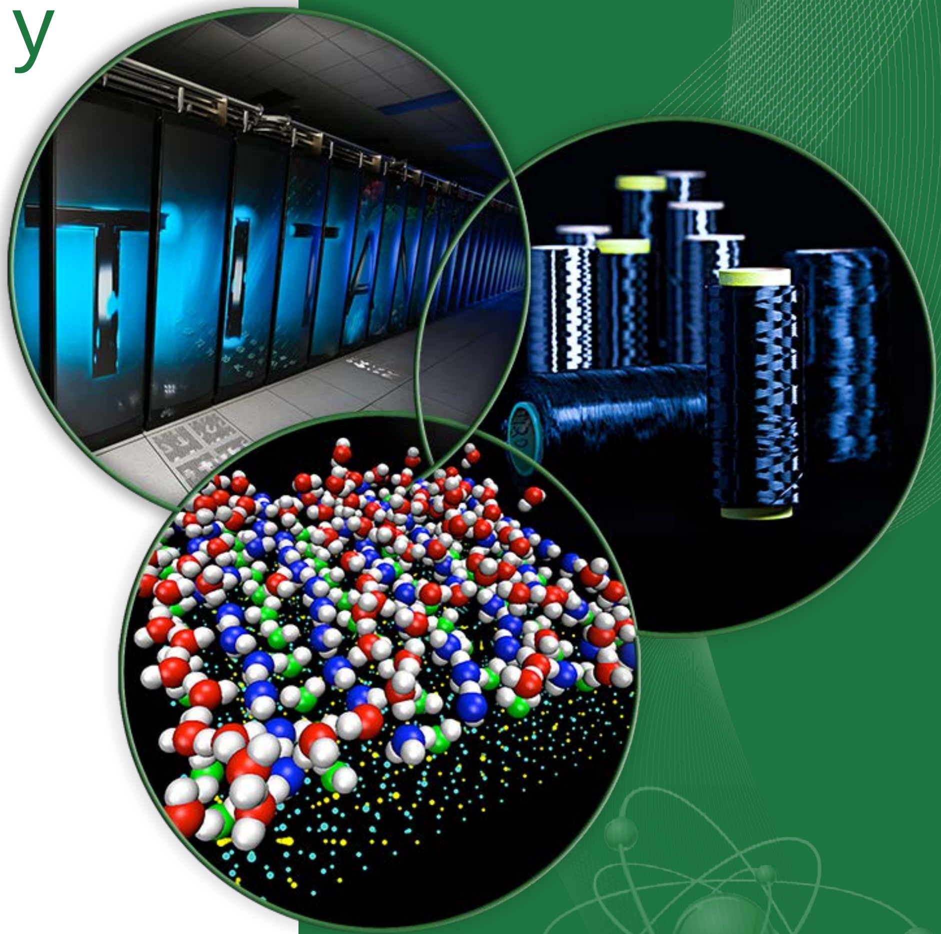
Pu-238 Supply Project–Technology Demonstration

Robert Wham

Presented to

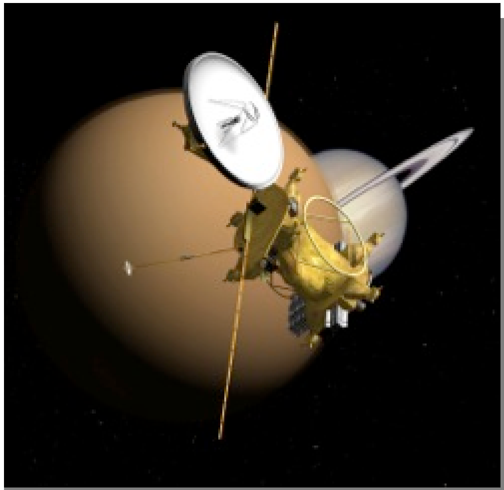
Knoxville AIChE/ANS

February 19, 2015



Prepared by OAK RIDGE NATIONAL LABORATORY, Oak Ridge, Tennessee 37831-6283 managed by UT-BATTELLE, LLC for the U.S. DEPARTMENT OF ENERGY under contract DE-AC05-00OR22725

Radioisotope Power Systems



- Enable and enhance missions by providing electrical power to explore remote and challenging environments where solar power is unavailable
  - Spacecraft operation
  - Instrumentation
- Converts heat from a Radioisotope into electricity
  - Heat is the product of the natural decay process of the isotope

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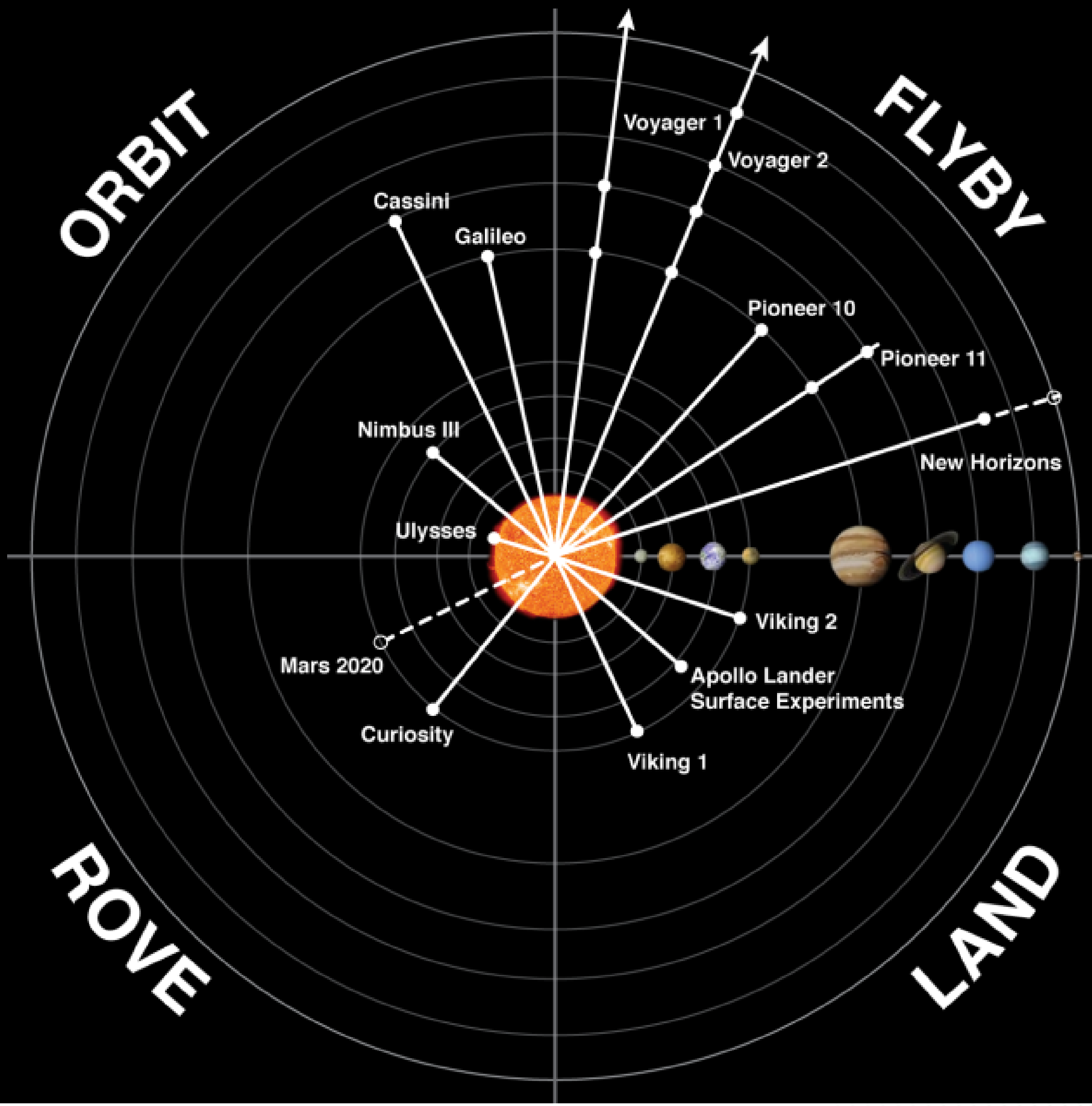
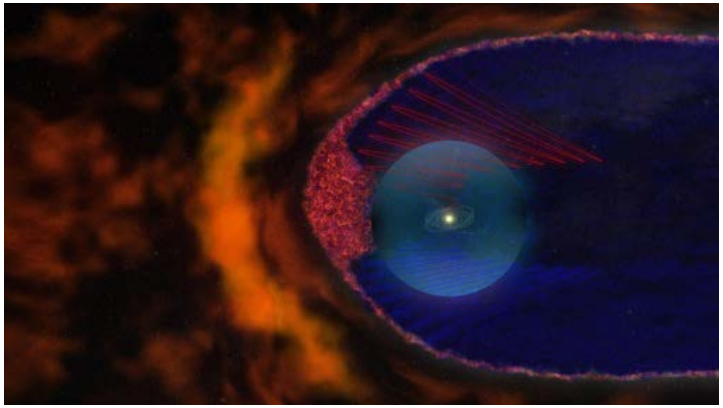
Over 50 years of RPS Missions

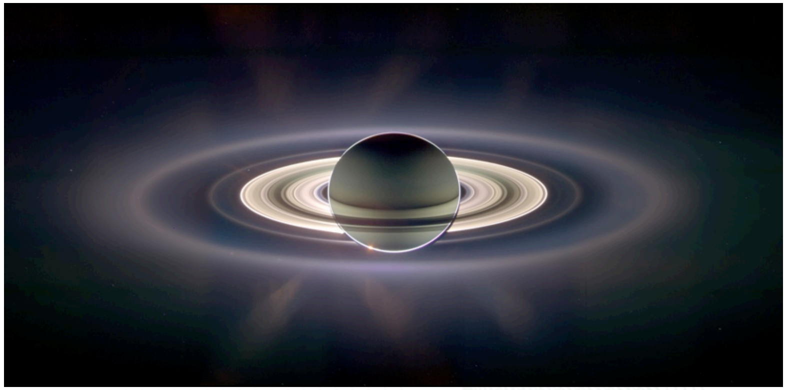


Operational Missions

- Voyager 1 & 2– *Extended Operations*
  - Launched: August 20, 1977 & September 5, 1977
  - Arrival at Jupiter, Saturn, Uranus, Neptune: 1979, 1980/1981, 1986, 1989
  - Science Mission duration: 35+ yr science
  - Power Source:
    - Three MHW-RTG
    - 474 W_e BOM



- Cassini – *Extended Operations*
  - Launched: October 15, 1997
  - Arrival at destination: July 2004
  - Science Mission duration: 7 yr. cruise
  - 10+ yr science
  - Power Source:
    - Three GPHS-RTG
    - ~885 W_e BOM



Operational Missions

- Pluto/New-Horizons – *On its Way (still)*
  - Launched: January 19, 2006
  - Arrival at destination: July, 2015
  - Science Mission duration: 9.5 yr cruise
  - Power Source:
    - One GPHS-RTG
    - 243 W_e BOM; ~200 W_e at arrival
- Mars Science Laboratory – *Extended Mission*
  - Launched: November 26, 2011
  - Gale Crater: August 6, 2012
  - Science Mission duration: ~ 2 yr (1 Martian year)
  - Power Source:
    - One MMRTG
    - ~110 W_e BOM; ~105 W_e at arrival

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Where is New Horizons?

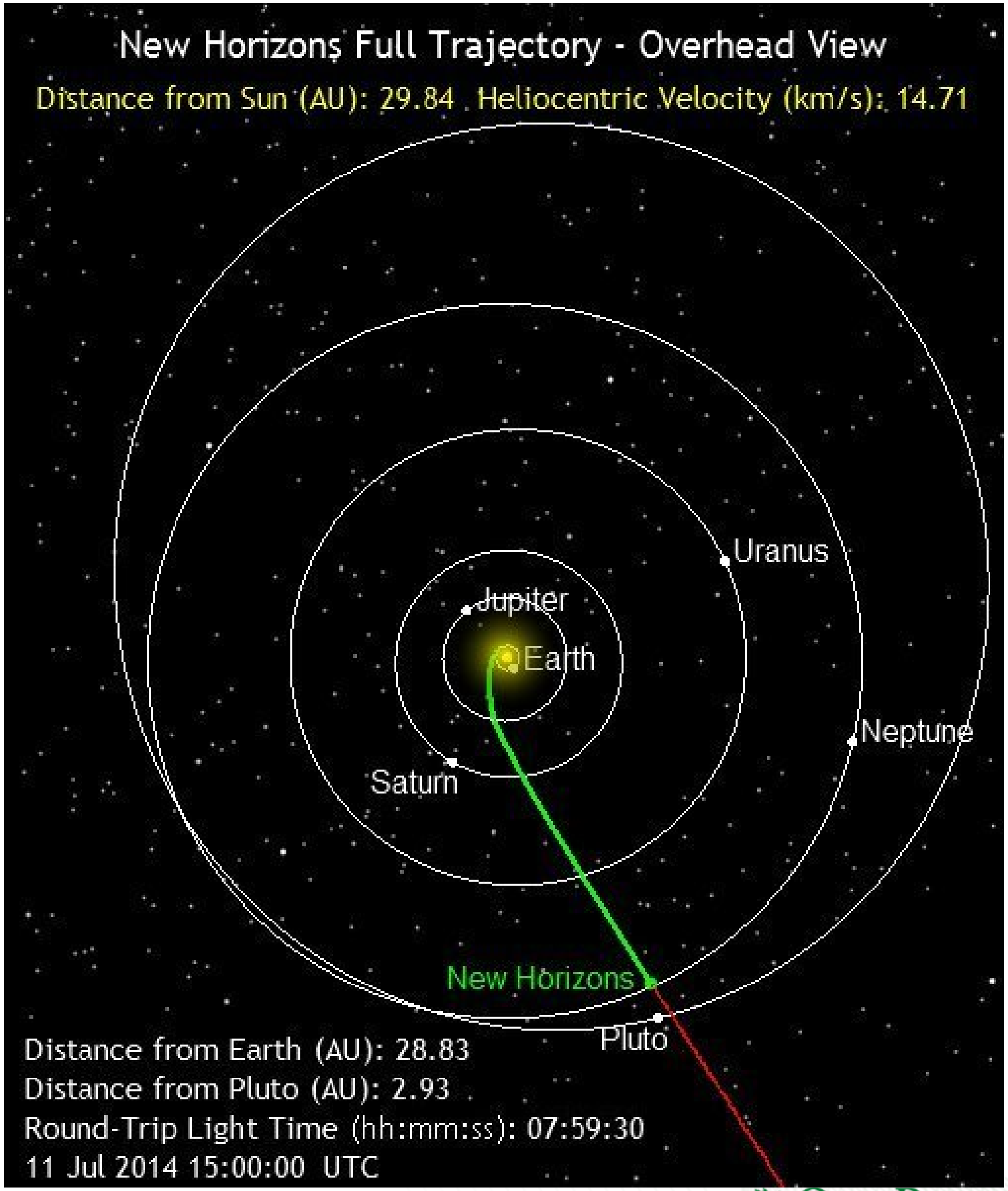
- As of July 11th

Neptune

- New Horizons crossed the orbit of Neptune on August 25, 2014 — exactly 25 years after Voyager 2 made its historic exploration of that giant planet.

Projected Orbit Crossing Dates

- Pluto: July 14, 2015



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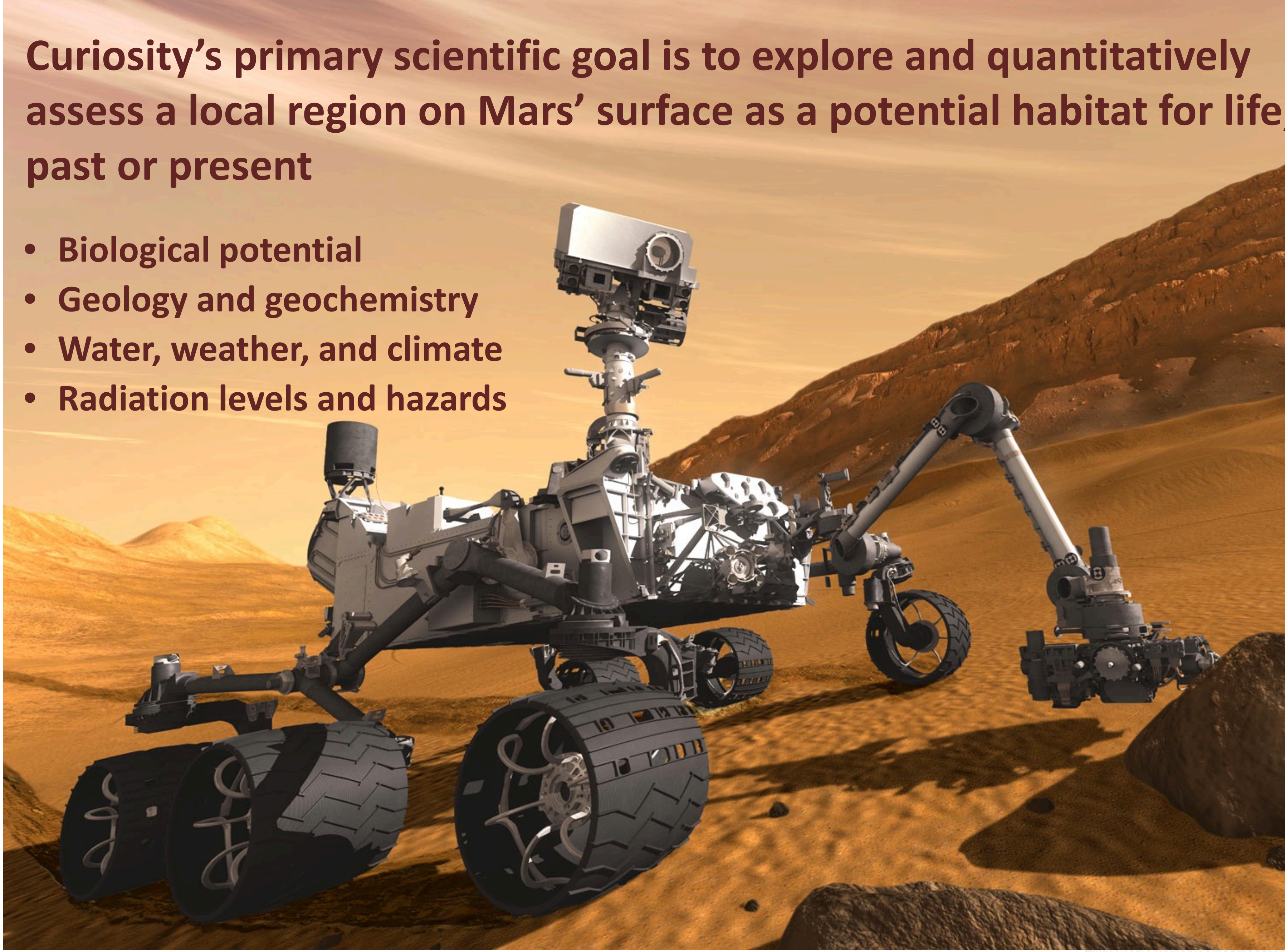
Operational Missions

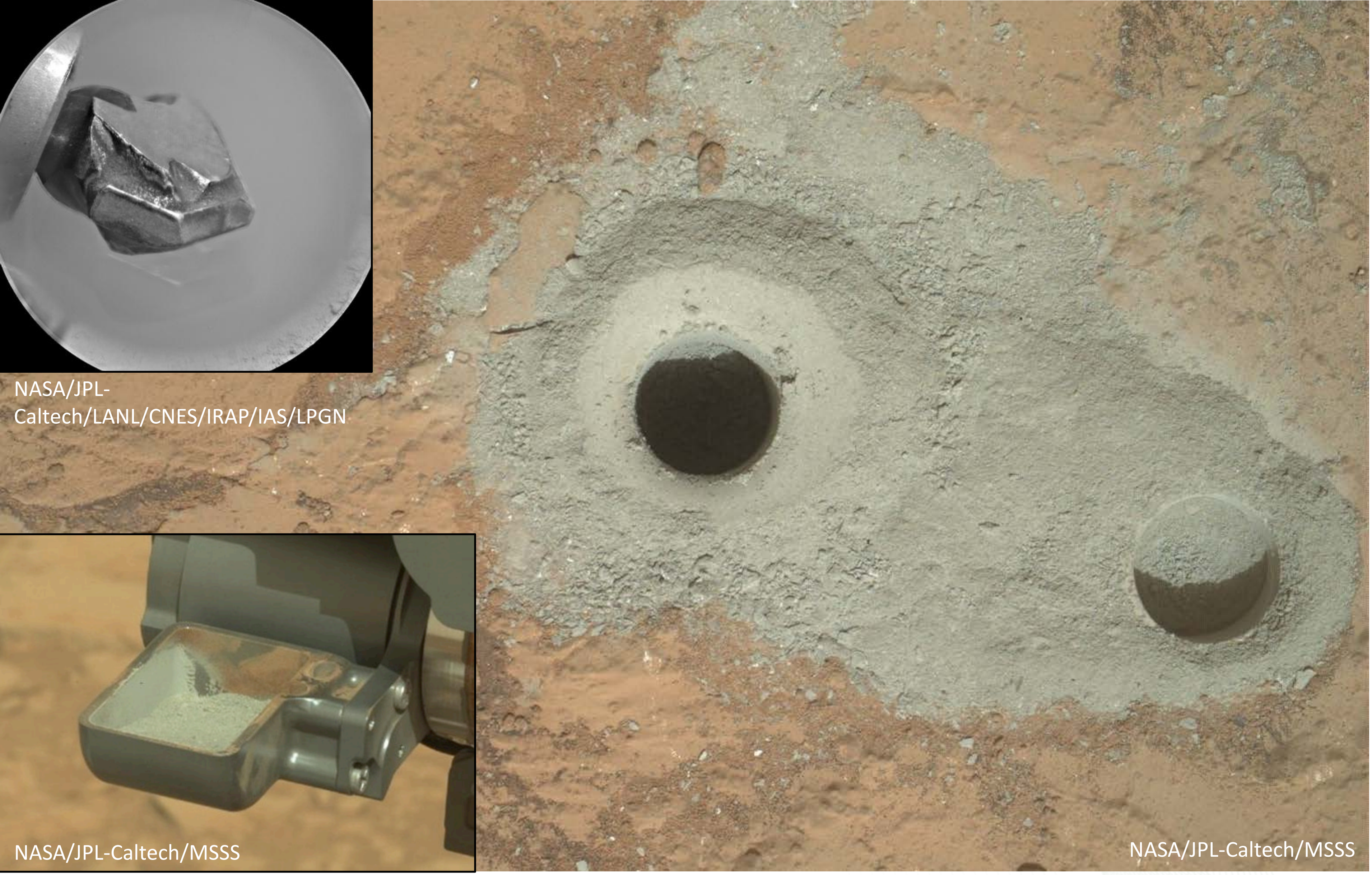
- Pluto/New-Horizons – *On its Way (still)*
  - Launched: January 19, 2006
  - Arrival at destination: July, 2015
  - Science Mission duration: 9.5 yr cruise, 5 yr science
  - Power Source:
    - One GPHS-RTG
    - 243 W_e BOM; ~200 W_e at arrival

- Mars Science Laboratory – *Extended Mission*
  - Launched: November 26, 2011
  - Gale Crater: August 6, 2012
  - Science Mission duration: ~ 2 yr (1 Martian year)
  - Power Source:
    - One MMRTG
    - ~110 W_e BOM; ~105 W_e at arrival

Curiosity’s primary scientific goal is to explore and quantitatively assess a local region on Mars’ surface as a potential habitat for life, past or present

- Biological potential
- Geology and geochemistry
- Water, weather, and climate
- Radiation levels and hazards



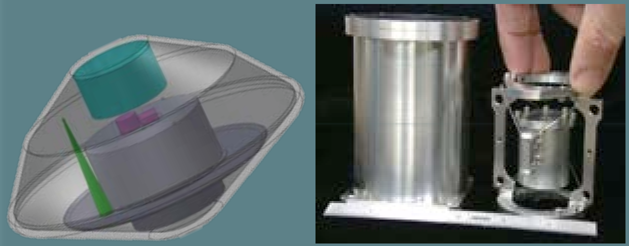


NASA/JPL-Caltech/LANL/CNES/IRAP/IAS/LPGN

NASA/JPL-Caltech/MSSS

NASA/JPL-Caltech/MSSS

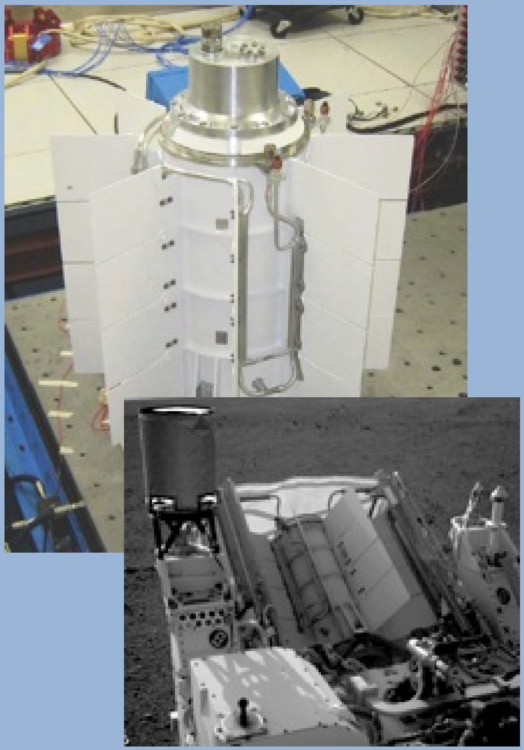
Radioisotope Power Systems



1-RHU mW-RPS Prototype

Radioisotope Heater Unit

- 300 RHUs have been used in 10 missions since 1969 to provide heat to the spacecraft
- Studies are underway to evaluate their use as a power source



Radioisotope Thermoelectric Generators

- 46 RTGs have been used spanning 27 missions
- MMRTG is currently powering Curiosity
- Voyager, Cassini, and New Horizons are RTG-powered as well
- Passive power source that provides reliable energy conversion

Stirling Radioisotope Generators

- Ground-based development
- Dynamic power systems provide increased efficiency and decreased mass

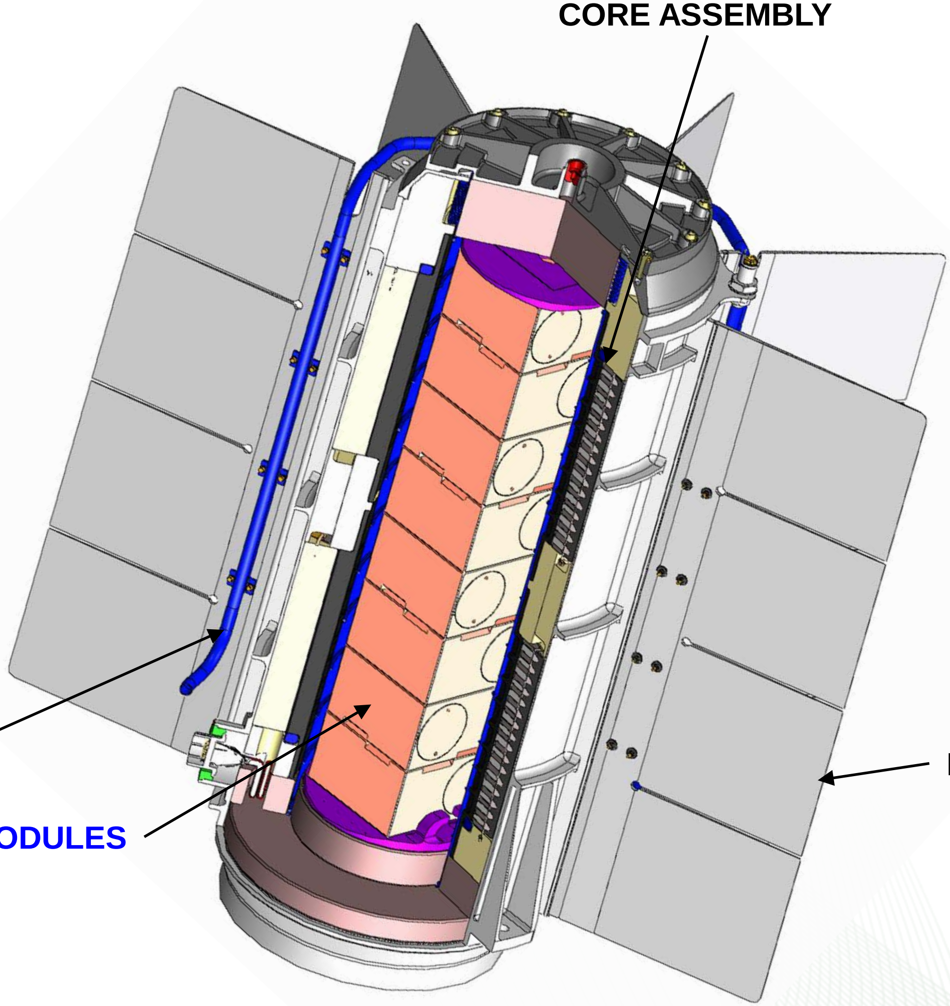
Mars Rovers

Spirit/Opportunity (2004) Sojourner (1997) Curiosity (2011)

MMRTG: Cutaway view

The MMRTG is designed to use heat from General Purpose Heat Source (GPHS) modules.

The MMRTG contains a total of 4.8 kg (10.6 lb) of plutonium dioxide that initially provides approximately 2,000 watts of thermal power and 120 watts of electrical power.



Next Potential RPS Mission

- An Multi-Mission Radioisotope Thermoelectric Generator is baselined to power the “Curiosity heritage” **Mars 2020 rover**
- Enable Mission Goals:
  - Exploring an ancient environment
  - Seeking any signs of past life on the planet
  - Gathering a scientifically compelling sample cache for possible future return to Earth
  - Demonstrating key technologies for future robotic and human exploration

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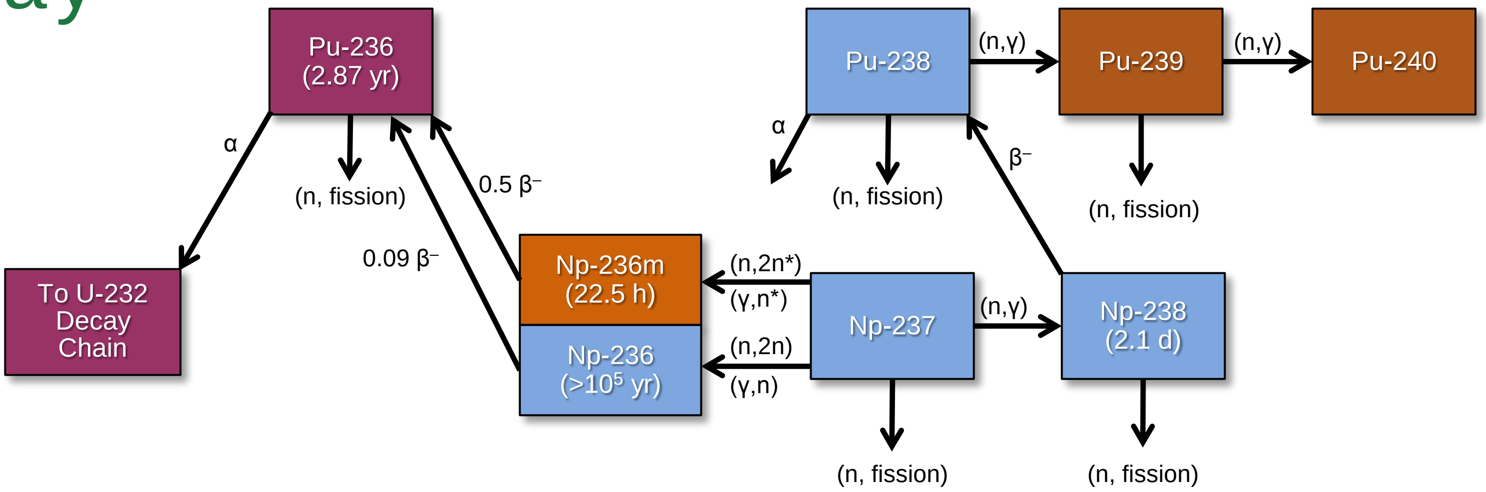
Plutonium-238 is Produced in a Nuclear Reactor via Neutron Capture and Beta Decay



Reactor Characteristics Desired for Efficient ^{237}Np Conversion to ^{238}Pu

Characteristic	Desired to maximize ^{238}Pu	Desired to minimize ^{236}Pu impurity
Neutron spectrum	High thermal flux O(10^{14})	Minimize high energy flux (>7 MeV)
Photon spectrum	N/A	Minimize high energy flux (>7 MeV)
Target size	Large diameter	Small diameter
Neptunium loading	Maximize loading	Minimize loading

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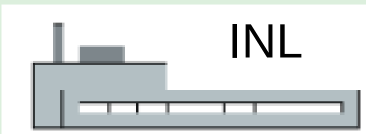
2012-016 RMW

Pu-238 Was Produced Using the Weapons Production Infrastructure at SRS

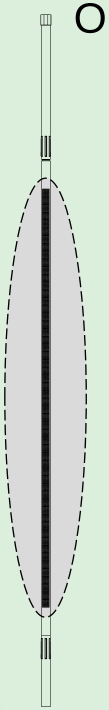
- Irradiation of neptunium oxide mixed with aluminum powder in aluminum clad targets to produce Pu-238
- Target fabrication was based on larger annular (~3" O.D., ~12' long) targets that were designed for heavy water moderated production reactors
- Reactor target volume allowed large batches of Pu-238 to be made in a single campaign (~12 kg batches)
- H-canyon was used for recovery of Pu-238 as product and Np-237 for recycle
- Funding for operations was incremental to baseline weapons programs

The US DOE and NASA Have a Project Underway to Re-establish a Domestic ^{238}Pu Production

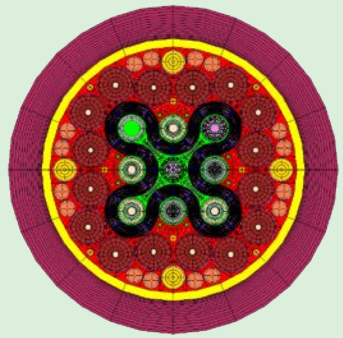
Storage of ^{237}Np

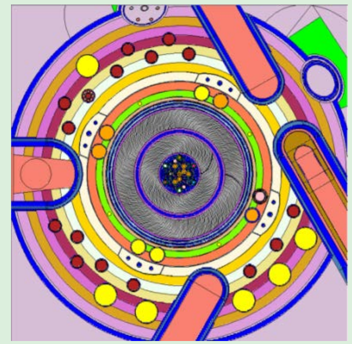


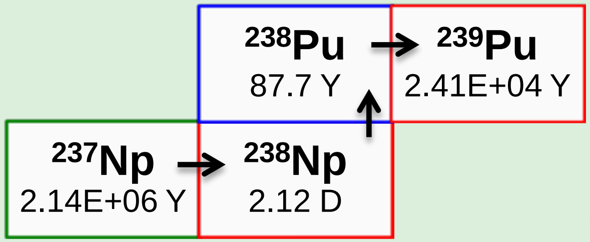
Target fabrication at ORNL REDC



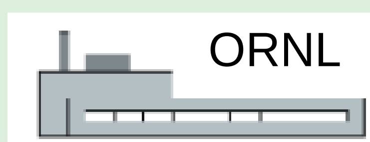
Irradiation of NpO_2/Al pellets
ATR at INL and HFIR at ORNL

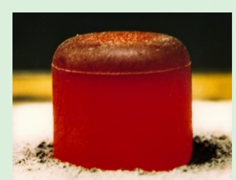




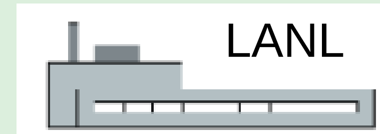


Chemical processing

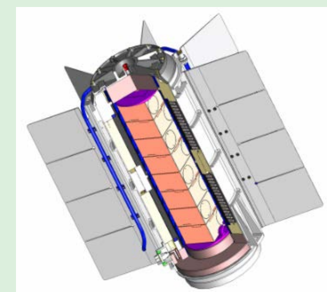




Pu powder → PuO_2



Power source (i.e., MMRTG)



Robotic rover (i.e., Curiosity)

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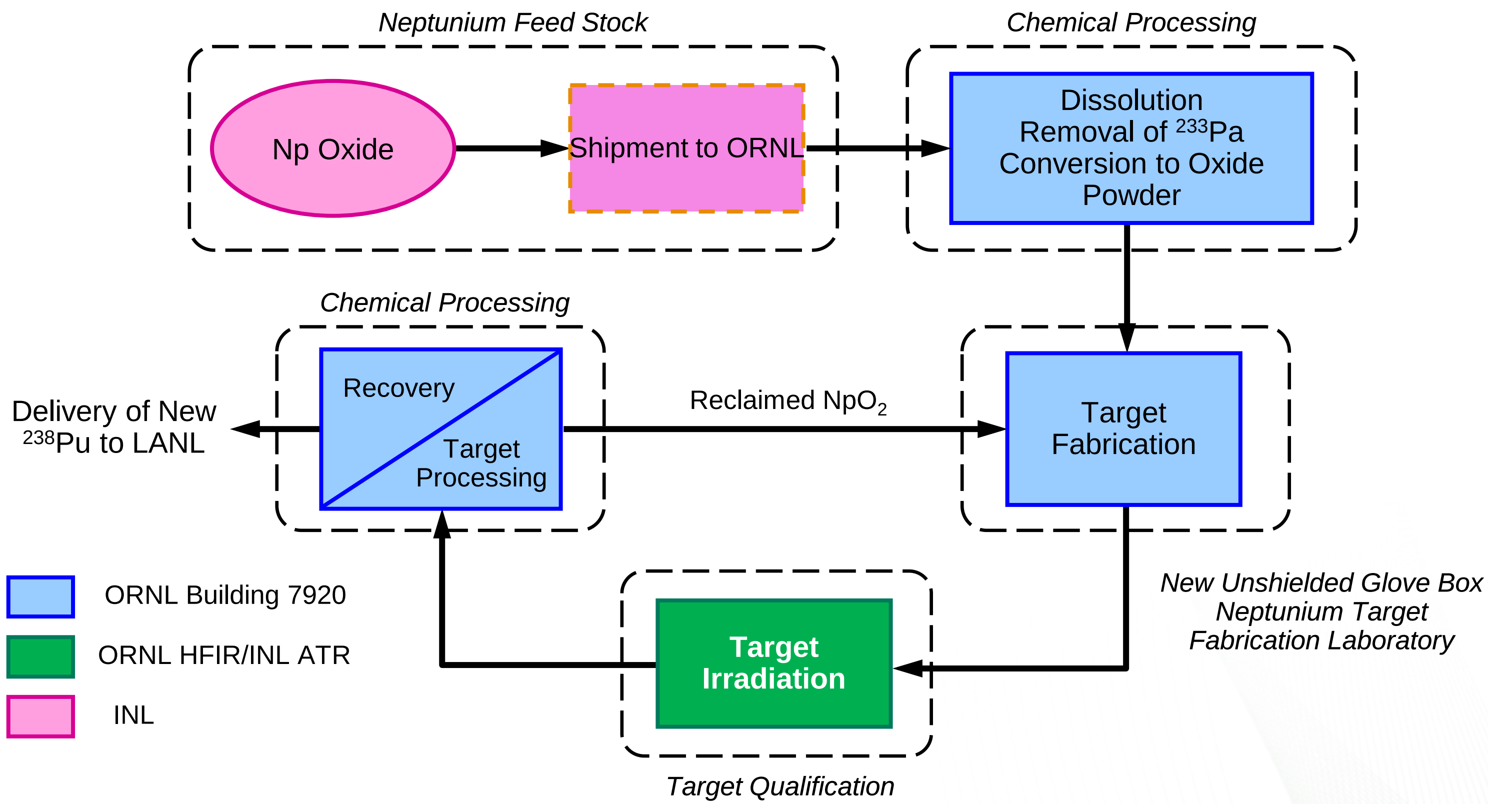
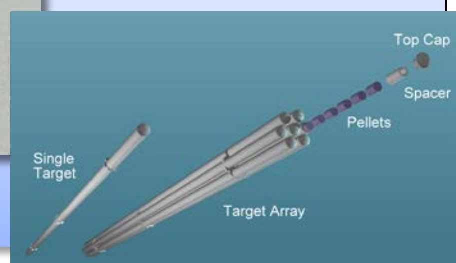
The Path to ^{238}Pu Production Requires Integration of Several Process Steps Using Existing DOE Facilities

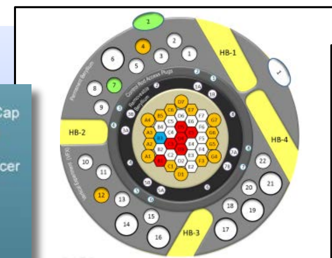


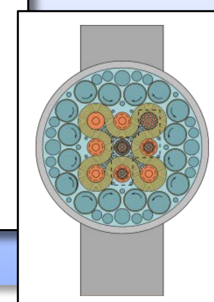
There are Several Stages of Development That Need to be Accomplished to Produce ^{238}Pu

Neptunium Conversion to Oxide

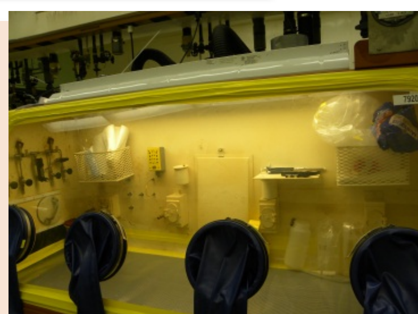
Target Fabrication, Irradiation, and Post-Irradiation Examination

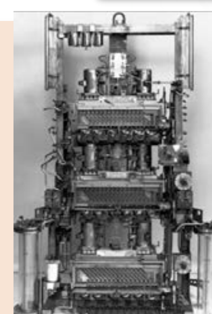


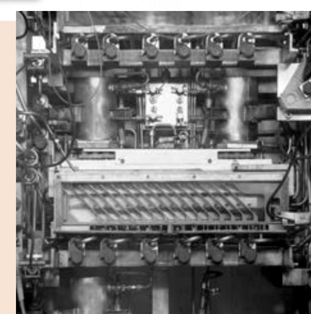




Chemical Separations







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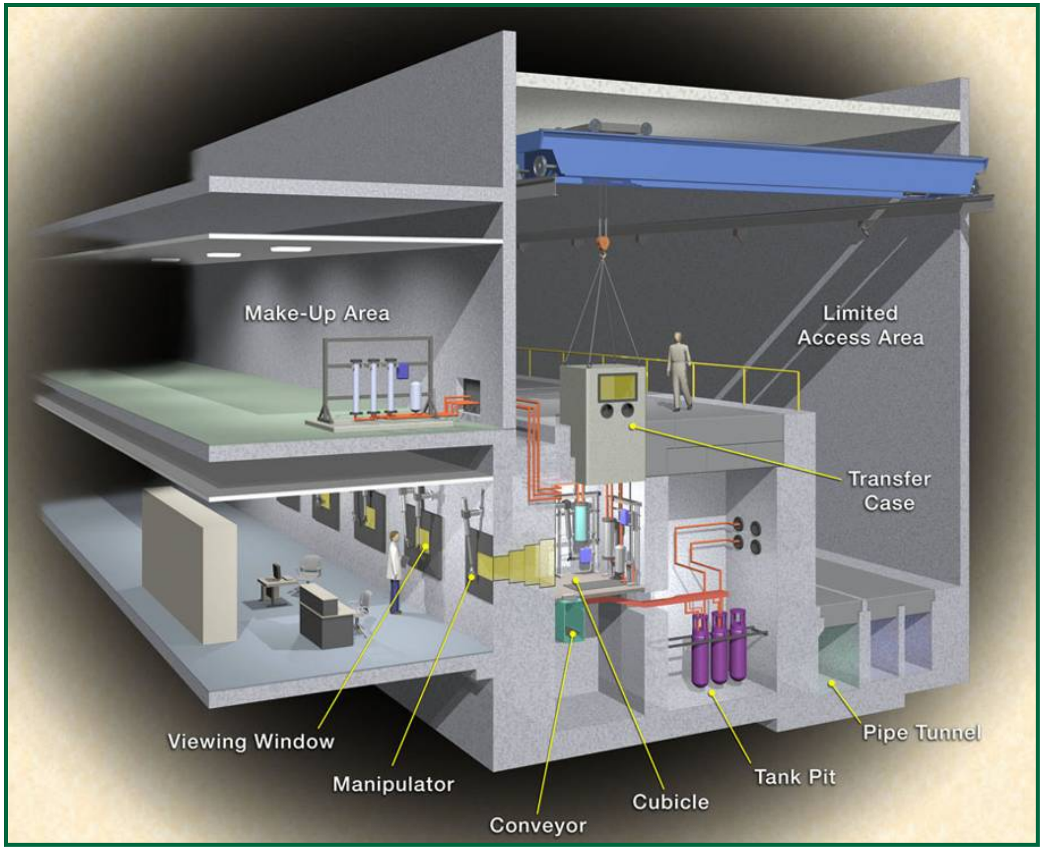
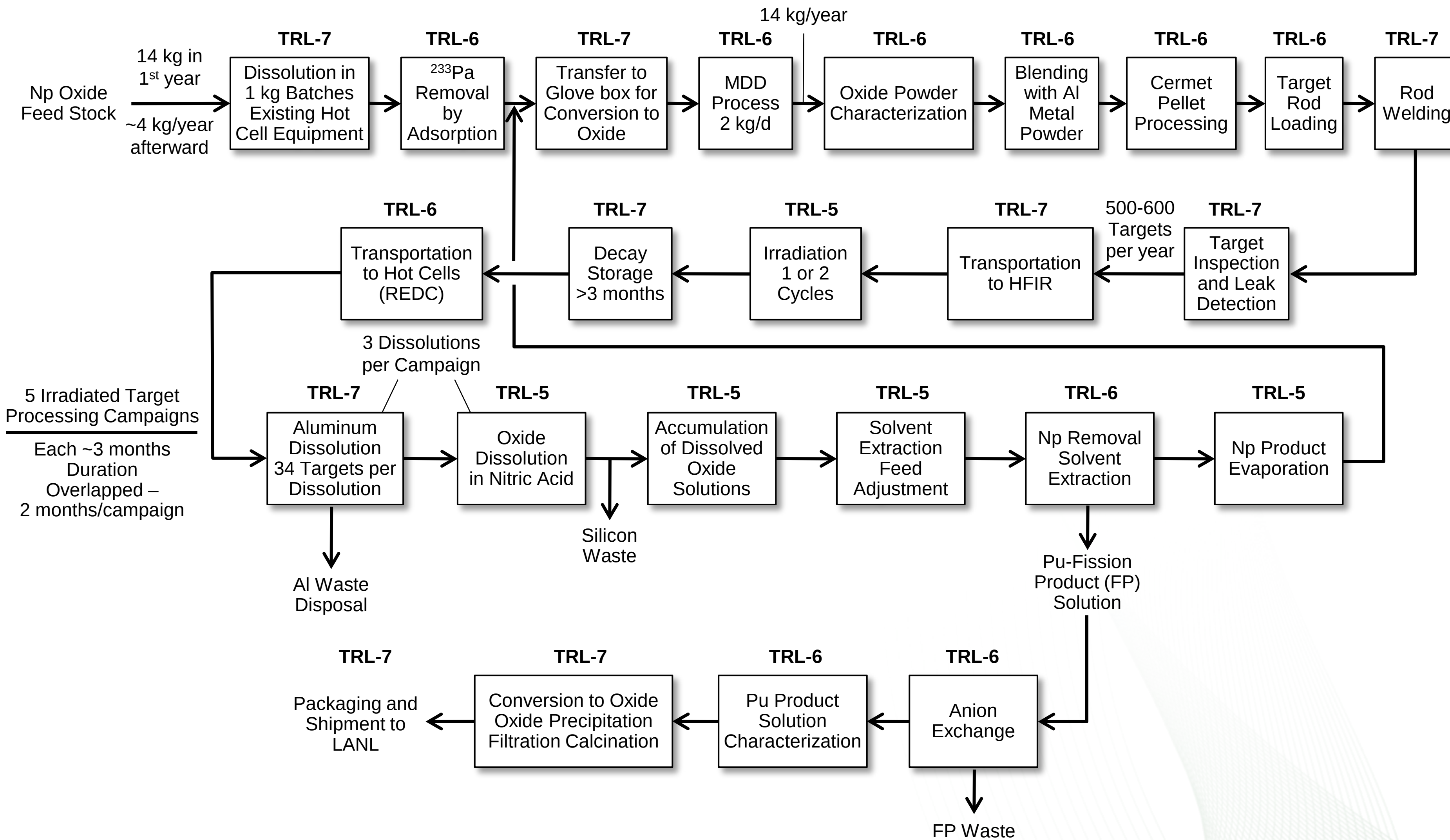
2011-044A RMW

Chemical Processing Study Results: Existing REDC Hot Cells Can Meet Current Projections for ^{238}Pu Production



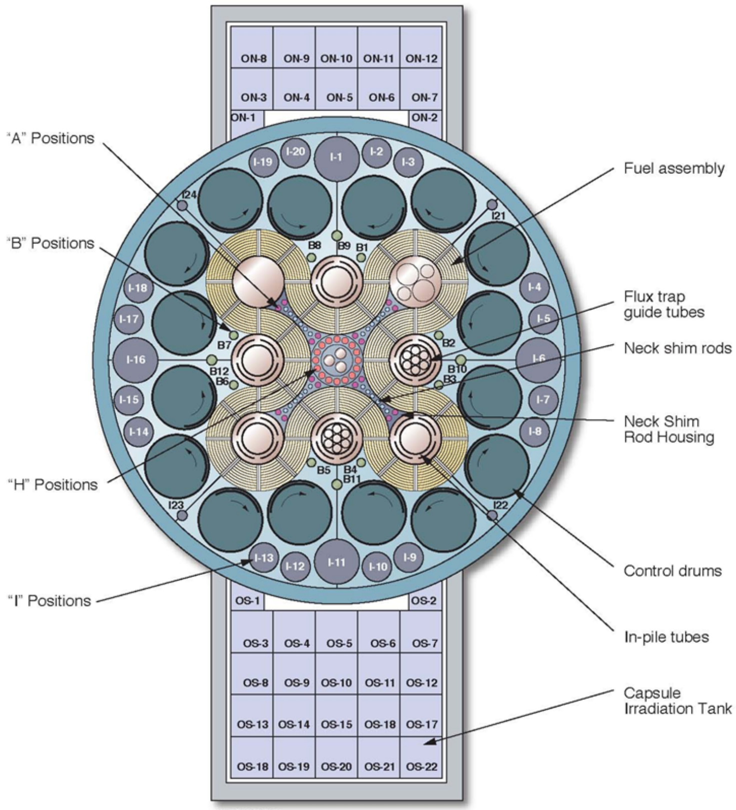
- Currently operating with approved DOE Category 2 Safety Basis – Pu-238 production requires SAR update with similar safety envelope
- Process equipment in place to dissolve, separate, recover and purify Np/Pu products and dispose of fission product wastes
- Fully remotely operated and maintained
- In-house analytical chemistry to support initial R&D activities
- Optimization studies should be conducted to determine opportunities to enhance operations

Production of 2 kg/year of ^{238}Pu in Existing Facilities at ORNL



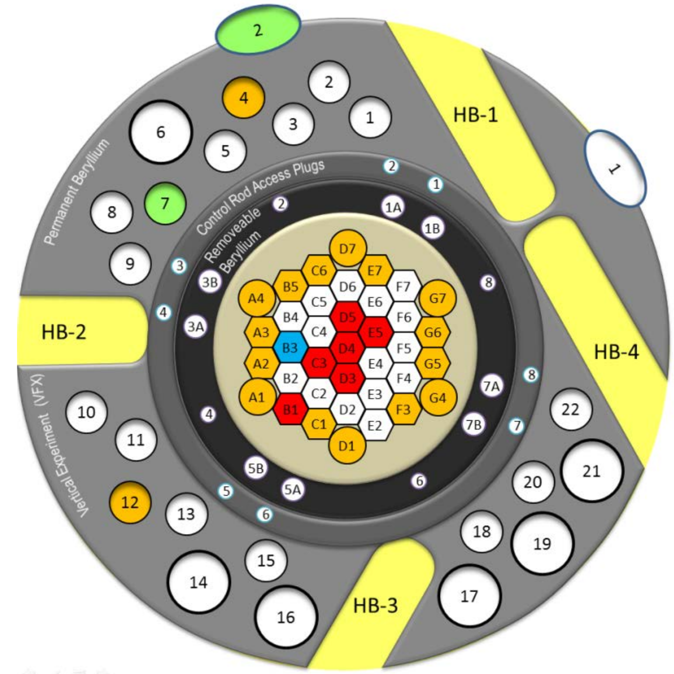
The Advanced Test Reactor and the High Flux Isotope Reactor Will Both Be Used to Produce ^{238}Pu

Advanced Test Reactor (ATR)



Reflector positions and flux traps can be used to irradiate NpO_2 at ATR

High Flux Isotope Reactor (HFIR)



Reflector can be used to irradiate NpO_2 in the HFIR

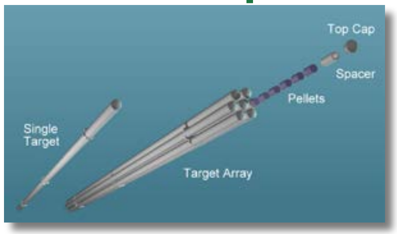
Both Reactors Produce Radioisotopes for DOE

2012-015 RMW

INL has Installed a Neptunium Oxide Repackaging Glovebox

- Installation is complete
- The first shipment anticipated in mid to late FY 2015

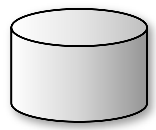
Target Design and Irradiation Focused on Development of Full Length Target Design



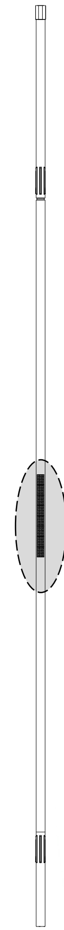
Tensile Strength

Do interactions occur between clad and pellets that reduce clad strength?

Single Pellet Targets



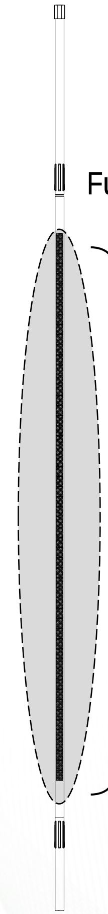
Targets must not fail due to melting or cladding breach.



Partially Loaded Targets

What are the impacts of flux depression on product quality?
What is the fission gas release?

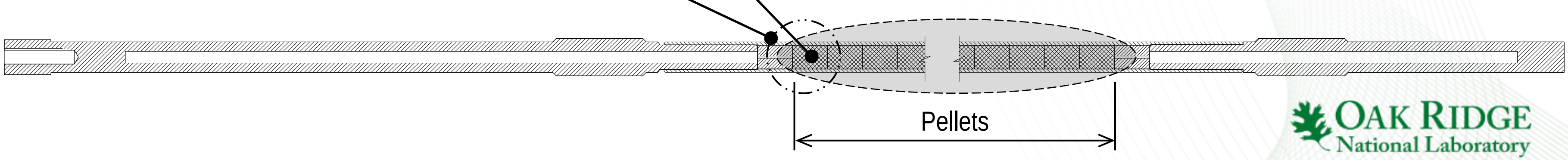
Partial ~ 3”



Fully Loaded Targets

What are the yields from a full length target?

Full ~ 20”



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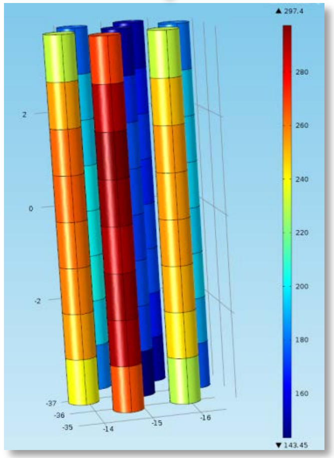
2012-037 RMW

Target Irradiation Has Been Scaled Up By >100X

Starting with NpO_2

Single pellets were irradiated in FY2012 (~ 0.6 g NpO_2)



Multi pellet test targets were irradiated and analyzed

Leading to fully loaded test targets

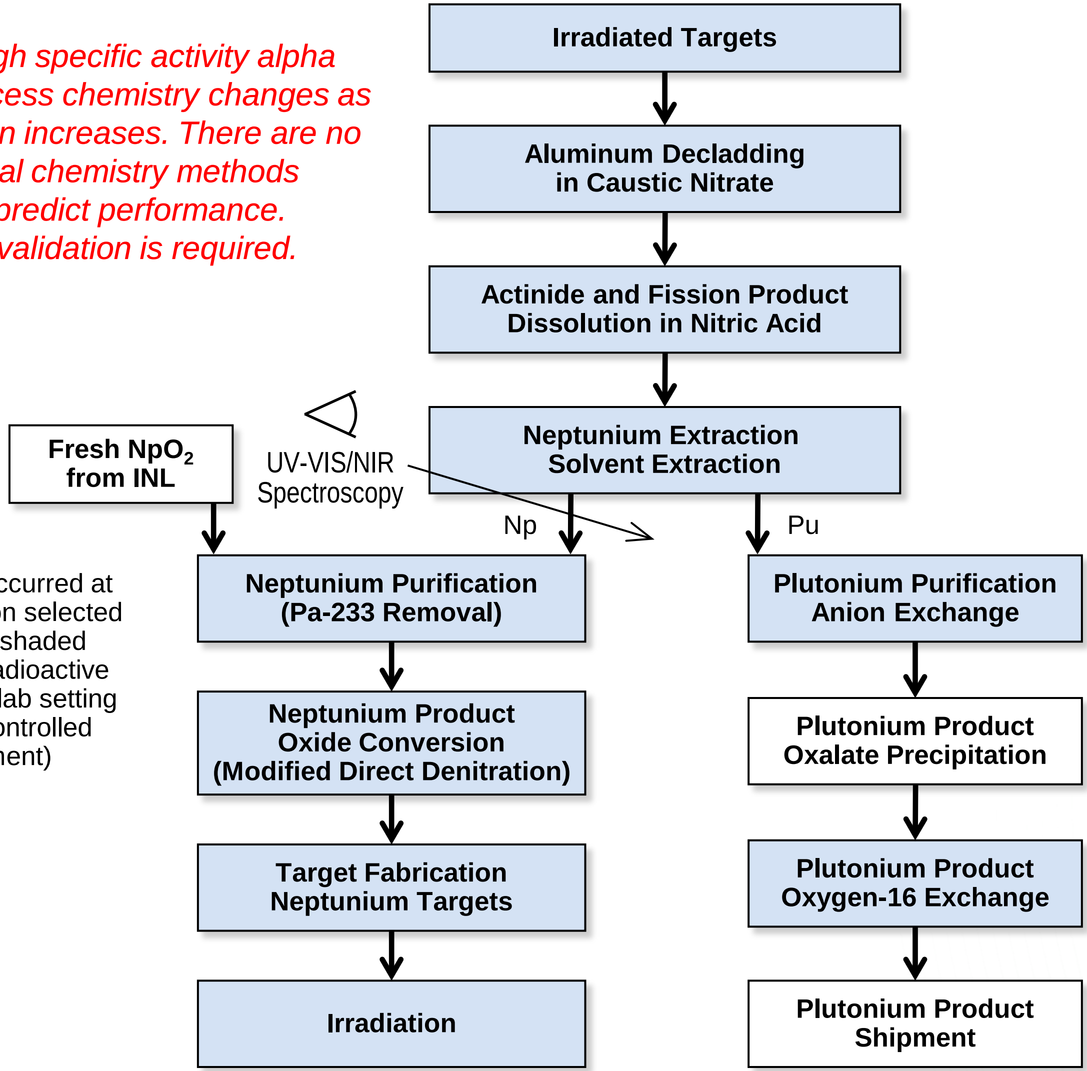
About 1100 g of NpO_2 will have been irradiated at the conclusion of the November irradiation cycle

Several Chemical Separations Are Needed for Efficient Plutonium and Neptunium Recovery

- Separate Pu and Np from fission products
  - Maximize recovery
- Recover Pu-238 product
  - Meet LANL product specs
- Recover Np-237 for recycle
  - Low fission product content for use in shielded gloveboxes
  - Pu-238 content <300 ppm
- Approach:
  - Solvent extraction for first-cycle separations
  - Purification of Np and Pu products by anion exchange or second-cycle solvent extraction

Process Chemistry of Np, ^{238}Pu needs to be Developed and Demonstrated to Ensure Delivery of 1.5 kg/year

^{238}Pu is a high specific activity alpha emitter. Process chemistry changes as concentration increases. There are no computational chemistry methods available to predict performance. Testing and validation is required.



Testing has occurred at a small scale on selected processes (shaded boxes) with radioactive materials in a lab setting (e.g., well controlled environment)

Integrated end-to-end testing (with the exception of ^{16}O exchange) is scheduled to start summer 2015 with irradiated targets.

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2014-025 RMW

Current Tasks Focus on Chemical Processing to Recover Np/Pu

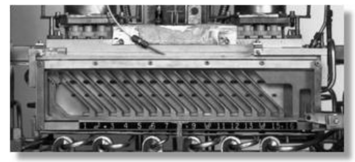
Dissolution

Target

Actinides in nitric acid solution

Can we dissolve with existing equipment?

Partitioning

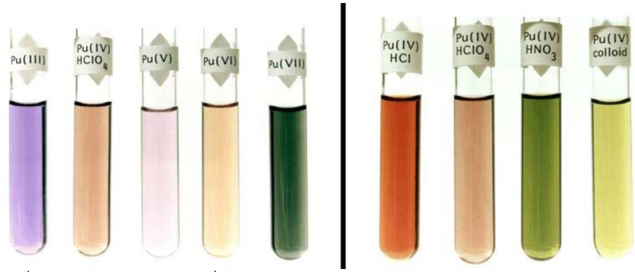


Np

Pu

Fission Products

Can we partition into components efficiently?



Pu Valence

Purity

How ORNL ensures that LANL can use new ^{238}Pu in their existing process line (product purity, neutron emission rate)

How do we recycle Np into additional targets? (decontamination from Pu, Fission products)

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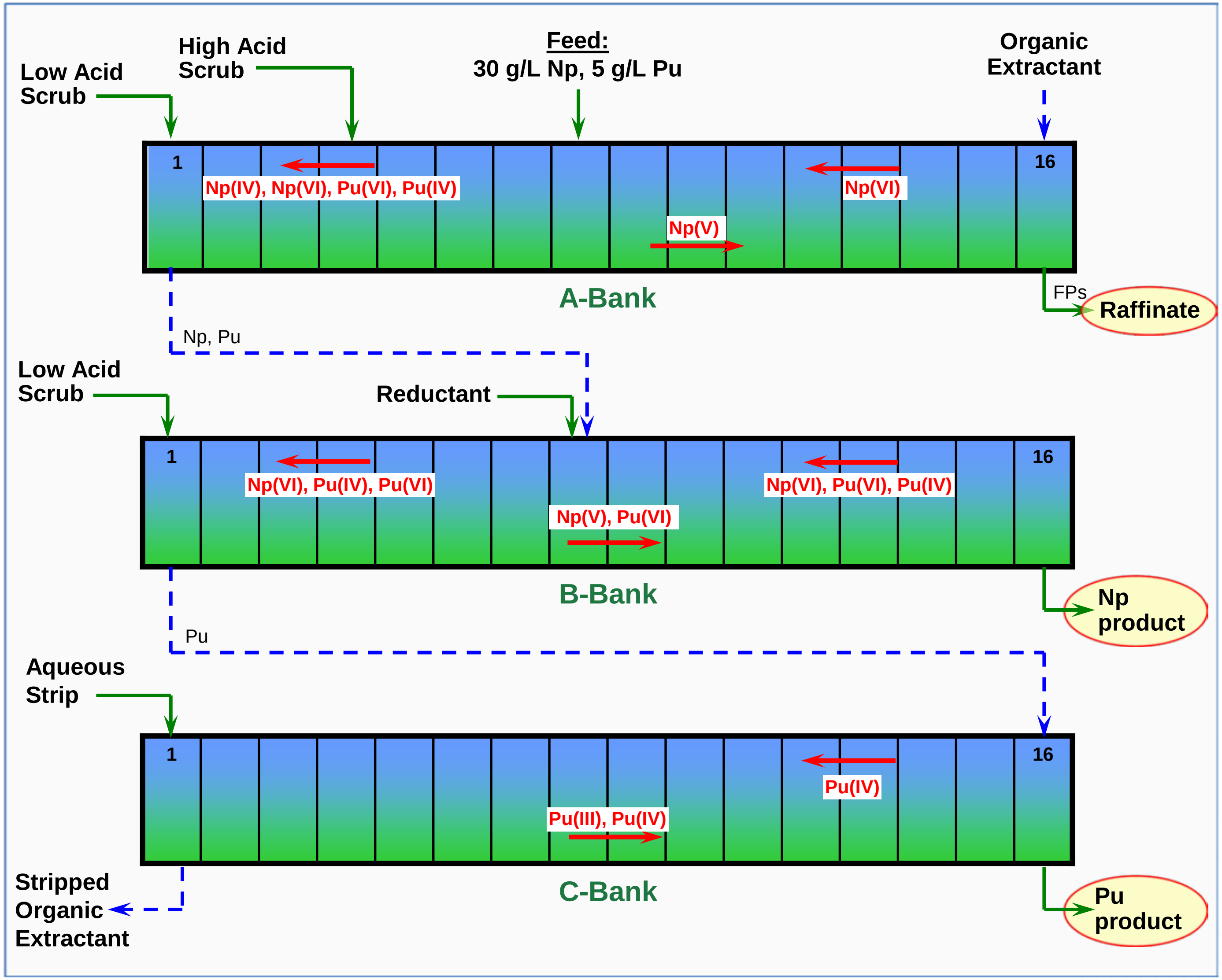
2014-005 RMW

Co-extraction and Partitioning was Tested to Evaluate Pu and Np Recovery



1. Coextraction

- Remove FPs
- Oxidize Np(V)
- Recover Np and Pu

2. Partitioning

- Reduce Np and strip in aqueous phase
- Retain Pu in organic phase

3. Stripping

- Reduce Pu and recover in aqueous phase

Good Progress Has Been Made In Solvent Extraction Testing

- Excellent recovery of Pu and Np in coextraction
  - Oxidation works with high Np concentration and presence of Pu-238
  - Model predicts oxidation performance reasonably well
- Good partitioning performance with sufficient nitrite
  - Np product has low Pu concentration (20-35 ppm)
  - Up to 97% recovery of Np in first-cycle Np product
- There is opportunity for improvement of kinetic models for Np reduction (a significant amount of Np went with the Pu product)
- Next testing will focus on
  - Fission-product decontamination factors
  - Second-cycle purification of Np product

There Have Been Several Significant Accomplishments Since the Project Started

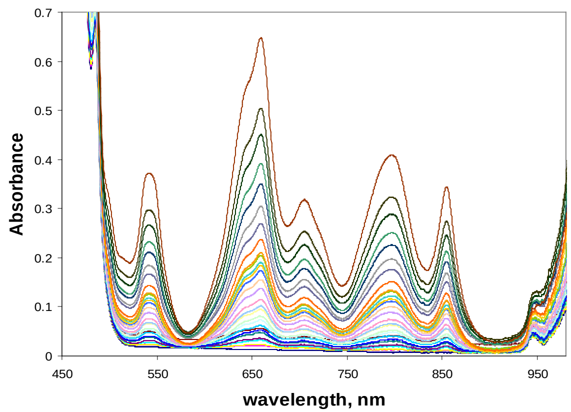
- Neptunium oxide repackaging capability for shipping containers to ORNL has been obtained at INL.
- Target design and fabrication have been scaled up to prototypical targets; approximately 50 targets have been irradiated to date.
- Chemical processing tests using approximately 500 gm Np/Pu at concentrations expected for production are underway to develop improved methods for separation and recovery of both neptunium and plutonium.

Acknowledgements

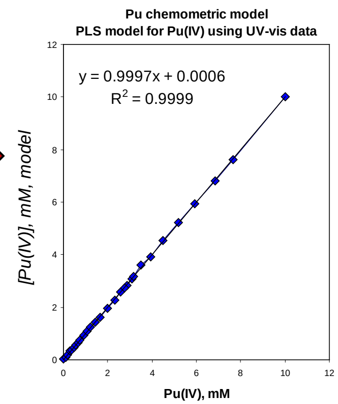
- NASA Planetary Sciences Directorate
- Dozens of ORNL employees in eight ORNL Research Divisions
- US DOE Office of Space and Defense Power Systems, NE-75

Methodology for Online Process Monitor Development

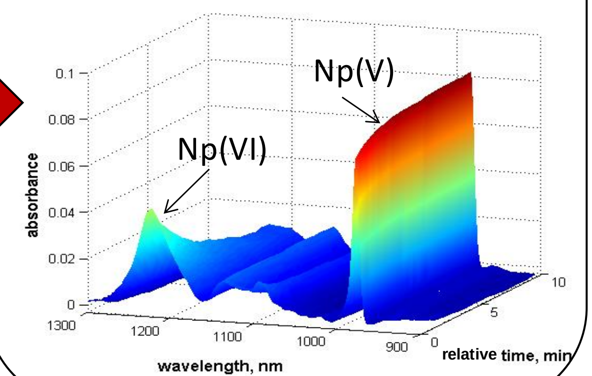
Static measurements: Model training database



Chemometric model development



On-line model verification and translation



Integrated software for data collection, processing, storage and archiving

Real-time on-line concentration data display

Bryan et al. 2012.

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