FABRICATION OF SOL GEL MICROSPHERES FOR SPACE NUCLEAR POWER J. A. Katalenich,[1] M. R. Hartman[1], R. C. O'Brien[2], and S. D. Howe[2], [1]University of Michigan Department of Nuclear Engineering and Radiological Sciences, 2355 Bonisteel Blvd., Ann Arbor, Michigan 48109, contact: jkatalen@umich.edu, [2]Center for Space Nuclear Research, 995 University Blvd., Idaho Falls, Idaho 83401

Introduction: The use of nuclear power for space applications provides unique system capabilities and enables missions that would otherwise not be feasible. In situations where conventional batteries cannot operate for the necessary duration and solar cells cannot provide sufficient power, nuclear devices find their niche. Radioisotope thermoelectric generators (RTGs) powered with plutonium-238 (Pu-238) have proven to be reliable sources of tens to hundreds of watts for times spanning decades. While nuclear thermal rockets (NTRs) and extra-planetary nuclear reactors have less heritage than RTGs, they hold promise for a future generation of more ambitious manned space exploration. These grand visions for a space-faring human race have faded, however, in recent years with the reluctance to deploy space fission systems and a declining national inventory of Pu-238.

With limited budgets for planetary exploration, there is a need to get more science per dollar spent on any given mission. In the early days of RTG development, RTGs were re-designed and re-qualified to be tailored for different missions. Indeed, in a relatively short period of time, there were many iterations of RTG designs deployed on spacecraft. Realizing the impact of RTG re-designs on cost, the goal became to create a more generic RTG design that could be used for multiple missions [1]. In fact, RTGs being deployed today are even named "multi-mission" RTGs. In keeping with these trends and ideals, a current proposal by the Center for Space Nuclear Research (CSNR) is to develop a universal nuclear fuel form that is applicable to RTGs as well as NTRs and space reactors. The method proposed by the CSNR involves a "universal" encapsulation of nuclear materials in a tungsten matrix. While the CSNR is advancing the technology for tungsten encapsulation methods [2, 3], work has begun at the University of Michigan, in collaboration with the CSNR, to develop a "universal" fabrication method for the nuclear feed particles that are to be encapsulated.

The Neutron Science Laboratory (NSL) at the University of Michigan (UM) has been pursuing the use of internal gelation sol-gel methods for the fabrication of uranium dioxide and plutonium dioxide microspheres. It is anticipated that fabrication of UO_2 and PuO_2 microspheres and subsequent encapsulation will be both simpler than the current methods utilized and result in a final product that is equally safe and more applicable to a variety of uses in space nuclear power applications.

Current RTG systems utilize pure plutonium dioxide (PPO) pellets, clad in iridium and surrounded in a layered graphite matrix to ensure that in the case of atmospheric re-entry the pellets themselves will not ablate and remain intact upon surface impact. Processes at the Los Alamos National Laboratory to fabricate these PPO pellets involve the ball milling of plutonium dioxide powders to sub-micron sizes [4]. Additionally, some stages of pellet production require radiation workers to perform glovebox operations [4]. These fine plutonium powders exhibit mobility, contaminating gloveboxes and posing a potential radiological hazard [5]. Therefore, the primary aim of sol-gel process development is to demonstrate automated microsphere production without the generation of fine powders.

Production of nuclear fuel particles by the sol-gel method is not a new concept. The external gelation sol-gel technique was used to make plutonium microspheres for the SNAP-27 RTGs used in the Apollo Program [6] and there has been ongoing research into internal gelation techniques at Oak Ridge National Laboratory (ORNL) and the Bhabha Atomic Research Centre (BARC) [7, 8]. Existing research has been largely aimed at the production of various combinations of thorium, uranium, and plutonium microspheres that can be subsequently pressed into fuel pellets for terrestrial nuclear power reactors. Research being conducted by the NSL and CSNR is aimed at producing microspheres suitable for encapsulation in a tungsten matrix capable of managing the buildup of helium from decades of Pu-238 decay.

Methodology: A general internal gelation sol-gel process begins with the combining of chilled solutions of first uranyl nitrate and then hexamethylenetetramine (HMTA) and urea. It is believed that urea in the chilled solution complexes the uranium [9]. Upon dripping the feed uranium solution into a column of hot, immiscible silicone oil, the microspheres formed begin to heat. Once the microspheres reach a critical temperature, the urea decomplexes the uranium, allowing the HMTA to undergo protonation and the result is gelled microspheres [9]. Upon extraction, the microspheres are typically washed and then prepared for heat treatments to drive off impurities and reduction into a dioxide form.

Experiments at the Neutron Science Lab and the Center for Space Nuclear Research have used cerium as a non-radioactive surrogate for uranium and plutonium. It is anticipated that once preliminary investigations have provided sufficient insight into the process and testing of the apparatus, studies will continue using depleted uranium.

Results: Preliminary experiments have been performed to assess the gelation parameters for cerium. Like the internal gelation process for uranium, cerous nitrate solutions mixed with optimal proportions of HMTA and urea resulted in the creation of a hard wax when heated as a bulk mixture in a beaker, and formation of soft, gelled spheres after falling through a column of hot silicone oil. A photograph of gelled cerium microspheres at the bottom of the gelation column is shown in Figure 1. These microspheres appeared to have good size and shape characteristics.

Figure 1: Soft, gelled cerium spheres fabricated by the internal gelation sol-gel process at the NSL

Work is continuing on the construction of a sol-gel testing rig for automated production of microspheres. The apparatus consists of a jacketed column, oil heater and circulator to heat the oil in the column, and a chiller to cool the feed solutions. Additionally, an assembly for producing uniform spheres is being developed. The testing rig is shown in Figure 2.

Conclusions: Initial investigations have shown that the internal gelation sol-gel method previously demonstrated with uranium is also capable of producing gelled microspheres of cerium. These microspheres appear to have a near-uniform size distribution. It is anticipated that spheres with diameters in the ranges of interest can be fabricated and heat treated to yield cerium dioxide microspheres for encapsulation in tungsten as a surrogate for uranium dioxide and plutonium dioxide. After characterization of the cerium spheres is completed, future work will include the fabrication of depleted uranium microspheres and encapsulation of microspheres in tungsten.

Figure 2: Sol-gel testing rig at the NSL

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