

# Advanced Radioisotope Heat Source and Propulsion Systems for Planetary Exploration

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## ADVANCED RADIOISOTOPE HEAT SOURCE AND PROPULSION SYSTEMS FOR PLANETARY EXPLORATION

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The exploration of planetary surfaces and atmospheres may be enhanced by increasing the range and mobility of a science platform. Fundamentally, power production and availability of resources are limiting factors that must be considered for all science and exploration missions. A novel power and propulsion system is considered and discussed with reference to a long-range Mars surface exploration mission with in-situ resource utilization. Significance to applications such as sample return missions is also considered. Key material selections for radioisotope encapsulation techniques are presented.

### I. TUNGSTEN CERMET HEAT SOURCES

#### I.I BACKGROUND

The encapsulation of radioisotope materials such as  $^{238}\text{PuO}_2$  and  $^{241}\text{AmO}_2$  within tungsten cermets offers many advantages for the production of radioisotope heat sources from both a radiological and thermo-mechanical standpoint<sup>1</sup>. It has previously been estimated that a significant reduction in the  $\alpha$ -n neutron yield from  $\alpha$  emitting radionuclide compounds can be achieved through the production intimate mixing of such compounds with heavy inert materials such as tungsten. For  $^{241}\text{AmO}_2$  it has been estimated that a 65.9% reduction in the  $\alpha$ -n derived neutron yield for a source of given activity can be achieved by mixing its compound with tungsten (60% by volume)<sup>1</sup>. The self-shielding properties for x-ray and gamma-ray emission of tungsten encapsulated sources is also most advantageous<sup>1</sup>. As an example, the gamma-ray emission for an  $^{241}\text{Am}$  source (primarily 60 keV) can be attenuated to 1% of its initial intensity by a 0.8mm thick tungsten jacket. For comparison, such a source would require a 15.52 cm thick aluminum encapsulating jacket to achieve the same degree of attenuation. The effects of source encapsulation within tungsten has been evaluated by Monte Carlo modeling<sup>1</sup>.

For heat source interfaces, the thermal conductivity between the source material and the application is highly important. For tungsten this is

174  $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . When considering operational temperatures and temperatures achieved under non-nominal conditions such as launch failure and de-orbit, it is also important to select an encapsulating material for the radionuclide compounds that will preserve capture of the enveloped nuclear materials under all foreseen eventualities. Tungsten offers strength and ductility at high temperatures, and its melting point at 3422 °C may be sufficient to prevent the release of encapsulated materials at high temperatures<sup>1</sup>. The use of Tungsten also presents resistance to cutting and ensures that the radioisotope materials cannot be successfully extracted by unauthorized parties via pyrolytic processes.

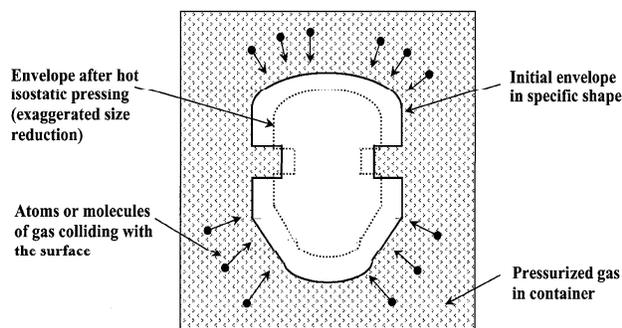
Significantly, through the use of tungsten cermet technology, a high power heat source may be produced with a specific volume ( $\text{kW}/\text{cm}^3$ ) that is up to 4 times greater than traditional heat sources that require a separate aero-impact shell.

#### I.II TRADITIONAL SINTERING TECHNIQUES

Traditional powder metallurgical sintering techniques for the fabrication of encapsulating media can be divided into two categories; with applied pressure (hot pressing) and without pressure (pressureless sintering). Pressureless sintering is achieved at temperatures close to the melting temperature ( $T_m$ ) of the processed powdered metals and

results in a product that has minimized density variations throughout its structure. The resultant density of products sintered via pressureless sintering is determined by a combination of sintering temperature and processing time, which is typically of the order of several hours<sup>2</sup>. Hot pressing, otherwise known as sinter-forging, is a technique in which the powdered media to be processed is loaded into a die, which in turn is loaded into a heating chamber and pressure is applied to the die via a unidirectional ram<sup>3</sup>. Sinter-forging typically results in products with a higher density than those produced via pressureless sintering<sup>2</sup> but this densification is typically non-uniform throughout the sintered matrix due to friction between the walls of the die and the processed materials<sup>3</sup>. The processing temperature, dependant upon the material being processed, is typically reduced, although this is still close to  $T_m$  through forge sintering and the respective processing time is reduced by the order of 10 times over pressureless sintering<sup>2</sup>.

Hot Isostatic pressing (HIP'ping) is a sintering technique that eliminates the need for rams, dies and external frictional forces. Here, the powdered materials to be processed are loaded into thin walled cans/envelopes that are shaped to the desired finished product and are made from materials that deform plastically at the processing conditions (temperature and pressure)<sup>3</sup>. During densification of the powdered materials, the volume occupied contracts. Since the envelopes/cans are under isostatic pressure during the sintering process, its volume contracts synchronously with the contained material (see Figure 1). Given that a photographic reduction in volume and shape can be achieved through HIP'ping, uniform densification and void removal can easily be achieved while producing net complex sintered product geometries. The typical processing temperatures for HIP'ping are material specific and are usually in excess of  $0.7 T_m$ <sup>3</sup>.



**Figure 1:** Illustration of the photographic reduction in shape of an encapsulating envelope and the processed powdered materials during Hot Isostatic Pressing (HIP'ping)<sup>3</sup>.

### L.III SPARK PLASMA SINTERING

#### L.III.I Principal & applicability

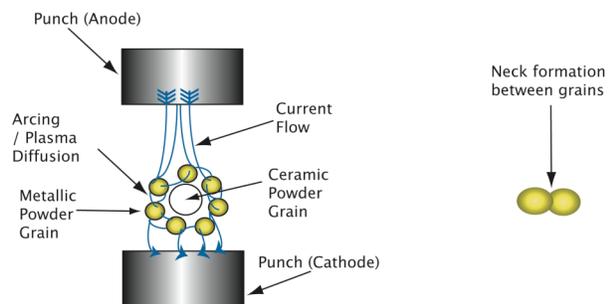
It has previously been proposed that plutonium dioxide or americium dioxide could be encapsulated within a tungsten-based cermet using the Spark Plasma Sintering (SPS) technique<sup>1</sup>. The fundamental rationale for the trial of SPS processing for these materials was that traditional sintering techniques, such as Hot Pressing, required the materials to be processed at high temperatures that would exceed the dissociation temperatures for the radioisotope oxides. The result would then be the formation of complete or partial non-cermet (metallic) regions such as those indicated within the fuel element development summary in the General Electric report on the development of the 710 High-Temperature Gas Reactor<sup>4</sup>. Given that Spark Plasma Sintering is capable of processing materials at greatly reduced temperatures<sup>5</sup> while minimizing the grain growth typically attributed to traditional sintering techniques<sup>3</sup>, SPS is highly suited to the production of cermets with temperature sensitive ceramics such as  $PuO_2$  and  $AmO_2$ .

In the SPS process, a powder mixture is heated by Joule heating which is a result of the passing of electric current through the powder matrix that is to be consolidated. This is effectively the same as conventional resistive heating with the exception that the temperature is varied during the process by the modulation of current pulses. There are therefore two distinct operating temperatures for an SPS furnace; the average temperature, and a much higher temperature that is only reached during the flow of the current pulses. The average temperature is tuned to be lower than the melting point of the materials in the matrix. This therefore reduces or prevents the dissociation of the ceramic materials that are to be encapsulated within the cermet.

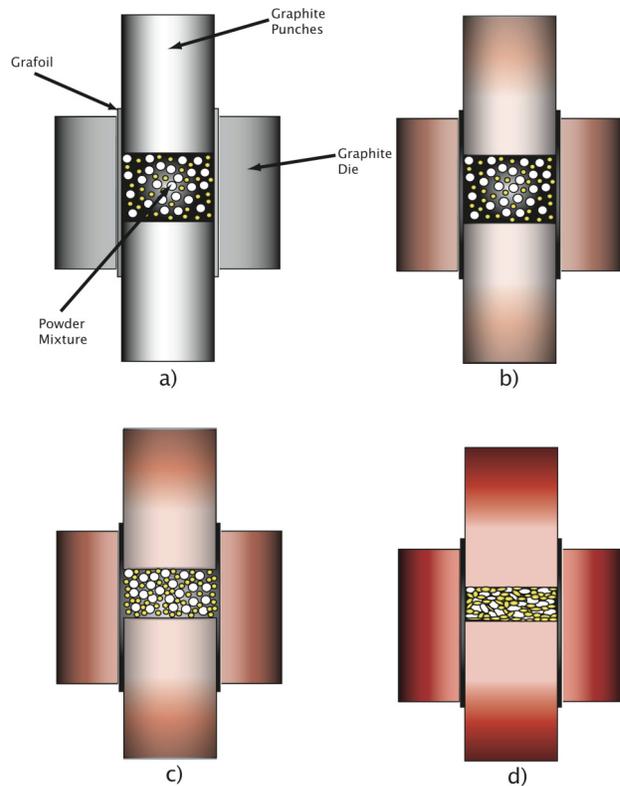
During each current discharge, metallic material is transported as plasma by the spark propagation across the pores of the matrix (see Figure 2 – Left). When the current is switched off, the matrix undergoes rapid cooling resulting in the condensation of the metallic vapors within the regions where there is mechanical contact between powder grains<sup>6</sup>. This condensation of vapor produces necks that enhance these joints (See Figure 2 – Right). The material transport in subsequent spark pulses is accentuated due to the greater electric current density in the necks and contacts than inside the body of each powder grain. The rate of material transport is enhanced through the application of an external compressive force<sup>7</sup> resulting in the plastic deformation of the powder grains at each interface<sup>3</sup> resulting in a flatter joint with lower electrical

resistance. This process propagates throughout the matrix, enhanced by the external pressure which induces plastic flow of the material to form a sintered cermet with a density that is very close the full theoretical density. Densification throughout the resultant matrix is uniform due to the SPS effect, applied axial force and the radial electromagnetic pinch force caused by the passage of high current. This process propagation is similar to that exhibited in the consolidation of matrices by hot isostatic pressing<sup>3</sup>.

The overall SPS setup consists of a conductive (graphite) die into which the powder mixture is formed, a press and a high power pulsed DC circuit. The filled cavity in the die is capped on the top and bottom with graphite punches that are free to move in the axial direction, facilitated by the use of grafoil to lubricate the side walls of the die. The die, punches and, ultimately, the powder mixture itself, form the completing component for the DC circuit. The geometry of the die determines the radial geometry of the pressed/sintered cermet. For the formation of cermets, ceramic powder (radioisotope compound) and metal powders are mixed to the specified volumetric ratio prior to loading into the graphite die. The die is then put under compression in between two electrodes in a hydraulic press<sup>8</sup> in order to compact the mixture. At this point a current is passed with a discharge pulse of the order of several kA, and its duration is of the order of several microseconds<sup>6</sup>.



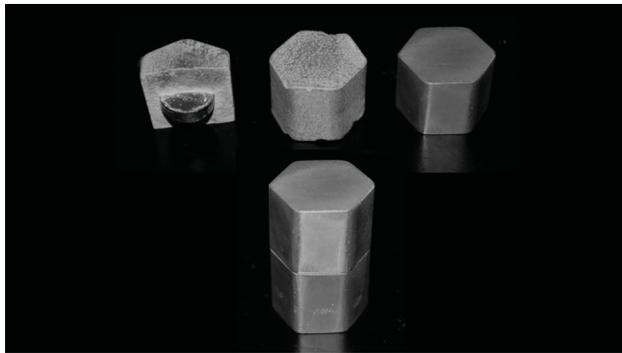
**Figure 2:** The principal of Spark Plasma Sintering (SPS). **(Left)** Illustration of the effect of the application of a current pulse to the powdered materials between the two electrode punches. Current discharge causes arcing between conductive grains resulting in rapid localized heating which in turn results in the formation and diffusion of plasma between the conductive grains. **(Right)** As the current pulse is terminated, the diffused plasma cools resulting in the formation of necks between the material grains.



**Figure 3:** **a)** Punches and die assembly lined with grafoil containing mixed  $\text{CeO}_2$  and Tungsten powder. **b)** The assembled die upon loading into SPS furnace (see figure 2.1). Application of minimal pressure and initiation of heating results in small expansion of the materials. **c)** Application of pressure to the punches via the hydraulic rams of the SPS furnace facilitates initial densification by closing of voids. **d)** Neck formation and continued application of pressure achieves final densification of the bulk materials resulting in cermet production.

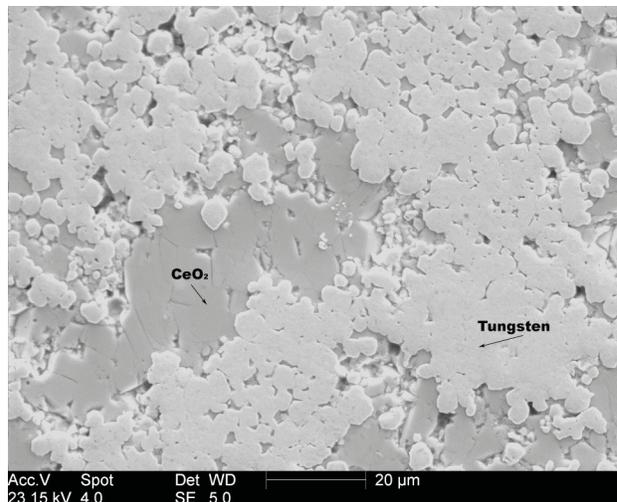
#### 1.III.II. Experimental Demonstration

Practical research conducted by O'Brien et al at the Center for Space Nuclear Research has demonstrated the capability of SPS to encapsulate simulated radioisotope compounds within tungsten cermets. Other non-tungsten based cermet fabrication has also been successfully demonstrated. Some tungsten cermets produced within this research were completely encapsulated in a tungsten jacket via a two-step SPS process. Examples are given in Figure 4 Verification of the encapsulation was performed by x-ray diffraction and SEM imaging with electron induced x-ray fluorescence capabilities. Figure 5 is an SEM image of a tungsten cermet loaded with  $\text{CeO}_2$  as an analogue for  $\text{PuO}_2$  or  $\text{AmO}_2$ .  $\text{CeO}_2$  was selected as the analogue for these compounds due to their similar kinetic properties, melting temperatures and Gibbs free energies.



**Figure 4:** Examples of variable porosity fully encapsulated inert heat source prototypes produced via a 2 step SPS process.

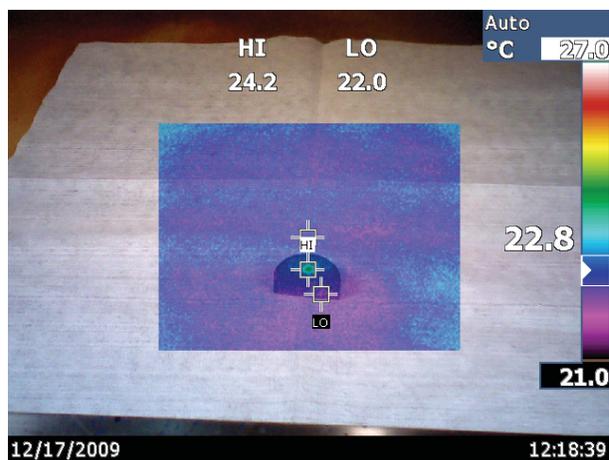
The resultant product cermet densities were found to be in excess of 94 % theoretical density for processing temperatures of  $0.44 T_m$  for tungsten, and porosity was tunable by adjustment of processing time in conjunction with the tungsten volumetric ratio and particle size used, allowing for the compensation for radiogenic gas production to be built into cermet design. Figure 5 clearly shows the polycrystalline nature of the  $CeO_2$  particles. It would appear that some cracking of the individual  $CeO_2$  particles occurred during the application of high pressure to the mixture within the die. The voids created by this action were mostly filled by the tungsten particles, although a small volumetric fraction remained unfilled, accounting for some of the deficit in full densification of the cermets. Further enhancement of densification could be achieved by the minimization of shearing and cracking of the ceramic materials during the application of pressure. Future investigations will examine the production of, and the advantages of using spherical ceramic particles over granular ceramics. It is anticipated that spherical particles will be capable of resisting shear from the surrounding tungsten particles.



**Figure 5:** Scanning Electron Microscope (SEM) image of a tungsten-ceria cermet cross-section. The cermet under examination was composed of 40%  $CeO_2$  and 60% W by volume. This volumetric ratio is evident in the product cermet from the inference of proportional areas of each material.

#### I.III.III. Chemical Compatibility study

The use of a surrogate compound for initial SPS encapsulation trials was prescribed by the absence of a surrounding glovebox during the early setup of the SPS furnace. Clearly, a strong argument may be presented for the demonstration of the SPS process with  $PuO_2$  or  $AmO_2$  prior to significant elevation of its technology readiness level due to subtle differences in the chemical behaviors of each individual compound.

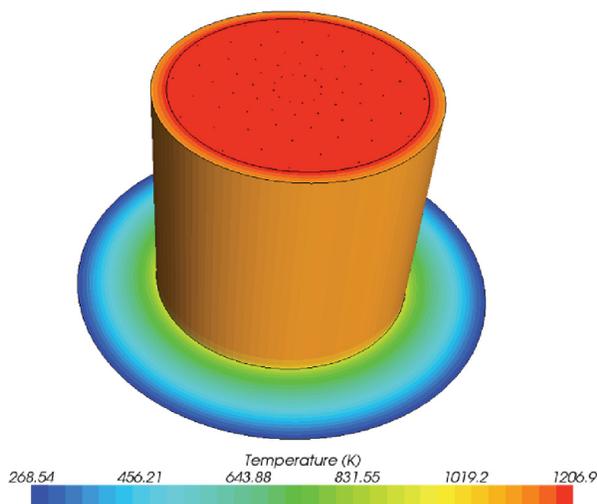


**Figure 6:** Composite optical and thermal image of americium oxide-tungsten cermet heat source indicating heat production at a rate of 5.9 mW and a specific thermal power of  $^{241}Am$  oxide of 0.093 W/g. Data verified via Differential Scanning Calorimetry.

In order to expedite the determination of the feasibility of the encapsulation process, a series of experimental chemical compatibility tests were performed by the author at Idaho National Laboratory using  $^{241}\text{AmO}_2$  and tungsten powders of identical morphology to those within the surrogate system. Here, green pressed tungsten cermet pellets were examined within a Differential Scanning Calorimeter (DSC) under a temperature profile identical to that of the SPS furnace. DSC data sets were examined for evidence of chemical alteration of the compounds and compared to both X-Ray Diffraction measurements and SEM data. The results from the compatibility study will be presented within a future paper.

## II. THERMAL ENERGY STORAGE SYSTEMS

Research performed at the Center for Space Nuclear Research examined an alternative approach to the utilization of nuclear decay heat. As an alternative to the continuous coupling of a radioisotope heat source to an energy sink or power conversion system, an intermediate thermal energy storage mechanism can be employed that can be thermally isolated from external systems at the end of periodic thermal discharge cycles. In doing so, energy is accumulated from an integral compact cermet based radioisotope heat source in between thermal discharges; hence the temperature of the heat source is recovered. The use of a thermally capacitive heat source allows for a smaller radioisotope inventory to be used while achieving higher initial heat source temperatures. Due to the nature of the thermal cycling process, a period of time is required for the temperature of the heat source to recover, thus the usage frequency is significantly dependant upon the recovery time and hence thermal design.



**Figure 7:** Steady state model of a thermal capacitor in an example insulation scheme.

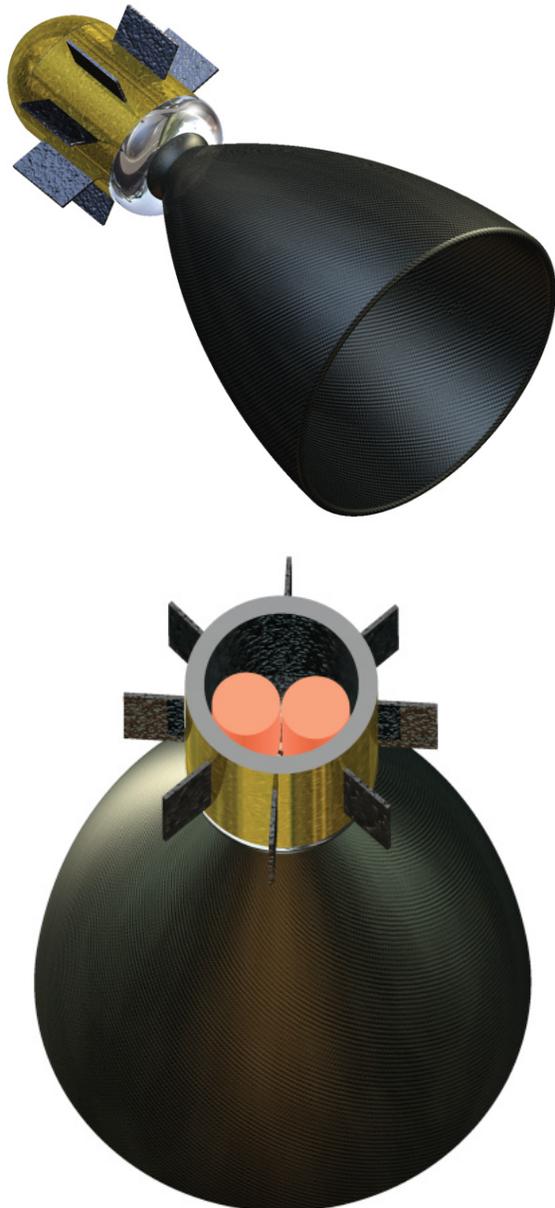
Initially, the capacitive heat source is thermally isolated from the outside environment via an insulation scheme such that the decay heat produced by the radioisotope raises the temperature of the heat capacitor. Once the desired operational temperature has been achieved, energy may be extracted from the system via either momentary direct coupling to a heat engine or through the passage of a working fluid through an integrated heat exchanger. Once the systems temperature is lowered to a predetermined minimum, the heat capacitor must once again be thermally decoupled from external systems so as to re-initiate the heat accumulation process.

Radiated heat that would otherwise be lost to the environment may be harnessed through Thermophotovoltaic, infra-red antenna arrays<sup>9</sup> or through thermoelectric converters coupled to a black body thermal absorber (graphite). Similarly, heat pipes may be used to transfer energy from a graphite absorber to a pair or multiple pairs of Free Piston Stirling engines.

The heat source design requires the use of a material that has a high specific heat capacity over the temperature range of interest, with a radioisotope loaded cermet nested inside the heat capacitor matrix. The use of tungsten-radioisotope cermets<sup>10, 11</sup> installed in a beryllium matrix is an appropriate solution to this requirement from a thermo-mechanical and safety standpoint. Sufficient capture of the source  $\alpha$  particles is also achieved via the tungsten matrix such that  $\alpha$ -n reactions at the Be interfaces are negligible. Beryllium is capable of providing significant shielding of neutron fluxes generated by the radioisotope source while offering excellent thermal conductivity and large heat capacity. Work performed by Karditsas et al.<sup>12</sup> at the United Kingdom Atomic Energy Agency Fusion Division has evaluated the specific heat capacity of Be as a function of temperature over the range of 300 K to 1500 K. The specific heat capacity of Beryllium may be approximated from the data presented by Karditsas et al.<sup>12</sup> by the function described in Equation 1 below.

$$C_p = 974.77 \ln(T) - 3668.70 \quad [1]$$

### III. AN ADVANCED RADIOISOTOPE THERMAL ROCKET (RTR) ENGINE CONCEPT



**Figure 8:** *Conceptual twin-capacitor Radioisotope Thermal Rocket (RTR) engine for use on a Mars Hopper platform.*

The concept of radioisotope thermal rocketry was first developed during the U.S. Space Nuclear Auxiliary Power (SNAP) program. This research culminated in the development of a bimodal system known as SNAPOODLE that was capable of delivering both electrical power and impulsive thrust via the passage of a stored propellant through the heat source<sup>13</sup>. The SNAPOODLE engine required a thermal inventory of several kilowatts of radioisotopic materials and as such drove the design to use <sup>210</sup>Po as a heat source due to the

cost and historical unavailability of other longer lived isotopes<sup>14</sup>. Thus, the significantly limited lifetime of the system was a major contributing factor to the termination of the research.

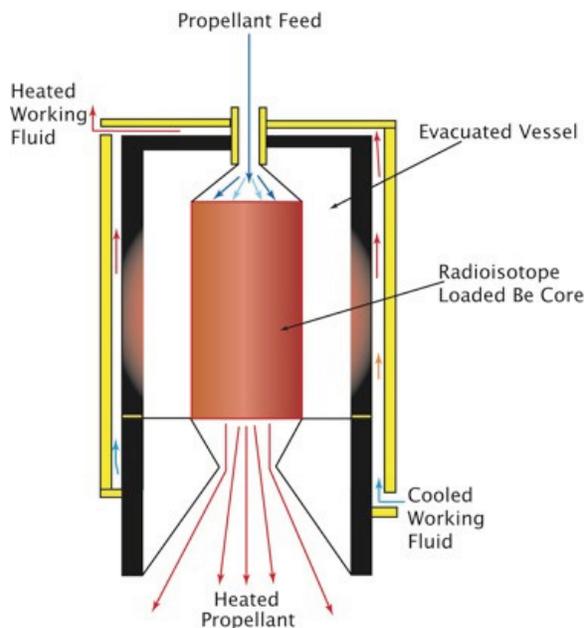
Unlike the SNAPOODLE system, a thermally capacitive radioisotope thermal rocket is proposed by the Center for Space Nuclear Research that will be capable of utilizing multiple propellant species, either stored or produced from in-situ resources while requiring a greatly reduced thermal inventory of radioisotopic materials. Specifically, the passage of a propellant through an appropriately designed beryllium heat capacitor allows for initial heat transfer rates of several tens to hundreds of kilowatts while requiring an isotopic loading of only hundreds of watts. Thus, thrusts that are 1 - 2 orders of magnitude greater than those achieved by the SNAPOODLE design are conceivable for up to only 1/3 of the radioisotope inventory.

Based on the heat capacitor principal described above, work undertaken at the Center for Space Nuclear Research has identified a novel mission concept that will facilitate long range mobility across the entire surface of Mars, limited only by the half-life of the radioisotope used to supply energy to the heat capacitor. In this concept, CO<sub>2</sub> is liquefied from the Martian atmosphere via a miniaturised compressor / refrigeration system and is stored within a tank while a thermally isolated heat capacitor is elevated in temperature by radioisotope decay heat. Once the core has achieved its target temperature, CO<sub>2</sub> is fed through a series of flow channels within the core, elevating the temperature of the gas prior to its exit through a rocket nozzle for generation of propulsive thrust.



**Figure 9:** *Artist's impression of a Mars Hopper platform enabled by the CSNR's RTR concept.*

Thermal losses via radiation may be harnessed for power generation applications using single or hybrid power conversion mechanisms. This may be possible through the use of a black body absorber on the inner surface of the evacuated vessel that provides a thermal interface between the heat radiated and the hot side of a converter such as thermoelectrics or Stirling engines. Alternatively, the black body absorber may replace the traditional heat exchangers used within a Brayton or Rankine cycle by allowing for the flow of a working fluid through its interstitial structure. Such methods of power generation are limited to the steady state and heating modes for the hopper core. Thus, it is implied that an alternative power source be used, such as a chemical battery where the core is below a threshold temperature, i.e. at the end of a propulsive operation. Figure 10 illustrates a potential configuration in which a working fluid is cycled through a black body absorber on the inside vessel wall. A similar approach may be used to preheat propellant prior to injection into the upper plenum of the RTR core.



**Figure 10:** Schematic of a heat capacitive Radioisotope Thermal Rocket (RTR) concept engine. Also illustrated is a potential heat exchanger schematic within the walls of the evacuated chamber that may be used directly within a dynamic power conversion system or for the preheating of propellants prior to injection into the upper plenum of the RTR core.

Further detailed discussion of the Mars Hopper mission concept is provided in a companion article submitted to section C3.5 of the IAC 2010 proceedings.

#### IV. CONCLUSIONS & FUTURE WORK

Our experimental research has successfully demonstrated the compatibility of the Spark Plasma Sintering process to the production of cermets consisting of simulated ceramic radioisotope materials and tungsten. This research has also found that for applications where it may be necessary to control the porosity of product cermets, the grain size and volumetric ratio of tungsten used in the cermets can be used to tune this porosity.

Future enhanced examination of encapsulated cermet structures will be performed to test the integrity of the materials at high temperature gaseous and shock conditions. At the time of writing, the Center for Space Nuclear Research is continuing to undertake experimental verification of the chemical compatibility of radioisotope oxide compounds, specifically those of  $^{241}\text{Am}$  and  $^{238}\text{Pu}$ , with tungsten encapsulation. The reduction of  $\alpha$ -n reaction yields for tungsten-encapsulated sources will also be verified. The findings of these experiments will be the subject of a future paper.

Clearly, thermally capacitive heat sources as proposed above require integration with a compact radioisotope source. Traditional isotopic heat sources that require an external aeroshell are unsuitable for such integration. Conversely, tungsten cermet based heat sources are expected to deliver a higher volumetric power density of the order of 4 times that of current heat source technologies. Therefore, it is envisioned that such advanced cermet based radioisotope heat source technologies are both enabling and essential for future power and propulsion systems and the future missions under which they will be used.

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