Sol Gel Production of Uranium and Plutonium Microspheres for Space Nuclear Power and Propulsion

Future Directions for Space Nuclear Power and Propulsion Workshop

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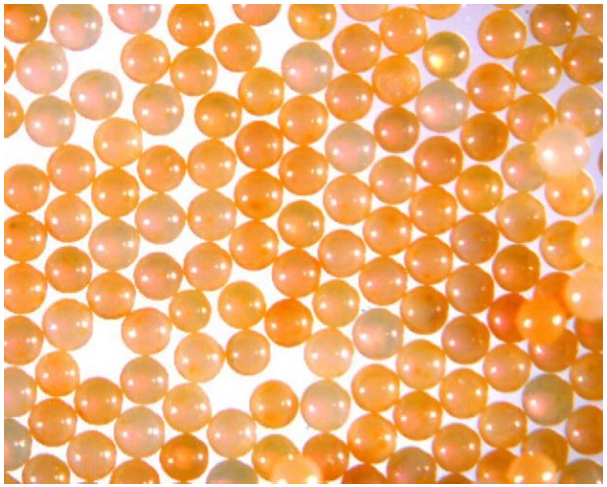
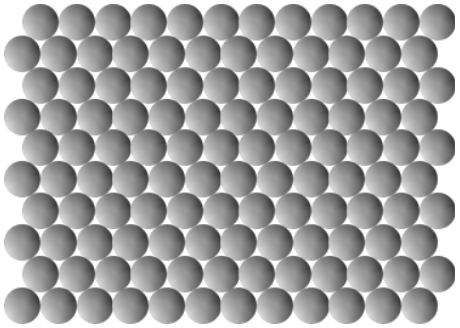
Overview:

- Concept Overview
- Why Sol Gel for Space Nuclear?
- External vs. Internal Gelation
- The Internal Gelation Process
- Process Capabilities
- Research at Michigan



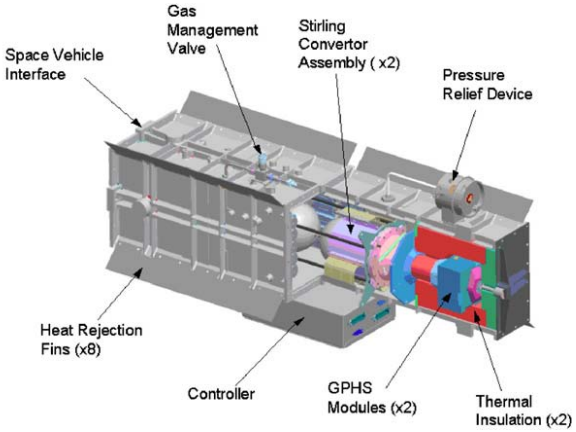
300µm Zr-Y Microspheres by ORNL [1]

Concept:

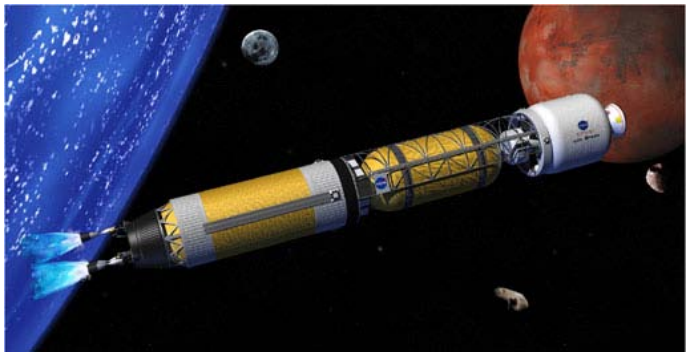


Sol Gel Oxide Microspheres of Uranium, Plutonium-238, Americium-241, or Curium-244

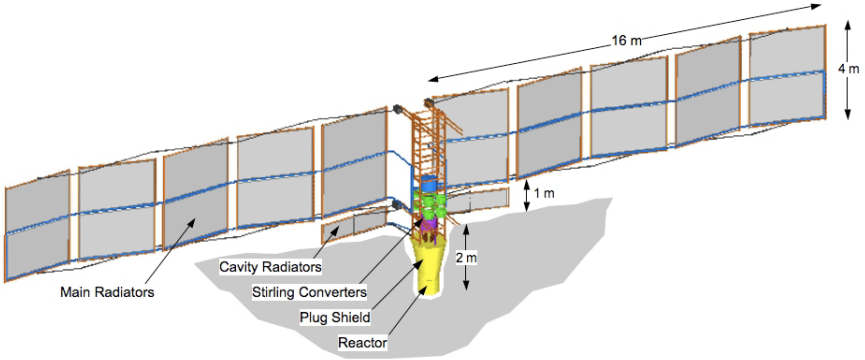
Tungsten Cermets Fabricated by the CSNR [2]



Advanced Stirling Radioisotope Generator [5]



Nuclear Thermal Propulsion [3]

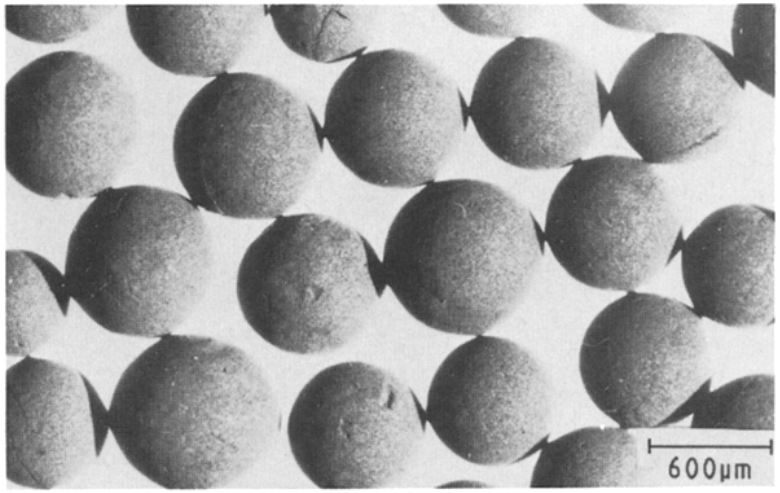


Fission Surface Power [4]

Introduction:

Why use sol gel processes for space nuclear power sources?

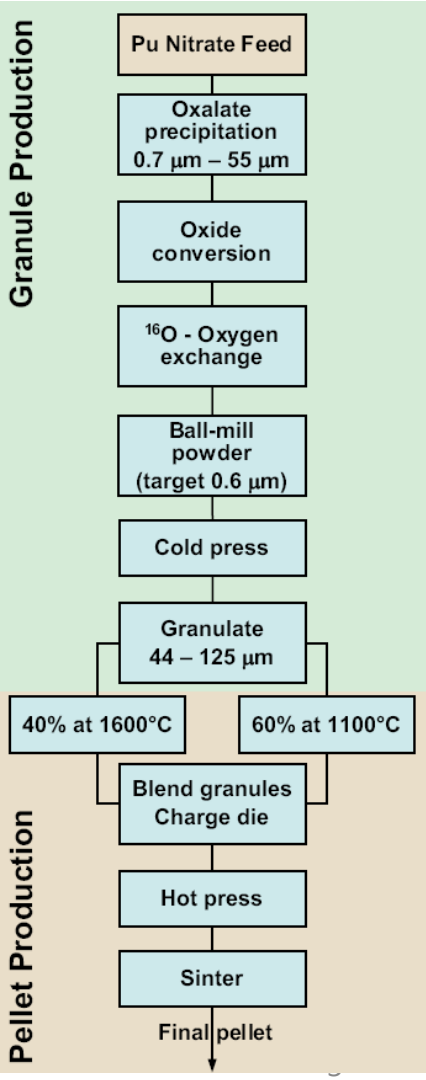
1) Improved Safety
2) Lends to Automation
3) Desirable for Advanced Encapsulation Methods



UO_2 Microspheres after Calcination [6]

Improved Safety:

- <u>Current Process Requires:</u>
  1) Ball milling to sub-micron powders
  2) Glove box operations with fine powders
  3) Many steps
- <u>Sol Gel Route Allows:</u>
  1) Direct route from nitrate solution to microsphere (dust-free) with fewer steps
  2) Free flowing microsphere product
  3) Less human handling → automation



Current GPHS Process Flowsheet [7]

Automation:

- System Input = Nitrate solution following Pu/Np separation
- System Output = Gelled particles

- Suitable for operation in a hot cell – reduce worker dose

- Hot cell operation also allows alternative isotopes to supplement Pu-238 supply:

  - Am-241 (59.5keV gammas)
  - Cm-244 (SF neutrons & gammas)

PuO_2: ~0.4 W/g, 87.7yr
AmO_2: ~0.1 W/g, 432.7yr
Cm_2O_3: ~2.2 W/g, 18.1yr

Remote Manipulators at INL's MFC [8]

Advanced Encapsulation:

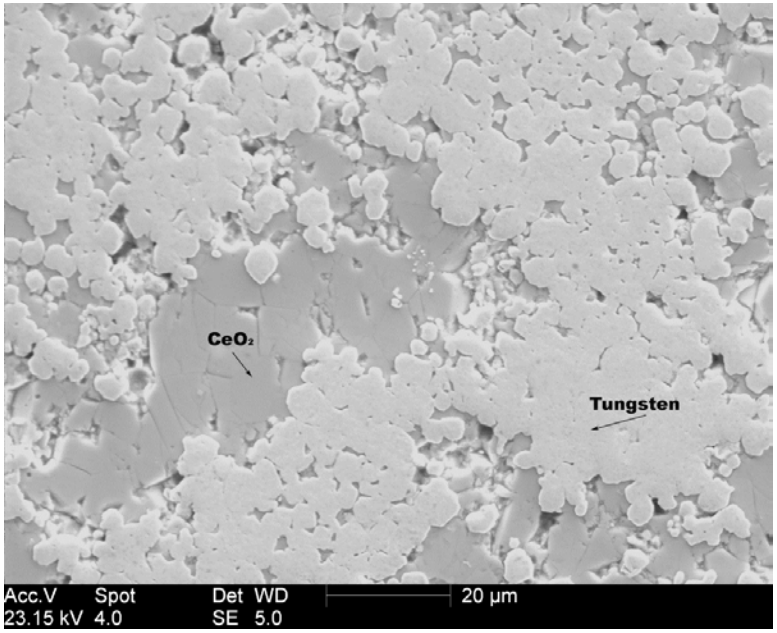
Neutron Science Laboratory

Studies by the CSNR indicate:

1) Universal encapsulation method using a tungsten matrix are attractive for a range of space power applications

2) Cerium (as actinide surrogate) can be microencapsulated in tungsten effectively via Spark Plasma Sintering

3) Higher product densification may be achieved via SPS using spherical actinide particles in 100µm size range

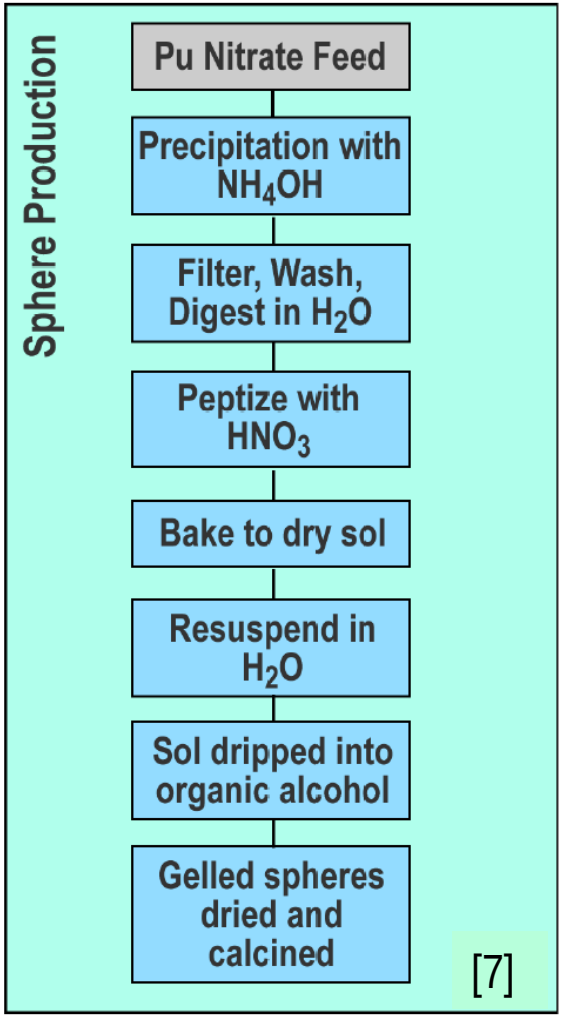


SEM Image of tungsten-ceria cermet [2]

External vs. Internal:

Neutron Science Laboratory

External Gelation



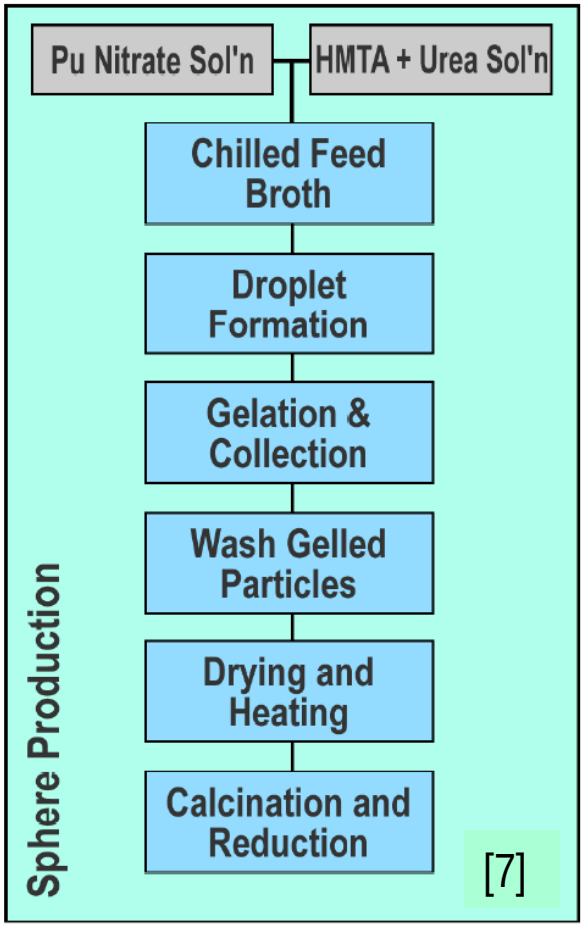
External:

- Higher density, low porosity
- Several steps to prepare Plutonium sol
- Solidifies in an organic solution
- Based on mass transfer through a boundary

Internal:

- Lower density, open porosity
- Pu nitrate solution from Np/Pu separation used directly and mixed with HMTA & Urea (chilled)
- Solidifies in silicone oil
- Based on decomplexation with heat addition

Internal Gelation

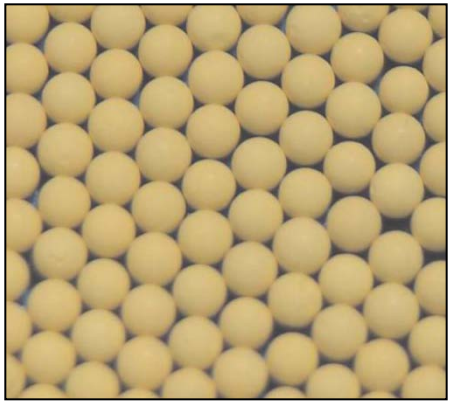


Internal Gelation Product:

- Tunable particle size from ~100 – 1000+µm
- Monodispersity in chosen sphere diameter

- Gelled spheres after washing: hydrated UO_3
- After calcination: UO_2

- Pure oxide pellet formation studies dominant
- Determination of optimal sphere properties for encapsulation via SPS to be determined



Air-dried $UO_3 \cdot 2H_2O$ microspheres with diameters of 1000 µm [9]

Research at Michigan:

Sol Gel Testing Rig at UM

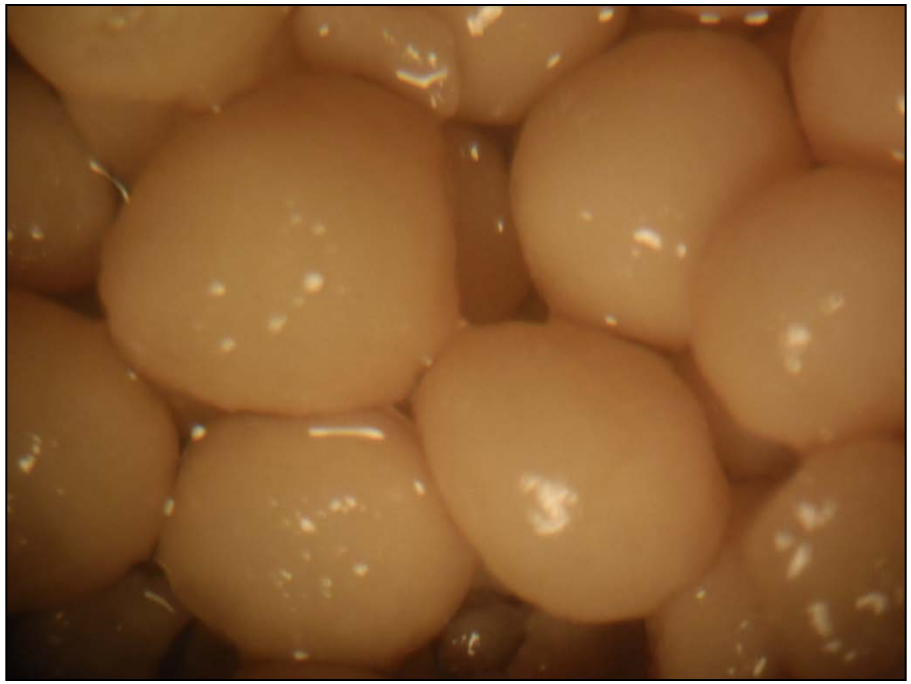
Efforts to produce (via Internal Gelation):

- Ceria microspheres as non-radioactive surrogates
- UO_2 microspheres
- PuO_2 microspheres
- Currently performing work with cerium and preparing for uranium work

Research at Michigan:

Neutron Science Laboratory

- Currently refining automated gelation column apparatus
- Basic chemistry demonstrated with cerium - gelled cerium particles obtained
- Heat treatment steps to achieve desired density/porosity to be completed



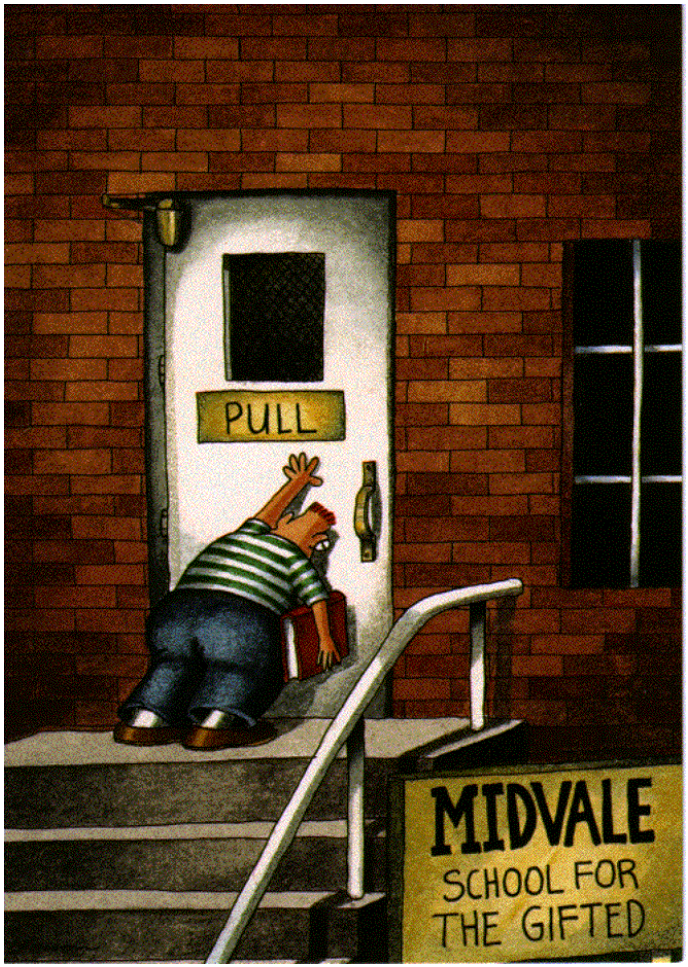
Preliminary Results: Gelled Cerium Microspheres

Future Work at Michigan:

Neutron Science Laboratory

- Determination of Achievable Sphere Size Distribution
- Production of Depleted Uranium Microspheres
- Production of Pu-239 Microspheres
- Refinement of Heat Treatment of Particles for Further SPS Processing
- Characterization of Density/Porosity, Post-Fired Oxygen Content, and Impurity Concentrations

Questions?



References:

[1] R.D. Hunt, F.C. Montgomery, and J.L. Collins, “Treatment Techniques to Prevent Cracking of Amorphous Microspheres made by the Internal Gelation Process,” Journal of Nuclear Materials, vol. 405, p 160-164, 2010.

[2] R. C. O’Brien et. al., “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, Atlanta, Ga., June 14-19, 2009.

[3] “NASA – Bimodal Nuclear Thermal Rocket,” NASA, 2 May 2008, Web, 7 Sept. 2011, <http://www.nasa.goc/centers/glenn/multimedia/artgallery/art_feature_002_BNTR.html>

[4] J. Werner, “A Lunar Fission Surface Power (FSP) System,” Presented at Nuclear and Emerging Technologies for Space 2009, Atlanta, Ga., June 14-19, 2009.

[5] T. S. Balint and J. F. Jordan, “RPS Strategies to Enable NASA’s Next Decade Robotic Mars Missions,” Acta Astronautica, vol. 60, Feb 2007, p 992-1001.

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[7] Borland et. al., “Evaluation of Aqueous and Powder Processing Techniques for Production of Pu-238 Fueled General Purpose Heat Sources,” Idaho National Laboratory, INL/EXT-08-14017, June 2008.

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[9] R. D. Hunt and J. L. Collins, “Uranium Kernel Formation Via Internal Gelation,” Radiochimica Acta, vol. 92, p 909-915, 2004.