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Fabrication of Cerium Oxide and Uranium Oxide Microspheres for Space Nuclear Power Applications

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Abstract. Cerium oxide and uranium oxide microspheres are being produced via an internal gelation sol-gel method to investigate alternative fabrication routes for space nuclear fuels. Depleted uranium and non-radioactive cerium are being utilized as surrogates for plutonium-238 (Pu-238) used in radioisotope thermoelectric generators and for enriched uranium required by nuclear thermal rockets. While current methods used to produce Pu-238 fuels at Los Alamos National Laboratory (LANL) involve the generation of fine powders that pose a respiratory hazard and have a propensity to contaminate glove boxes, the sol-gel route allows for the generation of oxide microsphere fuels through an aqueous route. The sol-gel method does not generate fine powders and may require fewer processing steps than the LANL method with less operator handling. High-quality cerium dioxide microspheres have been fabricated in the desired size range and equipment is being prepared to establish a uranium dioxide microsphere production capability.

Keywords: Sol-gel, internal gelation, space nuclear, nuclear fuel, microsphere.

INTRODUCTION

Nuclear power sources and radioisotope heaters have been used for decades to enhance or enable space exploration missions [1]. In regions where the solar flux is too low, or the radiation field is too high for solar cells to provide electricity, radioisotope thermoelectric generators (RTGs) have been used. Additionally, light weight heater units are used in strategic locations on spacecraft for thermal management. RTGs and heater units rely on decay heat from radioisotopes. More ambitious future space exploration missions propose the use of nuclear fission in nuclear thermal rockets (NTRs), nuclear-electric propulsion (NEP), and nuclear reactors supporting a lunar or Martian base. To date, the fuel used or developed for radioisotope systems, nuclear rockets, and space nuclear reactors are quite different.

Radioisotope Systems

All space radioisotope systems deployed by the United States have used Pu-238 as a heat source due to its favorable properties. With a half-life of 87.7 years, Pu-238 provides steady power levels over long mission times. Additionally, the use of Pu-238 minimizes system mass since it requires very little shielding to reduce radiation emissions to acceptable levels. To prevent the release of activity in the case of a launch vehicle failure followed by atmospheric re-entry and impact, the PuO₂ fuel pellets are encased in a welded iridium-alloy clad and placed in a carbon-fiber module to create what is called the General Purpose Heat Source (GPHS). Currently, the plutonium fuel is processed into pellets and clad in an iridium DOP-26 alloy at LANL and then assembled into the GPHS modules and RTG at the Space Battery Fabrication Facility at the Idaho National Laboratory (INL).

Plutonium-238 oxide fuels for RTGs have been fabricated at Mound Laboratory, the Savannah River Site (SRS), and are now processed at Los Alamos. Pellet production is a multi-step process that requires radiation workers and glove-box facilities to contain the high-activity plutonium [2]. Pu-238 powder is first dissolved in HNO₃/HF and

then precipitated as plutonium oxalate. Due to the size and morphology of the oxalate precipitates, ball milling followed by green pressing, granule formation with sintering, and final sintering of the compact is necessary to achieve high-quality sintered pellets [3]. Both the precipitated and ball milled plutonium oxide have a significant fraction of particles in the sub-micron to few micron size range [3]. Historically, fines were also produced during the subsequent granulation process as well as from sectioning, grinding, and polishing samples for metallographic analysis [4]. These fines are problematic from the standpoint of spontaneous contamination and, if released, inhalation by personnel.

The small size and high alpha activity of the Pu-238 oxide powders produced make it extremely difficult to contain [4, 5, 6]. Reports from personnel at the Plutonium Fuel Fabrication (PuFF) facility at Savannah River National Laboratory described Pu-238 oxide to exhibit gas-like characteristics, "flying" through the air, "walking" down pipes, and once released traveling hundreds of feet away from the breach rather than collecting directly downward on the floor [4]. It was noted at that time that Pu-238 oxide was unique from other fine oxide powders in that it did not exhibit the same tendencies to agglomerate into larger particles. The result was that submicron particles remained independent and were subject to very long settling times dictated by Brownian motion [4]. While it has been proposed that the propensity for the fine particulates to migrate was due to alpha recoil [4, 5], more recent analysis has suggested that the recoil energy from Pu-238 decay only exhibits a sufficient force to move particles with sizes of about 20nm or less [6]. However, the same study indicated that alpha decay tended to fracture particles into consistently smaller pieces, thus increasing their mobility. It was also theorized that alpha recoil is responsible for the migration of Pu-238 powders through HEPA filters, as was evidenced by the passage of multiple grams of Pu-238 through a first stage HEPA filter at SRS [6]. Workers at PuFF also found it necessary to change gloves on glove boxes regularly to prevent the release of Pu-238 [4].

Once released, the most direct pathway for plutonium oxide into the body is through inhalation. Particles smaller than 10µm tend to deposit in the lungs rather than being confined to the nasal passages [3]. The federal regulatory limit for Pu-238 inhalation for radiation workers is roughly equivalent to one 8µm diameter particle, making exposure limits very difficult to meet [3]. The corrosive and dispersive properties of $^{238}\text{PuO}_2$ have caused exposures in facilities at both SRS and LANL. After fabricating fuel for the Galileo and Ulysses missions, PuFF was placed in standby mode and decontamination of the hot cells was limited [6]. In 2000, $^{238}\text{PuO}_2$ powder was released from the hot cells and resulted in doses to seven workers, four of whom likely exceeded annual dose limits and underwent chelation therapy [3]. In 2003, the breach of a container due to corrosion resulted in the release of fine $^{238}\text{PuO}_2$ powder and exposure of two workers at LANL, resulting in delay in operations for eight months [3]. These accidents are attributable to the corrosive and migratory nature of fine plutonium powders and are a hazard to radiation workers as well as the schedule for programs using an RTG since decontamination of a facility is expensive and time-consuming.

Despite the difficulties of handling and containing Pu-238, approximately 300kg has been processed in the United States over the past five decades [6]. Space exploration missions utilizing radioisotope systems for power have had great success, as evidenced by the Voyager probes which have been operating since 1977. However, due to the contamination difficulties presented by Pu-238, it is desirable to assess aqueous routes to oxide fuels that don't involve the production of hazardous particulates.

Fission Systems

Over the past several decades, the United States has funded a number of development programs for nuclear thermal rockets and space power reactors. The use of a nuclear reactor is generally proposed for missions that would require tens of kilowatts of electrical power or more. For long-duration missions with high power requirements, especially far from the sun, the use of solar cells becomes increasingly difficult due to the surface area required. Furthermore, in locations such as the moon or Mars, long periods of night or dust storms complicate the use of solar cells. For propulsion applications, nuclear thermal rockets have been investigated due to their high specific impulse that allows for more efficient use of propellant and allows for lower system masses than those using chemical propellants. NTRs could also reduce transit times for manned missions, reducing astronauts' exposure to radiation and zero gravity [7]. In order to minimize mass, operate at elevated temperatures, and in some cases achieve high burnups, space power and propulsion programs have developed a wide variety of fuel forms for different fission applications.

Nuclear thermal rockets must be operated at extremely high temperatures and use hydrogen gas propellant to achieve a high specific impulse. In the ROVER/NERVA program of the 60s and 70s, uranium carbide (UC) fuel in a prismatic graphite matrix was used in order to push to temperatures of around 3000K [8]. During the same time period, the GE-710 Program and the ANL Cermet Nuclear Rocket program assessed the viability of uranium dioxide (UO_2) fuels in a refractory metal matrix [8]. Later, in the 80s and 90s, the Space Nuclear Thermal Propulsion (SNTTP) program investigated the use of a pebble-bed reactor with a TRISO-like fuel, which consisted of a UC kernel with pyrolytic carbon layers and a ZrC coating as a fission product barrier [8, 9]. In each of these programs, the fuel's ability to maintain integrity at elevated temperature without release of fission products was of importance.

Nuclear reactors designed to power space vehicles or a manned outpost can operate at much lower temperatures than an NTR, and thus more closely resemble mass-optimized experimental reactors where heat is transferred out of the core to an energy conversion system. Today, the SNAP-10A unit is the only fission power system that has been flown by the United States. Launched in 1965, SNAP-10A had Hastelloy N clad homogeneous uranium-zirconium hydride fuel with sodium potassium (NaK) coolant to transfer heat to thermoelectric converters, which produced about 500W_e [10, 11]. Many years later, there have been several programs aimed at developing space reactors in the 10-1000kW_e range that have not yet resulted in a test flight. The Space Power-100 (SP-100) initiative also planned to use thermoelectric conversion, but with a lithium working fluid and uranium nitride fuel in a rhenium-lined niobium-zirconium cladding [12]. The Safe Affordable Fission Engine-400 (SAFE-400) program focused on the use of molybdenum-sodium heat pipes to transfer heat from the reactor core to a Brayton converter for space power. The SAFE-400 was also designed using uranium nitride fuel with a rhenium coated cladding [13]. More recently, NASA's Project Prometheus was aimed at developing the capability for nuclear electric propulsion for missions like the Jupiter Icy Moons Orbiter (JIMO) before it was cancelled in 2005 [14]. The Prometheus reactor was to be gas-cooled with Brayton conversion and utilize uranium oxide in fuel pins; testing for the best cladding material was underway when the program was cancelled [14]. Currently, development is ongoing for the Fission Surface Power (FSP) unit, which would provide 40kW_e for a lunar or Martian base. FSP was designed to be modest-performance and low technical risk so as to provide for economic development and minimal risk in deployment [15]. Based on this design philosophy, the FSP reactor would use uranium oxide fuel in stainless steel cladding with NaK coolant to transfer heat to several Stirling converters [15].

In each of the NTR and space power programs, the fuel and cladding were chosen based on optimal adherence to mission-specific design philosophies. While each fuel/cladding combination has relative advantages and disadvantages in each application, the development, characterization, and flight qualification of multiple fuels and claddings have proven to be prohibitively expensive. For this reason, it has been proposed to assess the viability of a single fuel form for radioisotope systems, NTRs, NEP, and surface power reactors.

Universal Encapsulation

Researchers at the Center for Space Nuclear Research (CSNR) and the INL have been investigating the viability of fabricating tungsten-encapsulated actinides by spark-plasma sintering (SPS) for space nuclear power and propulsion applications [16, 17]. Studies have indicated that SPS fabricated nuclear fuels could be useful for a wide variety of applications. Encapsulation of cerium as a surrogate for plutonium or americium, and more recently encapsulation of depleted uranium powders [18], has provided encouraging results and indicated that the required cermet densities can be achieved at lower temperatures and shorter dwell times than fabrication methods used in previous fuel development programs [19]. In the past, tungsten cermet fabrication methods had issues with uranium oxide dissociation at elevated sintering temperatures that led to migration of free uranium through tungsten grain boundaries [20]. While the UO_2 /stainless steel fuels planned for the FSP unit cannot be used in an NTR, CSNR studies indicate that a tungsten-based fuel could be used in an FSP reactor and could actually decrease the combined reactor and shielding mass [21]. It is also anticipated that the combination of dust-free sol-gel fuel fabrication with SPS tungsten encapsulation may provide a safer route to fuels for radioisotope systems.

SOL-GEL FUELS

Sol-gel methods for fabrication of nuclear fuels from aqueous solutions have been investigated for several decades, including internal and external gelation of the elements uranium, thorium, plutonium, and curium. In addition to development for terrestrial nuclear fuel, typically in the form of a tristructural-isotropic (TRISO) particle, sol-gel fuels have also been used in space applications. Pu-238 microspheres made by external gelation were used in five SNAP-27 devices for the Apollo Lunar Surface Experiment Package (ALSEP) [22, 23]. While external gelation requires the preparation of a sol that is dripped into an organic solvent to gel by the extraction of water, the internal gelation process utilizes a chilled, mixed feed solution that undergoes gelation when the temperature is raised. In addition to being a dust-free process, one benefit of the internal gelation method is that the starting solution is in the nitrate form. Uranyl nitrate can be prepared readily and Pu-238 is already in a nitrate solution after being separated from neptunium following production in a reactor. Once Pu-238 production is re-established in the United States, sol-gel microsphere production could become a part of the back end of the process, eliminating extra dissolution, precipitation, and granulation steps required in the current fabrication route.

The flow sheet for internal gelation was developed at the KEMA Laboratory in the Netherlands in the early 1970s [24]. In this process, chilled aqueous solutions of uranyl nitrate and hexamethylenetetramine (HMTA) with urea are used as a precursor to gelled spheres. Passing the mixed feed solution through a needle and into a column of hot immiscible fluid, small spheres are formed that gel upon heating as they flow down the column. Gelled spheres are still quite soft and require washing and drying steps to cure prior to sintering.

Since its initial development, significant campaigns at both Oak Ridge National Laboratory (ORNL) and Bhabha Atomic Research Centre (BARC) have refined the understanding of the chemistry involved as well as worked to optimize the feed concentrations and processing parameters. In 2004, ORNL produced about 5.5kg of internal gelation uranium dioxide spheres for use to refine the pyrolytic carbon coating processes to create TRISO particles [25]. These spheres exhibited good size uniformity and excellent sphericity in 500 μ m and 350 μ m diameter spheres.

Due to the success of the ORNL and BARC programs to produce high-quality spheres, it was decided to construct an internal gelation sol-gel rig at the University of Michigan (UM) to produce cerium dioxide and then uranium dioxide microspheres to use for subsequent fuel studies with spark-plasma sintering in tungsten. In collaboration with the CSNR and INL, spheres produced at the University of Michigan will be mixed with tungsten powder and processed into pellets for examination with a new SPS device with an attached glovebox. As of October of 2012, the sol-gel rig is nearly complete and high-quality cerium dioxide spheres have been produced.

PROCEDURE AND EQUIPMENT

The internal gelation sol-gel rig constructed at the University of Michigan is shown in Figure 1. A Julabo heater and chiller can be seen on the right-hand side of the unit and the column on the left-hand side. Feed solutions are chilled and stirred independently and then mixed in a separate, chilled vessel to prevent premature gelation. From the mixed feed vessel, the feed is transferred through chilled lines to a needle immersed in flowing silicone oil. Oil flowing past the needle strips droplets of feed and transfers them into a heated glass column where gelation occurs. Oil and gelled spheres flow through tubing until the spheres are deposited into a mesh basket and the oil is recycled in a closed loop.

In the design and construction of the sol-gel rig, safety was a factor. In addition to processing radioactive material, the system requires the use of hot oil. Stainless steel vessels and tubing were used to contain the feed solution wherever it exists in liquid form. Also, vessels and lines containing liquid uranium are designed to vent either into sealed waste containers or through charcoal and HEPA filters. Additionally, the column was designed to operate at atmospheric pressure and such that the total volume of oil in the system will naturally drain into an oversized reservoir in the case of a failed pump. High temperature tubing was installed for lines transferring hot silicone oil and the connections were secured to avoid stressing the tubes at the glass connection and to eliminate any sharp bend radii. Acrylic doors were also installed to protect the worker from a spray of hot oil in the event of tubing failure. Personal safety equipment is also used when operating.



FIGURE 1. Sol-Gel Rig at the University of Michigan

After extraction of the microspheres from the collection mesh, they are cured in hot oil for about a half hour and then allowed to reach room temperature. The cooled spheres can be washed with a combination of solutions to dissolve any remaining silicone oil as well as to cure the microspheres. During the washing steps, the conductivity of the wash effluent is monitored with a Thermo Scientific Orion Star A215 multi-parameter meter until it reaches a constant level. After washing, spheres are transferred to an alumina crucible and allowed to air dry. Once sufficiently dry, the spheres are ready for heating and sintering to drive off any remaining reaction products and reach the desired density.

RESULTS

Preliminary results in the beginning of 2012 indicated that the chemical form of cerium used in the feed stock was of considerable importance, as was the relative concentration of the feed solutions. Without proper concentrations and ratios, premature gelation in the feed vessel, stainless steel lines, and the needle was frequently a problem. Once the appropriate parameters had been established and the lines were properly chilled, problems were still encountered with gelling in the needle. Using a variety of needles, it was determined that the needle quality was a factor in needle blockages. Additional problems were overcome related to flow characteristics in the column, bubble formation, and coalescence of spheres prior to gelation that resulted in polydisperse microspheres. Through more careful control of processing parameters, excellent flow conditions and uniformity were achieved in the desired size range. Figure 2 shows a comparison of microspheres before and after calcination in air. The image was taken using a Nikon optical microscope under 10x magnification. The larger, lighter colored spheres still contain water and reaction products that evolve during calcination, resulting in a significant reduction in diameter. Due to the large changes in diameter, heating ramp rates are kept low to prevent cracking. From the photo, it is apparent that good size uniformity is being obtained and that the darker, smaller spheres are near 110 μ m diameter.

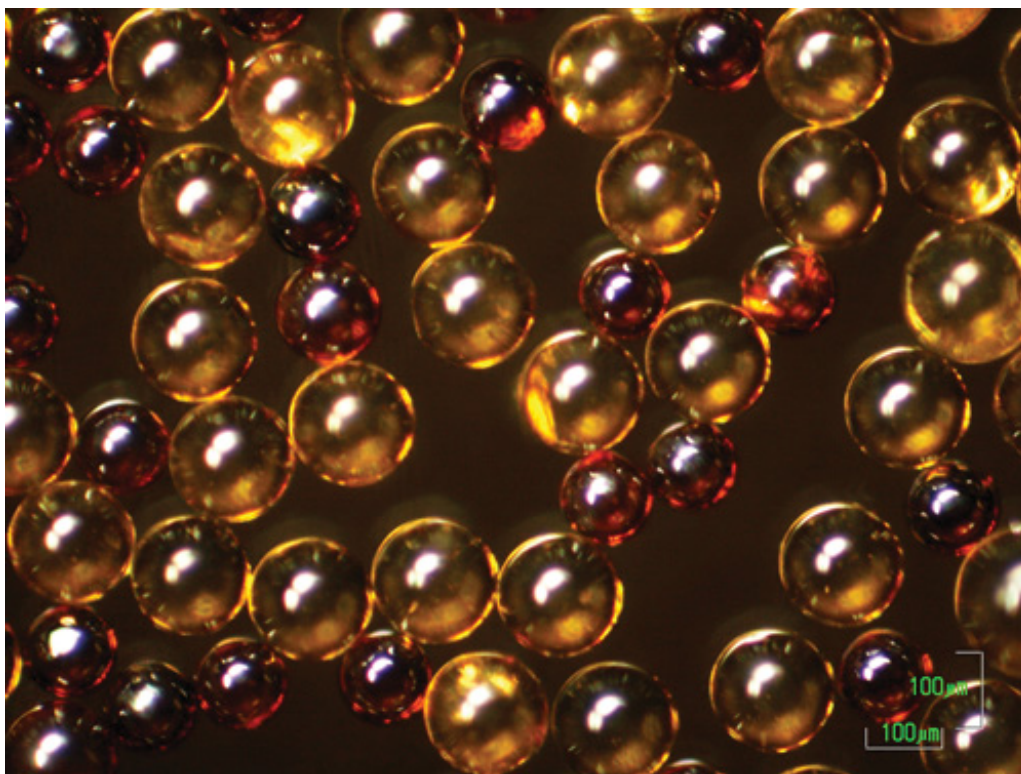


FIGURE 2. Sol-Gel Spheres Produced: Pre and Post Calcination

Work is currently underway to produce a supply of approximately 100g CeO_2 microspheres to be characterized in terms of size, sphericity, crystal structure, oxygen-to-metal ratio, and impurity content. The range of sizes achievable will also be determined. Preliminary results indicate that sintered diameters from 50-500+ microns should be achievable with minimal adjustment of the system's operating parameters. Once fabrication of the cerium spheres is complete, the rig will be used for production of depleted uranium (DU) microspheres. A kilogram of DU oxide has been acquired and procedures have been implemented in the UM laboratory to convert the stock material between U_3O_8 , UO_2 , UO_3 , stoichiometric uranyl nitrate, and acid-deficient uranyl nitrate. Based on the system throughput, production of a few hundred grams of DU microspheres should be possible and will be shipped to the CSNR for studies with tungsten encapsulation.

CONCLUSIONS

Space nuclear power and propulsion technologies are vital for continued exploration of our solar system and beyond, but require the use of unique fuels. Although the national laboratory infrastructure has significant experience processing Pu-238, fines produced during processing have proven difficult to contain and are hazardous to personnel if released. While radioisotopic systems have been an enabling technology for decades of space exploration, more advanced systems, especially those for manned missions, require the use of fission-based systems whether for power or propulsion. A number of development programs since the 1950s have advanced a variety of fuel forms, but none have been flight qualified and are ready for use today. A potential cost-saving solution is to create a universal nuclear fuel form that can be qualified and used for radioisotopic systems, NTRs, NEP, and surface reactors. It is envisioned that this universal fuel form may be realized by spark-plasma sintering encapsulation of sol-gel produced fuels into a tungsten matrix. In order to test this hypothesis, efforts have been undertaken at the University of Michigan to produce sol-gel fuels. Currently, a sol-gel rig has been constructed and the design has been iterated to the point where it can produce hundreds of grams of high-quality microspheres. Cerium dioxide spheres have been produced and will be characterized. Depleted uranium has been acquired and will be used to make hundreds of

grams of microspheres for research involving spark plasma sintering in a tungsten matrix. The resulting cermets will be analyzed to determine the quality of encapsulation.

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REFERENCES

- [1] Bennett, G. L., "Space Nuclear Power: Opening the Final Frontier," in proceedings of the 4th International Energy Conversion Engineering Conference and Exhibition (IECEC), San Diego, 2006, pp. 1433-1449.
- [2] Borland, M., Frank, S., Patton, B., Cowell, B., and Chidester, K., "An Evaluation of Alternative Production Methods for Pu-238 General purpose Heat Source Pellets," in proceedings of *Nuclear and Emerging Technologies for Space 2009*, Idaho National Laboratory, 2009.
- [3] Borland, M., et al, "Evaluation of Aqueous and Power Processing Techniques for Production of Pu-238-Fueled General Purpose Heat Sources," Idaho National Laboratory, INL/EXT-08-14017, 2008.
- [4] Congdon, J.W., "Physical Behavior of Pu-238 Oxide," SRT-MTS-96-3026, Westinghouse Savannah River Company, Aiken, South Carolina, letter to J. Herzog, Lockheed Idaho Technology Co., Idaho Falls, Idaho, 1996.
- [5] Icenhour, A. S., "Transport of Radioactive Material by Alpha Recoil," Oak Ridge National Laboratory, ORNL/TM-2005/22, 2005.
- [6] Duncan, A. J., and Kane, M. C., "Properties and Behavior of 238Pu Relevant to Decontamination of Building 235-F," Savannah River Nuclear Station, SRNL-STI-2009-00238, 2009.
- [7] Burkes, D. E., Wachs, D. M., Werner, J. E., and Howe, S. D., "An Overview of Current and Past W-UO₂ CERMET Fuel Fabrication Technology," in proceedings of Space Nuclear Conference 2007, MA, 2007.
- [8] Bhattacharyya, S. K., "An Assessment of Fuels for Nuclear Thermal Propulsion," Argonne National Laboratory, ANL/TD/TM01-22, 2001.
- [9] Haslett, R. A., "Space Nuclear Thermal Propulsion Program Final Report," Grumman Aerospace Corporation, PL-TR-95-1064, 1995.
- [10] Susnir, J., and Harman, T., "SNAP 10A Reactor Design Summary," Atomics International, NAA-SR-MEMO-8679, 1964.
- [11] Burdi, G. F., "SNAP Technology Handbook Volume II – Hydride Fuels and Claddings," Atomics International, NAA-SR-8617, 1964.
- [12] Makenas, B. J., Paxton, D. M., Vaidyanathan, S., and Hoth, C. W., "SP-100 Fuel Pin Performance: Results from Irradiation Testing," in proceedings of the 11th Symposium on Space Nuclear Power and Propulsion, NM, 1994.
- [13] Poston, D. I., Kapernick, R. J., and Guffee, R. M., "Design and Analysis of the SAFE-400 Space Fission Reactor," in proceedings of Space Technology and Applications International Forum – STAIF 2002, NM, 2002.
- [14] Ashcroft, J. and Eshelman, C., "Summary of NR Program Prometheus Efforts," Lockheed Martin, LM-05K188, 2006.
- [15] Poston, D. I., et al, "Reference Reactor Module Design for NASA's Lunar Fission Surface Power System," in proceedings of Nuclear and Emerging Technologies for Space 2009, GA, 2009.
- [16] Howe, S. D., and O'Brien R. C., "Recent Activities at the Center for Space Nuclear Research for Developing Nuclear Thermal Rockets," in proceedings of International Astronautics Congress, Prague, 2010.
- [17] O'Brien, R. C., Jerred, N. D., Howe, S. D., Samborsky, R., Brasuell, D., and Zillmer, A., "Recent Research Activities at the Center for Space Nuclear Research in Support of the Development of Nuclear Thermal Rocket Propulsion," in proceedings of Nuclear and Emerging Technologies for Space 2012, TX, 2012.
- [18] O'Brien, R. C., Jerred, N. D., "Spark Plasma Sintering of W-UO₂ Cermets," *Journal of Nuclear Materials*, Vol. 433 pp. 50-54, 2013.
- [19] O'Brien, R. C., Ambrosi, R. M., Howe, S. D., Bannister, N. P., and Atkinson, H. V., "Fabrication of Prismatic Fuel Elements for space Power and Nuclear Thermal Propulsion Reactors by Spark Plasma Sintering," in proceedings of Nuclear and Emerging Technologies for Space 2009, GA, 2009.
- [20] General Electric, "710 High-Temperature Gas Reactor Program Summary Report. Volume III. Fuel Element Development," General Electric, GEMP-600 (Vol. 3), 1968.
- [21] Craft, A. E., Silver, I., and Clark, C., "Radiation Shielding for Fission Surface Power Systems," Center for Space Nuclear Research, 2011.

- [22] D. L. Prosser, "SNAP 27 Radioisotopic Heat Source Summary Report," MLM-1698, 1969.
- [23] Plymale, D. L., and Smith, W. H., "The Preparation of Plutonium-238 Dioxide Microspheres by the Sol-Gel Process," Mound Laboratory, MLM-1450, 1967.
- [24] Bruggen, F. W. v. d., Noothout, A. J., Hermans, M. E. A., Kanij, J. B. W., and Votocek, O., "A U(VI) Process for Microsphere Production," in proceedings of the Symposium on Sol-Gel Processes and Reactor Fuel Cycles, TN, 1970.
- [25] Collins, J. L., Hunt, R. D., Del Cul, G. D., and Williams, D. F., "Production of Depleted UO₂ Kernels for the Advanced Gas-Cooled Reactor Program for Use in TRISO Coating Development," Oak Ridge National Laboratory, ORNL/TM-2004/123, 2004.