MICROSPHERE PLUTONIUM-238 OXIDE FUEL TO REVOLUTIONIZE NEW RADIOISOTOPE POWER SYSTEMS AND HEAT SOURCES FOR PLANETARY EXPLORATION

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ABSTRACT

We propose research and development activities supporting the advancement of improved radioisotope heat sources to support future NASA exploration missions such as those to Ocean Worlds. Missions to penetrate ice and explore oceans will require not only high specific power, but also the incorporation of radioisotope heat sources inside of the vehicle itself to use it for heat as well as power. To enable flexible geometries and high volumetric power loadings, microsphere-based heat sources are attractive. Sol-gel methods to produce radioisotope microspheres have the added benefit of being a much cleaner fabrication process, reducing hazards during heat source fabrication. Although preliminary efforts have demonstrated that microsphere heat sources can be produced, uncertainty remains around the integrity of coatings, which would significantly enhance the performance and safety of microsphere fuels, as the fuel ages. Continued research and development within the U.S. Department of Energy would provide the information needed to assess the feasibility of advanced, microsphere-based heat sources and to refine the design of future radioisotope power systems (RPS) for exploration missions.

INTRODUCTION

Microspheres of plutonium-238 (Pu-238) oxide, shown in FIGURE 1, were recently produced at the Pacific Northwest National Laboratory (PNNL) using the sol-gel process (Katalenich, 2017). Sol-gel methods allow for aqueous solutions containing Pu-238 to be converted directly into microspheres of the desired size without generating dust in the process. Conceptually, these microspheres could be pressed into conventional GPHS pellets, encapsulated as packed spheres, or individually coated to enhance thermal conductivity and reduce contamination risks during accidents.

FIGURE 1: $^{238}\text{PuO}_2$ sol-gel microspheres produced by the Pacific Northwest National Laboratory in 2017.

PNNL's successful production of these microspheres offers significant benefits in the near and distant future. In the near-term, such microspheres, which are both non-inhalable and non-respirable, can be used as a feed material to press plutonium-238 oxide fuel pellets for subsequent cladding at Los Alamos National Laboratory (LANL). Sol-gel processing produces virtually no dust contamination, which would reduce deterioration of LANL glove boxes, equipment, and the worker exposure hazard. In the longer-term, coated Pu-238 oxide microspheres would enable the

production of revolutionary radioisotope heat sources for RPSs with improved design integration, performance, flexibility, and safety (Sholtis et al., 1999). Compared to granules produced by powder processing, microspheres are smooth and round, promoting coatings with a thin compatibility layer(s) (e.g., porous and/or pyrolytic graphite) and a strong, high-temperature containment barrier (e.g., ZrC or WC). Although microsphere coating technology is demonstrated for TRISO uranium reactor fuels, the lifetime and performance of coatings on Pu-238 oxide microspheres remains to be determined.

Current U.S. radioisotope heat sources include the 250 Wt General-Purpose Heat Source module (GPHS), and the 1 Wt Light-Weight Radioisotope Heat Source (LWRHU). LANL maintains our nation's capability for processing Pu-238 into clad heat sources. Worker dose and fine particulate contamination increase the cost and risk for Pu-238 processing at the LANL PF-4 facility. The current Pu-238 processing flowsheet requires steps, such as ball milling and slugging, that generate small, dispersible particles that increase radiological backgrounds and equipment failure rates. Steps in the traditional flowsheet also require hands-on activities that, together with accumulated buildup, result in worker dose rates requiring workers to be rotated on assignments to prevent exceeding dose limits. Pu-238 oxide microspheres, produced by sol-gel methods, would eliminate the need for ball milling and its associated Pu-238 oxide dust generation. This will improve cleanliness and limit deterioration of glove boxes; in turn, reducing Pu-238 oxide recovery requirements and wastage, while increasing glove box lifetime and reducing hazards for glove box workers.

Pu-238 oxide pellets are clad with a ductile, refractory alloy (DOP-26 iridium (Ir) alloy for the GPHS, and platinum 30% rhodium (Pt-30Rh) for the LWRHU). These cladding materials are only ductile within a temperature range. Insulation is used to ensure clads don't get too hot or too cold during accident scenarios and remain ductile upon impact to prevent Pu-238 release. Coated Pu-238 oxide fuel particles would eliminate both the need for insulators within any of a myriad of heat sources that could be envisioned. Such new heat sources would require a reentry aeroshell to protect against fuel release and vaporization for an inadvertent reentry accident but could be a collection of coated Pu-238 oxide fuel particles within virtually any designed shape or size needed. This could enable specially designed compact heat sources for size-limited RPSs (e.g., cryobots; hydrobots; small communication and data transmission RPSs for icy moons with liquid ocean worlds beneath; shaped heater heads for Stirling Radioisotope Generators (SRGs); etc.).

SOL-GEL MICROSPHERES FOR NEXT-GENERATION RPS

Future exploration missions are likely to require Pu-238 heat and power sources that differ from what is currently available. For example, missions to penetrate thick layers of ice and to explore ocean worlds beneath them require vehicles with constrained size and aspect ratios. Such missions require heat and power sources to be integral to the vehicle, contrary to most probes to date which have attached radioisotope power systems (RPS) external to the vehicle. Compact heat sources designed to fit inside the vehicle, possibly with flexible geometries to facilitate use in different-sized craft, could be advantageous or even enabling given size, mass, and mission duration requirements.

Since microspheres can fill a void of any shape, sol-gel may enable more flexible RPS and RHU geometries. As an example, consider a Europa cryobot mission. Aside from producing heat sources meeting the unique length to diameter ratios required for such missions, microspheres could enable tailored heat source geometries to fit inside drill bits and shafts to allow melting whether or not the drill is functioning. This design flexibility could enhance the large RTGs inside the cryobot main

body and/or the smaller RTGs being deployed behind the cryobot to enable the communication link back to the surface. Additionally, microspheres could be loaded onto trays of horizontal K-1100 type graphite fibers to maximize radial heat flow, a major consideration for cryobots, and minimize centerline temperatures in the fuel that typically limit the dimensions of fueled regions.

We propose to initiate an R&D program to develop and evaluate the use of sol-gel methods to produce coated microspheres of Pu-238 oxide to address operations challenges at LANL and to create new RPS architectures for NASA exploration missions. Although the production of Pu-238 oxide microspheres has been demonstrated, additional development is needed to evaluate microsphere stability over time, determine the integrity of coatings over time, and to demonstrate the production of full-scale batches. Such studies can be performed as part of an ongoing RPS R&D program and the results used to determine the feasibility of envisioned RPSs. If results are promising, additional efforts would be needed to perform qualification testing of microsphere-based heat sources.

EXAMPLE USE CASES – CRYOBOT PROBE

The exploration of ocean worlds such as Enceladus and Europa is an ambitious goal that may involve a phased approach. Analogous to the approach to explore Mars, which used increasingly large and sophisticated exploration craft over time, missions to ocean worlds will likely grow in complexity and scope as we learn more about these planetary bodies and how to operate there. JPL is currently investigating the feasibility of an ice penetrator mission, called PRIME (Probe using Radioisotopes for Icy Moons Exploration). PRIME uses heat from the decay of Pu-238 to melt through ice, power onboard systems, and to power communication relays that are left in the ice as the vehicle descends.

The power density of a radioisotope heat source is very important for a cryobot because the physical size of the heat source strongly influences the size of the cryobot, which in turn drives the thermal inventory required. Due to this circular relationship, bulkier heat sources drive up power requirements and increase the amount of Pu-238 required. Minimizing the size and mass of the probe is also necessary because long probes would be more difficult to land on Europa and the mass we can deliver to the surface is limited. Increasing the power of the heat source also reduces risk by decreasing the mission duration, which is largely dictated by the transit time to Europa and the time required to melt through ice.

To support PRIME, JPL and PNNL generated conceptual designs for a high-power density radioisotope heat source to minimize probe size and the time required for ice penetration (Hockman et al., 2020; Katalenich et al., 2019). Initially, a variety of GPHS configurations within the cryobot were considered, as shown in FIGURE 2. However, as shown in blue in FIGURE 3, the cryobot mass and melt speed based on a stack of GPHS modules are undesirable compared to alternative configurations. Alternative heat source configurations, such as placing pellet-fueled graphite impact shell (GIS) assemblies into a common aeroshell or a microsphere-based heat source, depicted in FIGURE 4, provided geometries tailored to the cryobot optimal diameter and enabled more desirable melt speeds and system masses.

FIGURE 2: Various configurations of GPHS module stacks and the corresponding cryobot diameter required (Hockman et al., 2020).

FIGURE 3: Comparison of many heat source configurations and their resulting melt speed and system mass indicate that cryobot configurations based on the GPHS module (blue) are slower and heavier than non-GPHS configurations such as those that place GIS modules into a common aeroshell (green) or microsphere-based heat sources (red) (Hockman et al., 2020).

Based on these trades and constraints, JPL has expressed interest in investigating the development of radioisotope heat source configurations that have higher power densities than a stack of GPHS modules. Options that appear attractive include 1) a common, cylindrical aeroshell that accepts 6-7 graphite impact shells containing Pu-238 pellets, as depicted in FIGURE 4, or 2) a similar aeroshell configuration using Pu-238 microspheres to increase the power density.

FIGURE 4: Radioisotope heat source configurations inside a cryobot probe cross-section show the limitations of the General-Purpose Heat Source (A & B) compared to a common aeroshell containing GIS or microsphere-loaded canisters (C) (Hockman et al., 2020).

The microsphere-based heat source design results in a cryobot with the lowest system masses, smallest size, and fastest time to penetrate the ice, as shown in red in FIGURE 3. Although using a common aeroshell containing Pu-238 pellets in a GIS is likely more expedient than developing and qualifying a microsphere-based design for the first cryobot mission, JPL envisions a future with more missions that require the type of power-dense heat source enabled by a microsphere fuel. Additionally, microspheres facilitate the development of future heat sources with the kinds of flexible, modular geometries we anticipate needing to explore icy moons and planets where the heat source will need to be incorporated inside the vehicle.

COATED MICROSPHERE HEAT SOURCES – REQUIRED R&D AND QUALIFICATION

Despite the potential benefits of sol-gel processing and microsphere heat sources, changing heritage processes and systems poses technical and safety risks that must be addressed. In addition to converting portions of the PF-4 processing line to accommodate sol-gel and coating processes, some degree of qualification testing will be required to validate the safety performance of products made using a different approach. The level of investment required merits a detailed assessment of the extent of improvements expected using the sol-gel method as well as an evaluation of whether sol-gel processing and coating operations could be successfully deployed at PF-4.

While sol-gel technology is mature for applications such as producing large quantities of coated uranium oxide microspheres, used in tristructural isotropic (TRISO) fuel, the use of sol-gel to make Pu-238 oxide microspheres has only been demonstrated at the gram scale. Preliminary investigations used cerium oxide as a surrogate to perform detailed studies to develop necessary process modifications to the uranium flowsheet. Based on results with ceria, a flowsheet was developed and successfully demonstrated with gram quantities of Np-237, Pu-239, and Pu-238. Although work to date has answered many questions regarding the feasibility of using sol-gel methods to produce microspheres of $^{238}\text{PuO}_2$, additional efforts are needed to answer remaining questions regarding microsphere and coating stability and to evaluate the impact of using sol-gel at PF-4.

Development activities are needed to answer remaining questions regarding the use of sol-gel at Los Alamos as well as the use of microsphere-based heat sources for planetary science missions. For example, pellet pressing tests are needed to confirm that sol-gel microspheres can produce an

acceptable hot pressed GPHS pellet. A sol-gel flowsheet and hardware design is also needed to estimate worker dose during processing activities. For future NASA systems, an evaluation of whether $^{238}\text{PuO}_2$ microspheres can be coated with materials of interest and the stability of those coatings, would help answer questions regarding effective thermal conductivity and fuel containment robustness against launch failure scenarios.

We propose a prioritized and gated approach to sol-gel evaluation and development, led by LANL and PNNL. The first phase of proposed activities includes communication between LANL, PNNL, and JPL to share existing sol-gel knowledge, identify challenges and benefits for using sol-gel in PF-4, identify challenges and benefits for using microsphere-based fuels, and to prioritize the development activities needed to answer key questions. Findings from phase one, including estimates of the cost and schedule required, may be used as a basis for whether to proceed. The second phase includes development activities, with experiments prioritized by the importance of the questions answered. Reviews may be included throughout phase two in order to evaluate results and determine whether to proceed. The third phase includes definition and execution of qualification testing experiments. We envision that conversion of a processing line at PF-4 would not take place until after successful development and qualification testing.

Using the proposed approach, uncertainties and risks would be reduced and development could proceed at a pace commensurate with the likelihood of successfully implementing sol-gel at PF-4 as well as NASA's schedule requirements. Obtaining priority information in the near-term would help inform ongoing NASA planning activities and feasibility studies.

SUSTAINING SOL-GEL RESEARCH TO ASSESS MICROSPHERE FUEL FEASIBILITY

Considering the perceived benefit of a new microsphere-based heat source and the long lead time required to develop and qualify it, an effort should commence at the earliest opportunity to develop a detailed roadmap, schedule, and budget as well as perform priority experimental tasks to answer key questions and reduce risks. Such information is needed now to improve JPL's insights into the feasibility and cost of a microsphere-based heat source as well as when it could be available.

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