Economical Production of Pu-238: Feasibility Study

NASA NIAC Phase I

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5 Bankers Questions

- Who wants it?
  - Historical use
  - Potential users

- What is it?
  - Production methodology

- Where is the competition/collaboration?
  - DOE production program

- How is it Unique?
  - Benefits of the technique

- What does it cost?
Historical Use of Pu-238

- Pu-238 has been used in most space missions since the early days of Apollo
- RTGs still function on the lunar surface
- RTGs are on the farthest man-made object, Voyagers 1 and 2, now near 100 AU from Earth
- Domestic Production Ceased in 1988
- MMRTG powers Curiosity on Mars
- Any mission past the asteroid belt needs Pu-238 (JUNO exception)
- ASRG was intended to reduce demand by x4 compared to MMRTG - program stalled so next Mars Curiosity II may use a good fraction of the remaining Pu
Demand for Pu-238 NASA Planning

Missions to outer planets planned by NASA circa 2010

NASA mission plans assuming a 1.5 kg/yr production rate of Pu-238- circa 2011. (assumes ASRG)

The CSNR is developing future systems that require Pu-238

- **Mars Hopper**
  - Hop 6-10 km every 7 days for years – needs 2.5 kg Pu-238

- **Radioisotope Thermal Photo-Voltaic (RTPV)**
  - The use of micro or nano satellites offers the potential for cheaper exploration of the solar system
  - The smallest nuclear source available is the MMRTG at 125 w with a mass of 35 kg, i.e. No power source exists below the 100 w level
  - Pursuing RTPV development with NASA Ames – offers potential for 50-70 kg/kw (X2 reduction in mass versus ASRG; 6X reduction versus MMRTG)
  - RTPV could enable Cubesats to be sent throughout the solar system by universities and industry

- **Dual-mode, radioisotope powered propulsion**
  - 2014 NASA NIAC project
  - Enables small launchers, e.g. Leonides or Falcon I, to send probes to outer solar system
  - Relies on pulsed thermal mode (radioisotope thermal propulsion) and pulsed power mode (nuclear electric propulsion)
Other possible users

- **NSF** –
  - plans for 900 instrument stations in Antarctica to measure high energy neutrinos from space
  - Currently has 3 stations using less than 1 w through the winter-dormant mode
  - All stations rely on wind or solar and essentially go dormant in the winter

- **Other agencies** –
  - possible need for untended, long-duration, sensor packages

- So US demand of Pu-238 could very probably exceed 1.5 kgs per year
In 2007, Robert Lange, DOE NE-75 office director, told the ANS space nuclear conference that they were under contract to purchase the last 10 kgs of Pu-238 from Russia for $3M/kg.

2 years later, Russia broke the deal claiming the price was insufficient.

Verbal discussions indicate an asking price of $10 M/kg.

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Production mechanism

- Np-237 + n $\rightarrow$ Np-238 ($\beta$ decay 2.1 d) $\rightarrow$ Pu-238
- Losses
  - Np-237 + n $\rightarrow$ fission (.5 MeV threshold)
  - Np-238 + n $\rightarrow$ fission (large cross section)
  - Np-237 (n,2n) Np236 - contaminant
  - Pu-238 + n $\rightarrow$ fission

- Implies short exposure in high flux and then removal for decay to Pu
Plot of the energy dependent microscopic cross section for $^{237}$Np absorption in red, $^{237}$Np fission in green and $^{237}$Np to $^{236}$Np in blue.
Neutron spectra at the 1 MW TRIGA at Kansas State University
Production

Isotope levels versus irradiation time for 1 g of Np-237 in a flux of 1e14 n/cm2-s (courtesy of Dr. Ken Czerwinski, UNLV)
Basics of Alternative Approach

- Slightly alter the configuration of a large, e.g. 5 MW, licensed TRIGA to accommodate a loop around the core

- Continuously flow target material around the core
  - Residence time in the flux to be few days

- Allow Np-238 to decay for 5-10 half lives (up to 21 days) en route to processing facility.

- Separate Pu from other components in small, quantized batches using resin columns and established methods

- Re-inject run-off back into feed stream

- Allows small, university scale laboratory for processing facility- i.e. substantially reduced cost.
Concept

Produced by Brian Manning, CSNR Summer Fellow 2011
Continuous Target

- Allows short residence times and longer decay times
- Reduces fission product inventory and radioactivity levels
- Allows smaller processing lines
- Smaller facility footprint, i.e. lower cost
- Substantially reduced waste stream
- Allows for alternative isotope production
- Easily adjust to changes in demand
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DOE has initiated a new production program

- Targeted to produce 1.5 kg/yr (now 1.0 kg?)
- Uses the High Flux Isotope Reactor (HFIR) at the ORNL and, possibly, the ATR
- Estimated cost is $100 M
- Estimated production date is 2017 (now 2021?)
- Produces 1000s of gallons of radioactive, acidic, mixed waste per year

- Difficult to meet any demand above the 1.5 kg/yr mark
Issues with current method

- **Production issues**
  - Large mass of Np-237 is inserted into ATR or HFIR for long periods
    - Aluminum pins filled with NpO₂
    - Irradiated for 6 mo to 1 yr
  - Np-238 has a very large thermal neutron fission cross section – roughly 85% of the Np-238 created is fissioned
  - Long irradiation creates a large inventory of fission products
    - Requires dissolving large, radioactive masses in acidic solution
    - Requires a large facility to handle the mass and the high radioactivity levels

- **Fabrication issues**
  - Ball milling of sub-micron powders leads to exposures
  - Reconstitution of NpO₂ from solution involves handling

- **Costly and inefficient**
Potential for improved safety and handling in fabrication of PuO2 fuel pellets

- Current fabrication techniques of the fuel pellet for the General Purpose Heat Source (GPHS) involve ball milling materials.
- Sub-micron material is mobile and has accounted for the exposures at LANL over the years.
- Conceptually, fuel pellets can be fabricated using the INL RSPS furnace that will not involve ball milling.
Fabrication of PuO2 spheres for improved safety in handling

- The output from the ion resin columns is a nitrate solution containing Pu
- Univ of Michigan hired to fabricate spheres directly from a nitrate solution
  - CeO2 completed
  - DUO2 scheduled as a surrogate for PuO2
- CSNR submitted an LDRD in 2013 to fabricate GPHS fuel pellets (using surrogate) and match porosity and density profiles (rejected)
- LANL recently approached Thermal Technologies Corp. to buy a RSPS copy of the INL unit to investigate fabricating PuO2 fuel pellets
- CSNR can do so using DUO2 in a few months
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Benefits

- Significantly simplifies or eliminates target fabrication and target processing facilities
- Reduces time to material production
- Make more efficient use of Np stockpile (less fission losses)
- Provides ability to tailor Pu-238 quality
- More economical operations
- Allows for production of other radioisotopes for medical and industrial use (duo use mode / shared investment)
- Does not require government capital construction funding (commercialization option)
- Government only pays for product received (commercialization option)
Benefits of Continuous process

• Current process
  ◦ Target material is NpO2-20 vol% - reconstituted after separation
  ◦ Fuel clad interactions
  ◦ Fission gas generation
  ◦ 1000s gal of radioactive acidic waste per year*
  ◦ 10s of 5 gal drums of trans-uranic waste per year*
  ◦ Operating costs of HFIR and ATR are high

• Alternative Process
  ◦ Target material is solution that is compatible with separation process
  ◦ No cladding
  ◦ Fission is minimized
  ◦ Waste is estimated at gms/yr – nitric acid solution is recycled
  ◦ Reduced Pu236 content
  ◦ Operating costs of private small reactor are greatly reduced
Issues addressed

- **Production**
  - Impact on reactor operations from large amount of Np solution around core
  - Maximum concentration of Np possible and temperature dependence
  - Neutron spectral shift effect?
  - Residence and decay times for optimization
    - Fission product inventory time dependence and level versus amount of Pu-238 produced
  - Mechanical movement of hundreds of capsules

- **Political**
  - US government must own all SNM. How will price be determined?
  - Use of DOE sites if chosen?
Computational Results
Optimum mass of Pu-238 produced as a function of reactor power. The neutron flux used in the optimization is assumed to be linear with power.
Plot of the Figure of Merit versus irradiation time. The study concluded that 18 days was an optimum irradiation time.
The purpose of the report is to respond to the CSNR report and investigate the mechanics involving the delivery system.

1) the feasibility of fluid mechanics of the delivery system was determined.
2) the neutronics of the transport system was analyzed to confirm the previous analysis done by CSNR.
3) dose estimates were performed with regards to the capsule.
4) materials were recommended for the delivery system and a potential test facility for the system.
5) the more details for the test facility were developed and, regarding the facility, a cost estimate, and technical requirements were produced.
Proof of Concept experiment to verify production rate

- Difficult to calculate rate due to resonance region of cross section
- Verify production rate versus irradiation time
- Differentiate the production at two locations in the reactor
- 4 day irradiation in the Kansas State Univ 1 MW reactor
Successful completion of irradiation experiment at KSU TRIGA
## Summary of calculated yields and experimental results

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<th>Method</th>
<th>RSR yield (gms/gm)</th>
<th>CT yield (gms/gm)</th>
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<td>Hypothetical model (Fig. A3)</td>
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<td>KSU spectra model (Fig. A2)</td>
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<td>KSU experiment</td>
<td>7.47e-05</td>
<td>3.62e-04</td>
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Siting conclusions from study

- Build beside the Materials and Fuels Complex at the INL
- Once operational, move MFC fence around facility
- Share security costs with DOE
- Provide operators and technicians- possibly used by INL
- Costs?
  - Negotiate Pu price with DOE
  - Negotiate Np price with DOE
  - Lease of land from DOE - annual cost and duration
  - What are Costs
    - of security and ops
    - Start up of facility
    - Pre-construction costs – “Next steps”
Costing modeling

- Reactor power is a significant part of the capital cost of the facility.
- The reactor power level dictates the reactor diameter.
- Diameter dictates the amount of Np target material that can be irradiated.
- Thus, each power level has an annual production rate assuming a certain optimum irradiation time.
- Thus, price to meet costs and ROI are determined by the physical system.
Cost assumptions

- Reactor cost is $7 M/MW – determined from literature search for past TRIGA facilities. GA would neither confirm nor deny
  - Excludes cost to modify core

- Up front infrastructure costs - $10 to 40 M
  - Based on discussions with INL nuclear facility staff
  - Accounts for extraction line fabrication, physical security, transport lines, etc
  - Assumptions significantly impact cost estimate

- Operational annual costs - $5 M
  - 6 operators, 2 separation technicians, some administration, internal transportation

- Cost of Np (negotiated with DOE) - $200 K/kg

- ROI - 20%
Price

- In 2007, Robert Lange, DOE NE-75 office director, told the space nuclear conference that they were under contract to purchase the last 10 kgs of Pu-238 from Russia for $3M/kg.
- 2 years later, Russia broke the deal claiming the price was insufficient.
- Verbal discussions indicate an asking price of $10 M/kg.
- A price of $4 - 7 M/kg would appear possible.
Plots of Pu-238 mass produced, price per kg Pu for 20% ROI and price per kg PU for a 0% ROI versus reactor power.
Cost sensitivity to capital costs

Total initial cost = reactor cost + capital cost + Np cost
Total revenue required = operational cost + ROI
Alternative Option Conclusions

- Option for continuous target production of Pu-238 appears to be a viable, cost effective alternative to supplement the DOE program (load follow)

- Allows production quantities to be made in incremental stages- many kgs/yr

- Continuous production process allows small process footprint, minimal materials inventory, and greatly reduced waste stream

- Reduces government up front costs

- Places costs within reach of commercial venture
Next Steps

- Biggest uncertainties
  - Ability of feedline to mechanically move the 15 kg of Np loaded capsules through the line
  - Lifetime of the polymer shells – how many passes
  - Fabrication of GIS pellet using spheres and RSPS with minimal handling
  - Costs – Pu price, Np price, lease, security, permitting
Proposed approach

1) Build mechanical system, free standing, to demonstrate operation of the feedline

2) Irradiations – 4 day
   - Np in H2O-HNO3 solution
   - “ “ D2O “ “
   - Np in D2O filled container
   - Irradiations- long accumulation on polymer samples
   - One irradiation of 18 days for three samples vertically

3) Build time dependent code to model true irradiation history

4) Demonstrate separation chemistry with correct mass throughput using surrogates
Next Steps for USRA?

- Have requested a quote from Merrick Engineering to perform a cost assessment of the facility.

- Have requested a quote from Nuclear Associates to develop a MCNP based computational model of the 5 – 10 MW TRIGA.

- Would like to send a small team to INL in mid February to discuss various issues.

- Interested in forming a collaborative team between INL and industry with DOE participation to define the best path forward.