam, in.	Weld Diam, in	Shield Cup Diam, in.	Length, in.
SC0131	HF-1	9808-35-2339	1.165	1.169	1.165	1.184
SC0144	HF-29	9808-01-1941	1.168	1.172	1.166	1.173
SC0145	HF-31	9808-01-1942	1.169	1.168	1.163	1.176

^a Post-impact containment shell.

TABLE II. Hafnia-Fueled Test Clad UT Results

Test ID	Clad ID	Max. UT Ind., Equiv. mils ^a	Location, deg ^b	UT Capsule Disposition
BCI-HA1	SC0131	3.3	317	Accept
BCI-37	SC0144	2.9	94	Accept
BCI-38	SC0145	3.4	342	Accept

^a Equivalent mils—The UT response is calibrated against a standard with machined defects that simulate cracks. The UT indication here is the calibrated response to a defect measuring the listed mils (1/1,000 in.).

^b Angular location referenced to the weld start at 0 deg.

environment predicted for an orbital-decay reentry, a temperature of 975°C, and the terminal velocity of the bare clad plus 10%, 54 to 55 m/s.

The clads impacted in the bare-clad tests were placed on a support cradle fabricated of molybdenum sheet. This support cradle was designed to hold the clad in the desired impact orientation (with the axis of the clad parallel to the target) and was outgassed in a vacuum furnace at 900°C for 1 h before use. The cradle was placed in a tantalum can (previously outgassed under the same conditions as the support cradle) and welded shut in an inert atmosphere. The can was evacuated and backfilled with He to approximately 3 psia through a port in the bottom of the can and then sealed. The can was transported to the Los Alamos Isotope Fuels Impact Test (IFIT) Facility.⁽²⁵⁾ Once loaded, the test components were heated to the desired temperature and then impacted against a hardened steel plate. The temperature of the clad was monitored by means of a thermocouple contacting the side of the can. We assumed that the clad surface temperature was equivalent to that of the tantalum containment can because of the high efficiency of radiant heat transfer at these temperatures. The specific test conditions for each test are given in Table III.

4.4. Post-Impact Evaluation

After each impact test, the test components were removed from the catch tube. The tantalum can containing the bare clad was opened. Fines were collected by rinsing the can interior and collecting the resulting liquid in a separate container. All test components were photographed, and the clads were measured to determine post-impact capsule strains. The size and location of all clad failures and cracks were recorded.

After macroscopic examination, the clads were opened and the patterns of fuel fracture were photographed. All of the bare-clad impacted clads were then defueled, and the fuel was submitted for particle size analysis.

4.5. Deformation and Cracking Results

The individual tests and postmortem examinations are summarized below. Table IV lists the engineering strains and number of breaches experienced by the impacted clads.

4.5.1. SC0131 Impact Response

Simulant-fueled clad SC0131 was impacted at 56.12 ± 0.42 m/s on a 1.0-in.-thick structural, steel-plate target. The clad temperature was 974°C. The impact face was centered at approximately 135 degrees from the weld start, Figure 11. The clad was

TABLE III. Impact Test Parameters

Test ID	Velocity, m/s	Temperature, °C
BCI-HF1	56.12 ± 0.42	974
BCI-37	53.83 ± 0.05	971
BCI-38	53.42 ± 0.06	970

TABLE IV. Hafnia-Fueled Clad Impact Test Summary

Clad ID		Maximum	Minimum	Number of Cracks	Amount of Fuel Released, g
SC0131	Diametral:				
	Vent Cup	8.04	-11.28	6	0.29
	Shield Cup	5.74	-11.25		
	Weld	8.48	-10.71		
	Axial Length	20.12			
SC0144	Diametral:				
	Vent Cup	5.48	-7.96	14	NM*
	Shield Cup	8.23	-12.69		
	Weld	9.04	-6.31		
	Axial Length	15.26			
SC0145	Diametral:				
	Vent Cup	8.72	-11.80	3	None
	Shield Cup	8.17	-10.40		
	Weld	9.16	-10.02		
	Axial Length	14.29			

* Release quantity was too small to be weighed or sampled for particle size analysis.

breached by six cracks, and 0.29 g of fines were recovered from the impact can. There was no detectable deformation of the steel target. The capsule strains are listed in Table IV.

The clad deformation and breaches were atypical of urania-fueled bare fueled clads impacted at this velocity.⁽²⁶⁾ The deformation, particularly in the axial engineering strain, is very large. Typical maximum axial strains measure no greater than 17%, but the strain of the impacted clad is greater than 20%.

Clad SC0131 had six breaching cracks, but typically, bare clads, fueled with urania, and impacted under identical conditions, experience no more than three breaching cracks.

The largest breach was located in the vent cup and transected the vent. This crack measured approximately 18.66 mm in length and 2.73 mm at its maximum width. One end of this crack terminated at a transverse vent cup impact face crack. Four of the six breaching cracks were located on the impact

Figure 11a.

Figure 11b.

Figure 11c.

Figure 11d.

Figure 11. SC0131 was breached by six cracks. (a) impact face, (b) trailing face, (c) profile, (d) opposite profile, (e) vent end, (f) blind end, (g) fuel fragmentation.

Figure 11e. *Figure 11f.* *Figure 11g.*

face. Two transverse cracks were located on the radii of the ends of the impact face; one on the vent cup, the other on the shield cup. The vent cup crack measured approximately 23.05 mm long, 1.48 mm at its maximum width. The shield cup crack measured approximately 9.34 mm long, 0.42 mm at maximum width. Two hairline axial cracks were located directly next to each other on the impact face. One of these transected the girth weld and measured approximately 10.03 mm in length. The other measured approximately 5.94 mm in length, one end terminated at the weld centerline.

The weld was breached by a hairline weld centerline crack extending from approximately 60 to 110 degrees from the weld start. This crack measured approximately 12.41 mm in length.

Figure 12a.

Figure 12b.

Figure 12c.

Figure 12d.

Figure 12. SCO144 was breached by several cracks. (a) impact face, (b) trailing face, (c) profile, (d) opposite profile, (e) vent end, (f) flint end, (g) steel target, (h) fuel fragmentation.

Figure 12e.

Figure 12f.

Figure 12g.

Figure 12h.

4.5.2. SC0144 Impact Response

Simulant-fueled clad SC0144 was impacted at 53.83 ± 0.05 m/s on a 1-in.-thick structural steel-plate target. The clad temperature was 971°C. The impact face was centered at approximately 0 deg from the weld start. The clad had numerous cracks; three transverse and five axial impact face cracks, a weld centerline crack, and five axial cracks on the trailing face, Figure 12. Of these, a shield cup transverse crack and an axial crack that transected the girth weld on the trailing face appear to have breached the clad. A minute amount of fines was observed in the impact can. There was detectable deformation of the target. A small indentation, measuring approximately 10.69 mm long, 2.98 mm at its widest point, and 0.152 mm deep, was observed near the center of the target.

The impact response of this clad was also atypical. The bare-clad impact caused deformation of the steel target. This has not been observed in other uranium-fueled, bare-clad impact tests.⁽²⁶⁾ The total number of observed cracks is also atypical. The typical number of cracks range from 0 to 3 at this impact velocity.⁽²⁶⁾ The deformation of the clad is comparable to that observed in previous simulant-fueled, bare-clad impact tests. The maximum percent strain varies between 12 to 16 axially, and diametrically 6 to 18 in the vent cup, 6 to 18 in the girth weld area, and 5 to 9 in the shield cup.⁽²⁶⁾

There were three transverse cracks on the impact face. Two of the transverse cracks were located on the edges of the impact face on the vent-cup radius and the third on the shield-cup radius. The two hairline vent-cup cracks measured approximately 5.79 mm and 4.06 mm long. The shield-cup crack

measured approximately 10.59 mm long and 0.25 mm at its maximum width.

Five parallel hairline axial cracks were located on the impact face, each transecting the clad girth weld. Two of these were located at approximately 10 degrees from the weld start on the edge of the impact face. These measured approximately 13.44 mm and 6.22 mm. The larger of the two cracks was closest to the impact face edge. The other three impact face cracks were located on the opposite impact face edge at approximately 345 deg from the weld start. These cracks, listed in ascending order of proximity to the impact face edge, measured approximately 11.35 mm, 5.97 mm, and 5.00 mm.

One hairline weld failure was located on the trailing face, running approximately from 135 to 180 deg and measuring approximately 12.88 mm in length.

Five axial cracks were located on the trailing face of the clad, four centered across the girth weld. The largest of the transecting cracks was located at approximately 180 deg and measured approximately 11.79 mm long and 0.559 mm at its maximum width. The other three weld transecting cracks were located at approximately 120, 135, and 200 deg. These measured approximately 0.483 mm, 0.240 mm, and 6.45 mm long respectively. The remaining hairline axial crack was located at approximately 230 deg in the shield cup just below the girth weld centerline and measured approximately 2.39 mm.

4.5.3. SC0145 Impact Response

Simulant-fueled clad SC0145 was impacted at 53.42 ± 0.06 m/s on a 1-in.-thick structural steel plate target. The clad temperature was

970°C. The impact face was centered at approximately 0 deg from the weld start. The clad was breached by one transverse impact face crack and two axial cracks, one on the impact face and one on the trailing face, Figure 13. Both axial cracks transected the girth weld. No released fines were observed in the impact can. There was detectable deformation of the target. A small indentation, measuring approximately 20.02 mm long, 8.23 mm at its widest point, and 0.381 mm deep, was observed near the center of the target. A ridge was observed on the impact face and appeared to correspond to the target indentation. This ridge measured approximately 11.56 mm long and 6.25 mm at its widest point.

Although the target impact response from this clad was atypical, the clad deformation was the most comparable to the responses of $^{238}\text{UO}_2$ simulant-fueled and $^{238}\text{PuO}_2$ bare-clad impacts. The total number of cracks is also comparable. The bare clad impact, however, caused the greatest deformation of the steel target. Such deformation had not been observed in other uranium-fueled, bare-clad impact tests.

The transverse impact face crack was located on the vent cup at the cup radius. This crack measured approximately 11.56 mm long and 0.813 mm at its widest point. A hairline axial crack, measuring approximately 3.12 mm, was also located on the impact face. This crack was centered across the girth weld at approximately 15°.

The axial trailing face crack was located at approximately 200°. This crack extended from the vent cup grit blast line through the weld and into the shield cup and measured approximately 11.48 mm long and 0.51 mm at its maximum width. The breach appears to have been caused by the development of

tensile strains in the trailing face parallel to the girth weld resulting from simulant fuel fragment displacement. Numerous ridges that appear to have been caused by the pellet fragmentation and displacement were observed on the trailing face and profiles of the clad.

4.6. SUMMARY OF PARTICLE WEIGHT FRACTION DATA

An important aspect of an impact experiment or field test that results in a clad rupture is the fragmentation of the fuel. The degree of fragmentation, particle weight, and size distribution, will control fuel dispersal through the clad rupture. We have assessed how our hafnia fuel compares to the actual $^{238}\text{PuO}_2$ fuel and the current $^{238}\text{UO}_2$ simulant based on these and previous side-on impact experiments. The conditions used for the hafnia impacts have been discussed. In the case of the $^{238}\text{PuO}_2$ fuel, three bare-clad, side-on impacts had been performed previously at 1,090°C and, nominally, 55 m/s. Four bare-clad experiments had been performed using the $^{238}\text{UO}_2$ simulant fuel in a side-on configuration. The test temperature and impact velocity were 1,090°C and 55 m/s, respectively.

Particle weight fraction data from all of the experiments are summarized below in Tables V through VII. These tables contain not only the particle weight fraction distributions but the mean and standard deviation of the experimental data for a given sieve size. For the $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$, for which the experiments were identical, it is a straightforward process to generate the mean and standard deviation.

Figure 13a.

Figure 13b.

Figure 13c.

Figure 13d.

Figure 13. SC0145 was breached by one transverse crack and two axial cracks. (a) impact face, (b) trailing face, (c) profile, (d) opposite profile, (e) vent end, (f) blind end, (g) steel target, (h) fuel fragmentation.

Figure 13e.

Figure 13f.

Figure 13g.

Figure 13h.

TABLE V: Weight Fraction of $^{238}\text{PuO}_2$ Particles

Sieve Size (microns)	BCI-33 55.23 m/s FC0334	BCI-35 55.72 m/s FC0339	BCI-15 53.34 m/s FC214	Mean Particle Weight Fraction	1 Standard Deviation in Wt Fraction
5600	0.3342	0.3628	0.3837	0.36023	0.02485
2000	0.3962	0.3913	0.3677	0.38507	0.015238
850	0.1609	0.1316	0.1199	0.13747	0.02112
425	0.0412	0.0377	0.0412	0.040038	0.0020207
180	0.0199	0.019	0.0254	0.021433	0.0034646
125	0.0042	0.0045	0.0065	0.005067	0.00125
75	0.0081	0.0093	0.0092	0.008867	0.000666
45	0.0180	0.0215	0.012	0.017167	0.004805
30	0.0056	0.0029	0.0135	0.007333	0.005509
20	0.0058	0.0059	0.0113	0.007667	0.003147
10	0.0040	0.0055	0.0059	0.004167	0.001258
<10	0.0019	0.0080	0.0037	0.004533	0.003134

Total: 1.0 1.0 1.0 1.0

TABLE VI: Weight Fraction of $^{238}\text{UO}_2$ Particles

Sieve Size	CPV-1	CPV-5	CPV-9	CPV-10	Mean Particle Weight Fraction	1 Standard Deviation in Wt Fraction
5600	0.3765	0.3952	0.2482	0.3854	0.3513	0.069173
2000	0.3309	0.3482	0.4551	0.3657	0.3750	0.055274
850	0.1822	0.1504	0.2075	0.1758	0.1790	0.023457
425	0.0108	0.0556	0.0552	0.0436	0.0413	0.021081
180	0.0495	0.0178	0.0216	0.0156	0.0261	0.015779
125	0.0225	0.0039	0.0026	0.0031	0.0080	0.009665
75	0.0026	0.004	0.0018	0.0017	0.0025	0.001063
45	0.0018	0.0031	0.0016	0.0014	0.0020	0.000768
30	0.0009	0.0006	0.0000	0.0000	0.0004	0.00045
20	0.0014	0.0017	0.0000	0.0000	0.0008	0.000903
10	0.0047	0.0073	0.0003	0.0010	0.0033	0.003279
<10	0.0162	0.0122	0.0061	0.0067	0.0103	0.004797

Total: 1.0 1.0 1.0 1.0 1.0

The tabulated results for particle size are plotted in Figures 14, 15, and 16. We are plotting ± 1 standard deviation as an error bar versus sieve mesh size. The results for $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$ are shown in Figure 14. One can see that there is much less deviation in the plutonia data than in that for urania. However, the weight distributions as a function of sieve size are identical. With the exception of the $850\ \mu\text{m}$ sieve, the “error bars” for the plutonia lie within those of the urania. Because of the consistencies in all of the other data, we believe that this difference is the result of accumulated error and not a significant perturbation. The data for hafnia are plotted together with that for plutonia in Figure 15. We have plotted the data from each of the three hafnia impact experiments separately with the ± 1 standard deviation error bars for the plutonia impacts. One can see that there is no systematic variation of the hafnia data with material strength. Rather, the weight fraction distributions appear very consistent irrespective of strength. This result appears to conflict with Figures 10a and 10b where, at least visually, the particle distributions appear different, but one must remember that the impact experiments are dynamic. In a dynamic experiment there is kinetic energy associated with the impacting body. This energy is the same for all three hafnia tests and is much greater than the stored elastic energy in the particles. Thus, the energy available to fragment the pellets on impact is nearly equivalent for the three experiments irrespective of material strength, hence the consistent weight-fraction distributions.

Because the hafnia particle weight-fraction data are so similar, we have taken the three hafnia experiments as a single population group and calculated the standard deviations for the individual weight fractions. These results are plotted with the plutonia data in

Figure 16. One can see that the standard deviations in the hafnia data are comparable to those for the plutonia and urania. This validates our assumption that the results from the three hafnia experiments can be combined and viewed as a single experimental series. One can see that there are very distinct differences between the hafnia and plutonia results. There is a smaller weight fraction of material for the two largest sieve sizes and correspondingly a larger weight fraction of material for sieve sizes between $125\ \mu\text{m}$ and $850\ \mu\text{m}$. These data are systematic, and we do not believe them to be the result of experimental error or inconsistencies between experiments. Finally, when one examines Figures 14 and 16, one can see that the experimental error in the weight-fraction measurements, as represented by the standard deviation in the experiments, is equal to the mean for sieve-mesh sizes of $125\ \mu\text{m}$ and below. In other words, our error is as large as the quantity that we are measuring. Although it is important to try and assess the weight fractions of very fine particles, this is a difficult measurement because the quantities of material are so small.

4.6.1. Correlation between the Weight Fraction of Material and the Number of Particles

In order to know the importance of the differences in the HfO_2 and $^{238}\text{PuO}_2$ weight-fraction data, we must be able to estimate how many particles are associated with a given weight fraction of material, knowing the sizes of particles. For example, a very small difference in the number of large particles could produce a large difference in the weight fraction of material. Such a

TABLE VII: Weight Fraction of HfO₂ Particles

Sieve Size (microns)	SC131	SC144	SC145	Mean Particle Weight Fraction	1 Standard Deviation in Wt Fraction
5600	0.2076	0.1651	0.2232	0.1986	0.03007
2000	0.3642	0.2907	0.3450	0.3333	0.038121
850	0.1807	0.2426	0.2331	0.2188	0.03334
425	0.0814	0.0988	0.0838	0.0880	0.009430
180	0.0683	0.0444	0.0458	0.0528	0.013413
125	0.0406	0.0077	0.0068	0.0184	0.019457
75	0.0491	0.0070	0.0081	0.0214	0.023995
45	0.0066	0.0049	0.0058	0.0058	0.000850
30	0.0002	0.0000	0.0000	0.0001	0.000115
20	0.0001	0.0000	0.0002	0.0001	0.0001
10	0.0001	0.0000	0.0048	0.0016	0.00277
<10	0.0011	0.1388	0.0434	0.0611	0.070536
Total:	1.0	1.0	1.0	1.0

Figure 14. Experimentally measured weight fractions of plutonia and urania for the different sieve sizes.

difference in the number of a few large particles is likely to be relatively unimportant.

In order to calculate the number of particles associated with a particular weight-fraction population group, we must make three engineering assumptions: the particle shape, the size of particle that can fit through a given mesh, and the distribution of weights as a function of particle size for a given weight-fraction population group. For the purpose of this analysis, we will examine two different particle shapes: a particle with a 5:1 aspect ratio (a needle shaped particle) and a particle with a 1:1 aspect ratio, (a spherical particle). We believe that these two shapes represent bounding conditions for the particles that result from our impact experiments. The dimensions of a particle that can fit through a particular sieve are a function not only of the sieve size but of the particle shape. It is obviously more difficult

Figure 15. Experimentally measured weight fractions of plutonia and hafnia. Individual data points are plotted for the hafnia.

Figure 16. Experimentally measured weight fractions of plutonia and hafnia for the different sieve sizes.

to pass a needle-shaped particle through the sieve than a spherical particle. We assumed that the maximum diameter of a spherical particle that passes the sieve will be equal to the sieve size. In the case of the 5:1 aspect-ratio (needle-shaped particle), we assumed that the maximum diameter would be equal to 70% of the sieve size. Thus, the length of this particle is 3.5 times the sieve spacing. The sizes and shapes of the particles with respect to the sieve spacing, S_i , are shown schematically in Figures 17a and 17b.

Finally, we have assumed that the distribution of weights is uniform with respect to particle size within a given weight-fraction population group. Thus, because sizes of particles vary within a particular weight-fraction group, the distribution of the number of particles will not be uniform within a given grouping. This is depicted for the i^{th} weight-fraction population group, again schematically, in Figure 18. The distribution of weights is denoted W_i and the

number distribution is $n_i(x)$. That is, the number of particles is a function of their size, x . The weight-fraction population group is between sieve mesh spacings $D1_i$ and $D2_i$.

In order to estimate the number of particles in each weight-fraction group, we need to calculate the “mean” diameter of the particles in that group. Then, knowing the total weight of a population group and the density of the material, we can calculate the total number of particles in that group. We can calculate the average diameter using the principle of calculating a moment of the number distribution. The equations that we have used to perform these calculations are derived in Appendix I. The solutions to the equations give the number of particles for each particular weight-fraction group N_i , in terms of the total pellet weight, W_p , weight fraction of material, f_3 , the material density, ρ , and the mean particle $\bar{d}_i$. Note that the mean particle diameter will be a function of the

particle shapes because of our assumptions concerning the sizes of particles that will pass through the sieves:

$\bar{d} = 2/[(1/D1_i) + (1/D2_i)]$ $D1$, and $D2$, are the smallest and largest particle diameters for the i^{th} weight-fraction group. For the particles

with a 5:1 aspect ratio, $N_i = \frac{4}{5} \frac{f_i W_i}{\rho \pi \bar{d}_i^3}$, and in the case of the spherical particles,

$N_i = \frac{6}{\pi} \frac{f_i W_i}{\rho \pi \bar{d}_i^3}$, The densities, total weights, and mean particle diameters that were used in these calculations are compiled in Appendix II.

4.6.2. Numbers of Particles

Figure 19 is a plot of the number of particles, associated with the particular weight-fraction groups, plotted against the mean particle diameter of that group. The error bars are again ± 1 standard deviation. The data and calculations appear to be very reliable at and above a mean diameter of 180 μm . When the particle diameter is 125 μm and below, 1 standard deviation in the data is equal to the mean value. Thus, there are very large errors in the number of particles that we calculate. Because of the very small volumes of material associated with a particle that is 125 μm in diameter, even a small weight fraction of material (for example, 4%) constitutes over 10^5 particles. It is difficult to measure such a small amount of material accurately, and therefore the standard deviation can equal the mean value. Thus, the weight fraction can be between 0% and 4%. In this case, the scatter in the number of particles is between 1 and 10^5 . These difficulties can be clearly seen in Figure 19.

We now compare the estimated number of particles between the plutonia and the hafnia. There are very few particles associated with the group that has the largest size, the 5,600- μm sieve. In the case of the 5:1 aspect-ratio shape, this group has at most 15 particles. Thus, although the weight-fraction data can be very different, the difference in the absolute number of particles is small because each particle is massive. The weight fraction of plutonia of the largest size constitutes 12 to 14 particles with a 5:1 aspect ratio. In the case of the hafnia, there are between 6 and 9 particles. This is not a large difference. The potential difference as a result of particle shape is much more significant. If the particle shape is a sphere rather than a 5:1 aspect ratio cylinder there are four times as many particles.

There was a measurable higher weight fraction of hafnia in the smaller particle sizes, between 180 μm and 850 μm , above the error in the experiment. This higher weight fraction was only about 5%; however, in terms of the number of particles (5:1 aspect ratio 177 μm mean diameter) the difference is substantial, 15,000 particles of $^{238}\text{PuO}_2$ to 40,000 particles of HfO_2 . The difference between the numbers of cylindrical and spherical particles was again four times. For the smaller particles, the potential difference in shape is approximately equivalent to the difference in the weight-fraction data between the hafnia and plutonia.

5. DISCUSSION

5.1. Thermal Behavior

One cannot alter the thermal properties of a material as we did with the mechanical behavior of the HfO_2 by thermal cycling.

Figure 17a. Schematic of a 5:1 aspect-ratio particle with respect to the sieve spacing

Figure 17b. Schematic of a 1:1 aspect-ratio particle with respect to the sieve spacing.

Figure 18. The assumed weight distribution for a given weight-fraction population group. Note the number distribution.

Rather, one must work with the existing properties and select boundary conditions, temperatures, and heating rates that will give the desired results for the known density, thermal conductivity, and specific heat. This can be done in a relatively straightforward manner with the modern finite element codes, FEM. One can calculate the performance of the $^{238}\text{PuO}_2$ for the desired boundary conditions, heating rates, etc. These boundary conditions can then be altered in the FEM code until the same behaviors are predicted for the hafnia. In the case of the hafnia, the thermal characteristics are sufficiently different as to require this approach. Although this requires effort and computational time, it should not prevent the use of hafnia as a simulant fuel.

This procedure was unnecessary in the case of the $^{238}\text{UO}_2$ simulant fuel because the thermal properties of the urania and plutonia match so closely. In this respect, the urania is a very good simulant.

5.2. Strength and Impact Response

In the course of this study, evaluating hafnia as a simulant fuel for $^{238}\text{PuO}_2$, we have

also examined and summarized strength and impact data for $^{238}\text{UO}_2$. We have found that urania is an excellent simulant fuel. The strengths of $^{238}\text{UO}_2$ that we measured in laboratory compression experiments were nearly identical to those of $^{238}\text{PuO}_2$, as well as the variation of strength with temperature. This similarity of behavior was also reflected in the identical side-on impact response of $^{238}\text{PuO}_2$ and $^{238}\text{UO}_2$ fueled clads. The deformations, vent cup, shield cup, weld and length strains, were the same, irrespective of the fuel. In addition, the number and locations of clad breaches were the same. Finally, fuel fragmentation as measured by particle size analysis was equivalent for the $^{238}\text{UO}_2$ and $^{238}\text{PuO}_2$. The error bars for the plutonia particle weight-fraction data for a particular sieve size consistently lie within the error bars characterizing the urania.

From a practical viewpoint, the mechanical response of the urania and plutonia are indistinguishable. If there were no issues with the very low levels of radioactivity associated with the $^{238}\text{UO}_2$, it would be an excellent simulant for $^{238}\text{PuO}_2$ to be used in field tests.

Figure 19. The calculated numbers of particles of a given mean diameter for the plutonia, urania, and hafnia.

Hafnia's properties do not match those of plutonia nearly as well as do urania's. However, the differences are small enough that we believe there is the potential for its use as a simulant fuel if depleted urania is unacceptable from a radiological viewpoint.

We have been able to tailor the strength of HfO_2 to match that of $^{238}\text{PuO}_2$ at a particular temperature. This was accomplished by thermal-cycling the hafnia across a tetragonal/monoclinic phase boundary. Because of a volume change accompanying the phase transformation, microcracking is introduced. The extent of microcracking, which is a function of the number of thermal cycles, determines the material's strength. Thus, the strength can be modified to match that of plutonia. One difficulty with this process is that the strength of the thermally cycled HfO_2 increases with testing temperature. This is exactly the opposite of plutonia for which the strength decreases with increasing temperature. Thus, the hafnia must be given a very specific treatment depending on the temperature at which one wants to conduct a field experiment. We have seen that this is not an insurmountable task in preparing for the hafnia impact experiments at 975°C.

Our hafnia-fueled, single-clad impact tests approached the behavior that had been previously observed for plutonia-fueled clads, but there were some differences. The hafnia pellet fabricated by cold pressing and sintering followed with 20 thermal cycles came the closest to simulating the impact response of plutonia. The clad fueled with this pellet experienced the same strains, numbers and sizes of breaches, and locations of breaches that have been observed for clads fueled with $^{238}\text{PuO}_2$. There were differences, however. This particular fueled clad, cold pressed and sintered plus twenty thermal

cycles, left appreciable deformation in the hardened-steel target plate. This had not been observed in any previous plutonia or urania-fueled, side-on, bare-clad impacts. A weaker hafnia fuel, BCI-37 SC0-144, that produced an excessive number of breaches and fractures, also dented the target. We do not know the reason for this result at this time.

The particle sizes of the hafnia fuel after impact were also similar but not exactly the same as for the plutonia. The greatest difference in the weight-fraction distribution between HfO_2 and $^{238}\text{PuO}_2$ was for the largest particles. There was a smaller amount of the largest hafnia particles. This difference, however, was very small if one were concerned with the number of particles rather than their weight fraction. Corresponding to the smaller hafnia weight fraction of the largest particles was a slightly larger weight fraction of small-to-medium-size particles. This difference in weight fraction was not particularly large. However, because these particles are small, a slight difference in weight fraction corresponds to a larger difference in absolute number.

Although the thermal, mechanical, and impact performance of the hafnia is a reasonable approximation to plutonia, it is not close enough for us to recommend it as a substitute fuel at this time. The differences that we have observed give one reason to pause. However, the behaviors of HfO_2 and $^{238}\text{PuO}_2$ are close enough that we believe further experimentation is warranted. With more data, the evidence will likely be strong enough to justify using hafnia as a simulant fuel. We recommend that the additional follow-on experiments that focus on the impact behavior be run.

1. Duplicate experiments BCI-37 and BCI-38, cold-pressed and sintered hafnia

pellets that have been given 40 and 20 thermal cycles respectively. This test series is to confirm our previous results.

2. Based on the bare-clad, side-on tests, pick a single condition that most closely matches the impact response of the plutonia. Prepare four fuel pellets to this condition for loading into GISs and an FWPF aeroshell. Finally, conduct an impact test on the full aeroshell assembly. We recommend an edge-on impact configuration, with the edge containing the flight-control bevels as the impact face. This configuration will result not only in fueled-clad/target interactions but fueled-clad/fueled-clad interactions. This will be an extremely demanding test of the hafnia as a simulant fuel.

If our bare-clad, side-on impact tests are consistent with the previous results and the fueled aeroshell assembly experiment duplicates previous results for an identical $^{238}\text{PuO}_2$ test, we can confidently recommend hafnia as a simulant fuel for $^{238}\text{PuO}_2$ in field experiments.

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**PROPERTIES OF HAFNIUM OXIDE WITH RESPECT TO
ITS USE AS A SIMULANT FOR $^{238}\text{PuO}_2$ IN SAFETY STUDIES**

APPENDIX I

In order to interpret the importance of the differences in the weight-fraction data we have performed an engineering analysis to determine approximately the number of particles that correspond to each weight-fraction population. For this analysis, three assumptions are required: a particle shape; the size of the particles that can fit through a particular mesh; and finally, the distribution of weights over the particle sizes within a weight-fraction population group.

From our compression experiments, we found that the particles fractured into cylindrical shapes with a height-to-diameter aspect ratio (h/d) between 3 and 7. The compression experiments were end-on rather than side-on as were the impact tests, but we think that it is reasonable to assume that the fragments from these two experiments have roughly the same shape. Thus, we assumed, as an upper bound, that the particles were cylinders with a height-to-diameter aspect ratio of 5:1. To establish a lower bound for particle shape we have also performed the analysis for a spherical particle.

It is very difficult to know what diameter cylinder of high aspect ratio can fit through a particular mesh size. Obviously, the particle would have to have a diameter that is a fraction of the mesh spacing. We have concluded that it would be reasonable to assume that the largest cylindrical particle that could fit through a mesh would be one with a diameter 70% of the mesh spacing. In the case of the spherical particles, the largest particle would have a diameter equal to the mesh size. This was shown schematically in Figures 17a and 17b.

There is a range of particle sizes within each weight-fraction population group, and we do not know the mass distribution over particle size within the weight-fraction group. For the purposes of the calculation, we assumed that this distribution was uniform. In other words, the mass of large particles is equal to the mass of small particles within a single grouping. Thus, there will be a nonuniform number distribution of particle sizes within a single group. There have to be fewer large particles and more small particles so that the mass of all sizes is the same. Figure 20 shows both the weight fraction and number distributions for the i^{th} weight fraction group, containing particles with a minimum diameter of $D1_i$ and a maximum diameter of $D2_i$. The particle number distribution is indicated as $n_i(x)$ and the weight distribution by $W_i(x)$.

In order to estimate the number of particles in each weight-fraction group we need to calculate the “average” diameter of the particles in that group. Then, knowing the total weight of a population group and the density of the material, we can calculate the total number of particles in that group. We can calculate the average diameter using the principle of calculating a moment of the number distribution.

The weight of an individual cylindrical particle with diameter d and a 5:1 aspect ratio is given by

$$\rho \cdot 5d \cdot \frac{\pi d^2}{4},$$

where ρ is the materials density. For a spherical particle the weight is

$$\rho \cdot \frac{\pi d^3}{6},$$

The total weight of all of the particles with diameter d is $W_i(d)$ where,

$$W_i(d) = n_i(d) \cdot \rho \cdot 5d \cdot \frac{\pi d^2}{4},$$

or for spherical particles

$$W_i(d) = n_i(d) \cdot \rho \cdot \frac{\pi d^3}{6},$$

Because the weight distribution $[W_i(d)]$ was assumed constant across the entire population, we can write the following expression for the number of particles,

$$n_i(d) = \beta_i / d^3,$$

where β_i is a constant for the i^{th} group that is a function of the weight distribution and shape for that particular population of particles.

We can now solve for the “average” particle diameter, or centroid, of the i^{th} group. The principle behind this calculation of a moment is shown in Figure 20. The sum of

the moments of the number of particles at diameter d over d ($d \equiv x$) for all possible diameters is equal to the moment of the total number of particles over the average diameter $\bar{d}$. In integral form,

$$\bar{d} \int_{D1_i}^{D2_i} n_i dx = \int_{D1_i}^{D2_i} x n_i dx.$$

Substituting for n_i , one finds

$$\bar{d} \int_{D1_i}^{D2_i} \beta_i x^{-3} dx = \int_{D1_i}^{D2_i} x \beta_i x^{-3} dx, \text{ or}$$

$$\bar{d} = \frac{\int_{D1_i}^{D2_i} x^{-2} dx}{\int_{D1_i}^{D2_i} x^{-3} dx}.$$

Solving the integrals

$$\bar{d} = \frac{\frac{1}{D1_i} - \frac{1}{D2_i}}{\frac{1}{2} \left(\frac{1}{D1_i^2} - \frac{1}{D2_i^2} \right)}, \text{ and noting that}$$

Figure 20. A schematic representation of the use of the moment of particle number to obtain the mean particle diameter for a weight-fraction population group.

$$\frac{1}{D1_i^2} - \frac{1}{D2_i^2} = \left(\frac{1}{D1_i} - \frac{1}{D2_i} \right) \cdot \left(\frac{1}{D1_i} + \frac{1}{D2_i} \right),$$

we find

$$\bar{d} = \frac{2}{\left(\frac{1}{D1_i} + \frac{1}{D2_i} \right)}.$$

Thus, knowing the largest and smallest diameters of particles in each weight-fraction group we can solve for the average diameter of that group. The values of diameter that we will use for our calculation are based on 70% of the sieve-mesh sizes for the cylindrical particles and the mesh size for spherical particles. For the largest sieve size (5,600 μm) we assumed 6,000 μm was the largest particle diameter, $D2_i$.

From the average diameters, we can calculate the number of particles in each weight-fraction group. The total weight of the group will be the total number of particles in that group (N_i) times the density and average particle volume. The weight of the individual group divided by the total weight (W_i) will give us the weight fraction

(f_i). In algebraic form, for cylindrical and spherical particles respectively, the measured weight fraction can be expressed as

$$f_i = \frac{N_i \rho \cdot 5\bar{d} \cdot \frac{\pi \bar{d}^2}{4}}{W_i} \text{ and } f_i = \frac{N_i \rho \cdot \frac{\pi \bar{d}^3}{6}}{W_i}.$$

Solving for the total number of particles in the i^{th} group,

$$N_i = \frac{4f_i W_i}{5\rho \pi \bar{d}^3} \text{ (5:1 aspect-ratio cylinders)}$$

$$\text{or } N_i = \frac{6f_i W_i}{\rho \pi \bar{d}^3} \text{ (spheres).}$$

Using these relations we can now perform the calculations. Appendix II contains the values we used for total weights, densities, and diameters.

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APPENDIX II

The following physical constants were used to calculate the number of particles associated with particular weight fractions of material. One should note that the mean grain diameter $\bar{d}$ is a function of the sieve sizes and our assumptions concerning the particle shapes and sizes that will pass through a particular sieve. The values of $\bar{d}$ are

independent of material. To calculate the mean grain diameter for the largest sieve size, we had to assume a maximum particle diameter of the particles that did not pass through the 5,600- μm sieve. We assumed that this diameter was 6,000 μm .

<p>$\rho(\text{hafnia}) = 8.7659 \text{ g/cm}^3$, 90.56% theoretical density $\rho(\text{urania}) = 9.2064 \text{ g/cm}^3$, 84% theoretical density $\rho(\text{plutonia}) = 9.9596 \text{ g/cm}^3$, 87.21% theoretical density</p>		
<p>Average total weight, W_t: hafnia, 138.23 g urania, 144.75 g plutonia, 151.2706 g</p>		
Sieve Size	Mean Grain Diam (μm), $\bar{d}$, 5:1 Aspect-Ratio Cylinder	Mean Grain Diam (μm), $\bar{d}$, 1:1 Aspect-Ratio Cylinder
5600	4741.94	5793
2000	2063.16	2947
850	835.09	1193
425	396.67	567
180	177.02	252.9
125	103.28	147.5
75	65.62	93.75
45	39.375	56.25
30	25.2	36
20	16.8	24
10	9.33	13.3

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