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National Nuclear Security Administration
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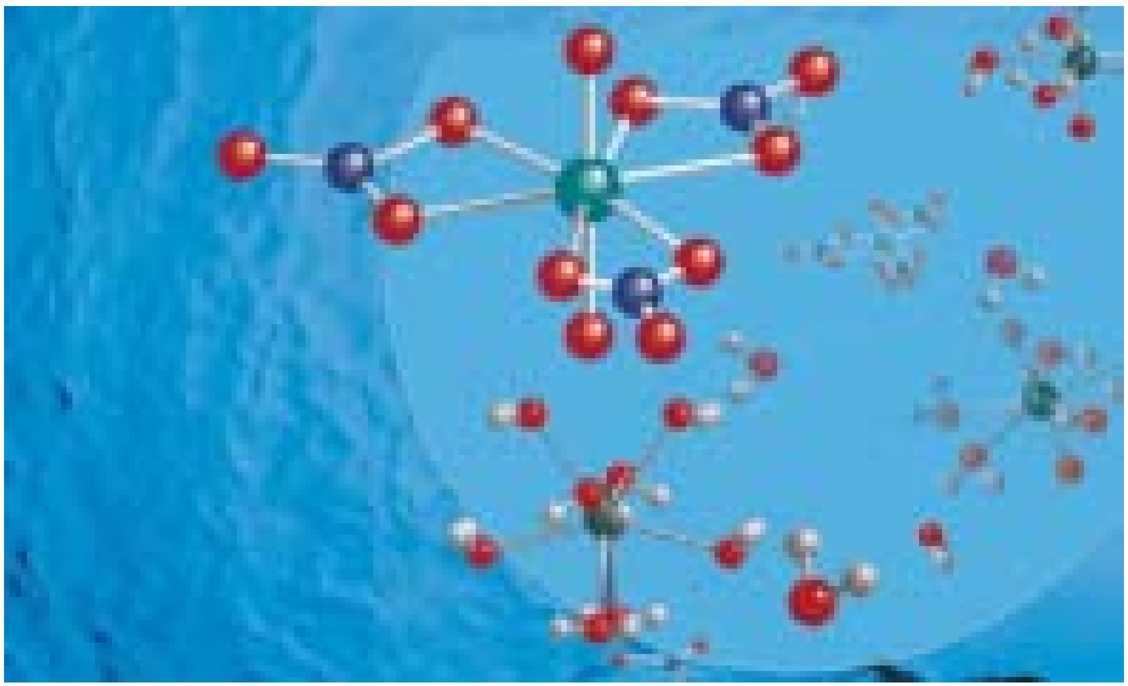
LA-UR-17-22053

Experimental Design to Study Criticality Effects of Plutonium Aging

IER-301



Theresa Cutler, Travis Grove, David Hayes, Mark Mitchell, Bill Myers, Rene Sanchez, Jessie Walker, Bob Margevicius

NCSP Technical Program Review

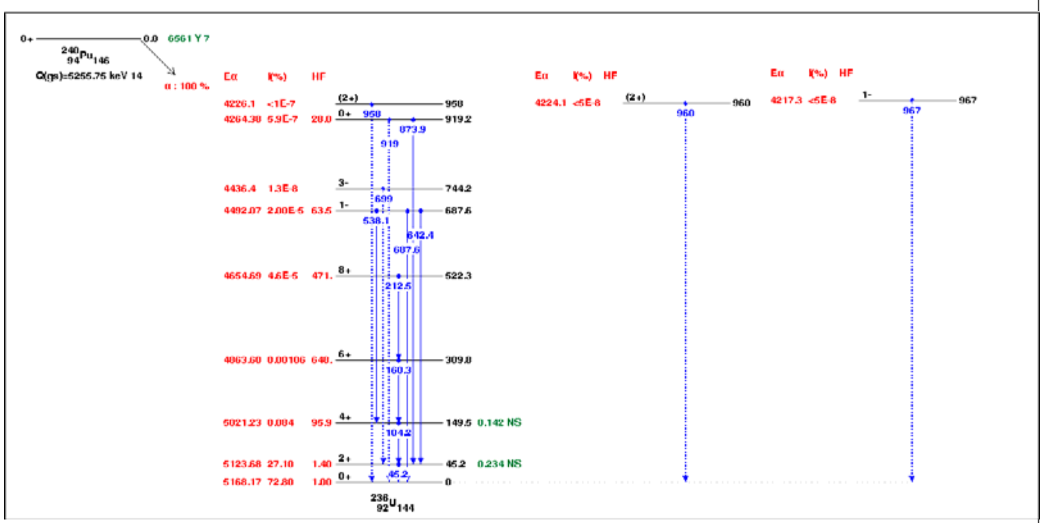
March 2017

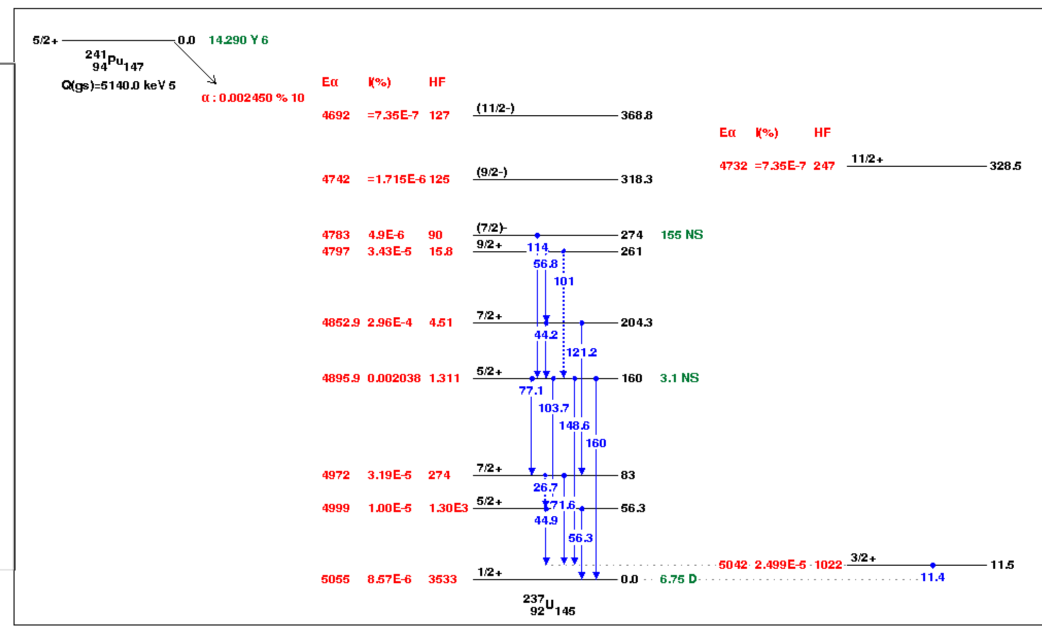
Overview

- Experimental Objectives
- Background and Previous Work
- Preliminary Design
- Final Design

Experiment Objectives

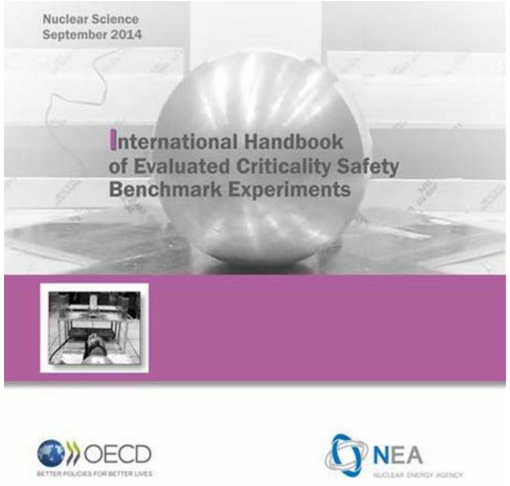
- **Examine the effects of plutonium aging on criticality**
  - It is known that the isotopic composition of plutonium changes with age, due to radioactive decay
    - Pu-241 → Am-241
  - He-3 ingrowth
- **The effects of criticality have yet to be examined in a systematic approach**
- **NEED integral data**





Previous Work

- **Metallurgical Effects have been investigated over the past few decades**
  - Much information has been obtained
- **He-3 atoms formed from alpha decay**
  - These atoms migrate toward each other forming sub-micron-size bubbles
  - Become effective “hardening agents” in the plutonium
  - Well documented in the last two decades
- **Accounted for in benchmark evaluations**
  - ZPPR critical benchmark experiments (1969→1980)
  - BeRP ball fundamental physics benchmark experiments (1980→2009)

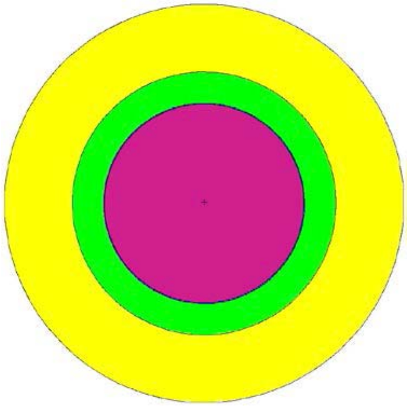


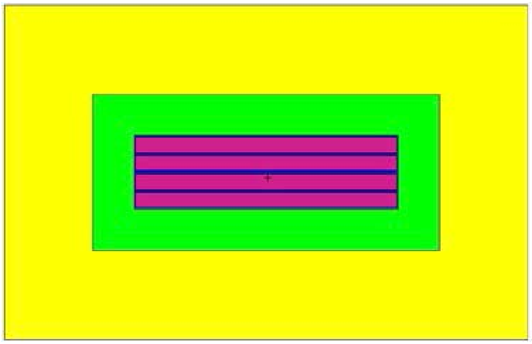
Preliminary Design

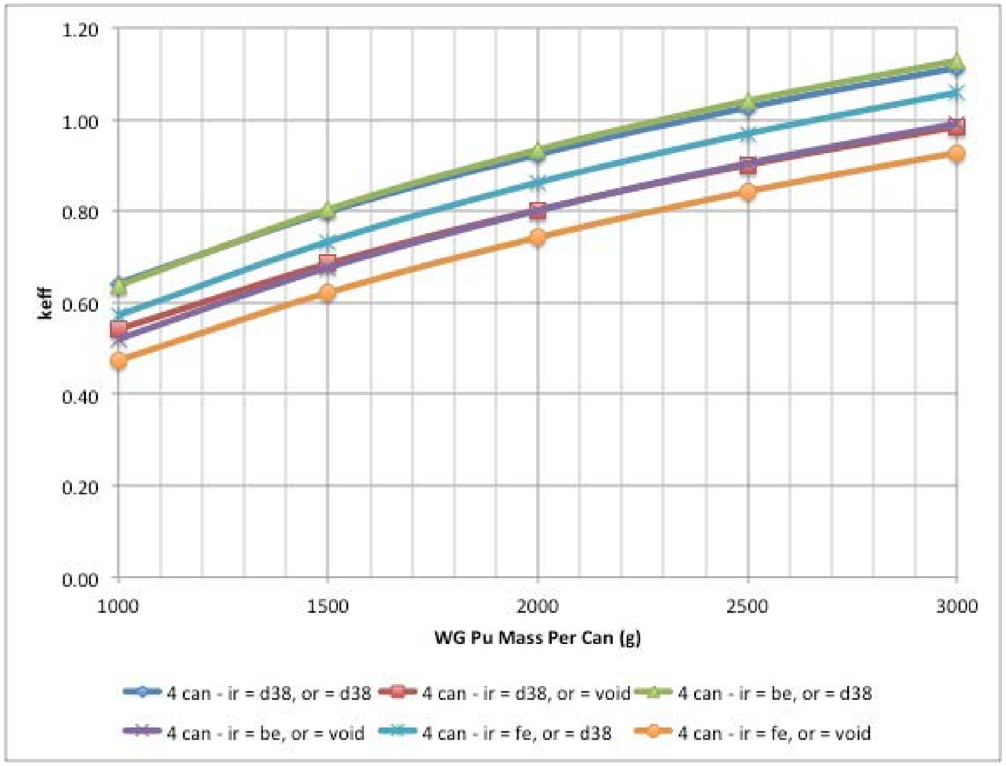
- **Design an integral experiment to compare new and aged plutonium**
- **Two approaches considered**
  - **Approach 1**: Design and build a completely new set of critical experiment parts to perform the comparison
  - **Approach 2**: Utilize currently available critical parts to perform small sample reactivity worth comparisons
  - Both options utilize the existing critical assemblies at NCERC

Preliminary Design: Approach 1

- **Manufacture 6" OD discs, matching current NCERC inventory and available reflectors (Be, DU, Fe, …)**
- **2 sets**
  - One with "new" isotopics
  - One with "aged" isotopics
- **All cladding would be of the same material type and thickness**





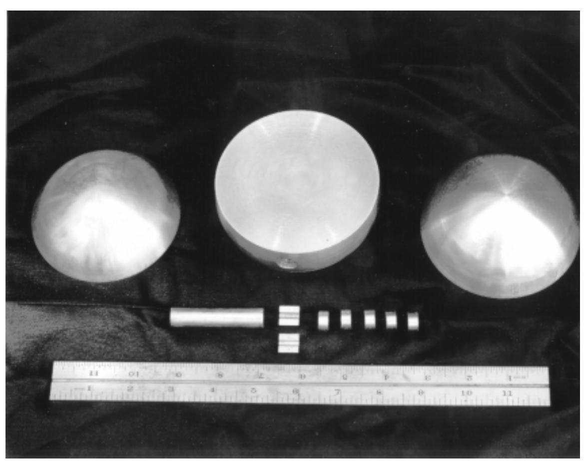


Preliminary Design: Approach 2

- **Utilize currently available critical parts to perform small sample reactivity worth comparisons**
  - Flattop glory hole pieces
  - BeRP ball
  - Thor core pieces
- **Used by R. Sanchez* to estimate the critical mass of Np-237 using Flattop**

BeRP ball

Flattop

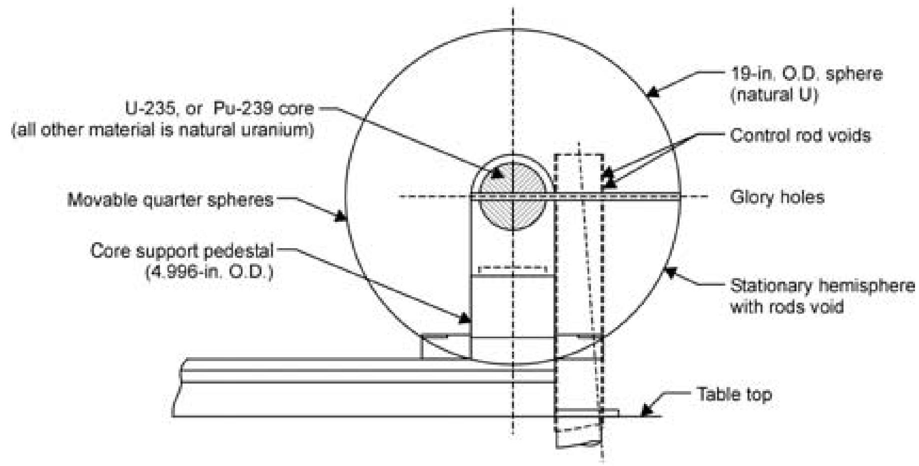


Thor

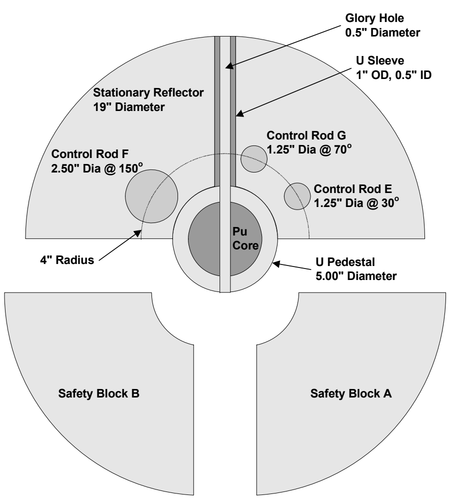
***SPEC-MET-FAST-003**, *Neptunium-237 and Highly Enriched Uranium Replacement Measurements Performed Using Flattop,* 1999-09-30.

Final Design

- **Small Sample glory hole replacement measurements in Flattop**
  - Chosen through systematic evaluation in MCNP and feasibility
  - Future work will likely build upon this with a new full Flattop Pu core
- **Use similar protocal for determining reactivity worth and critical mass to Sanchez Np-237 work in 1990s using the Flattop core**
- **Considered both HEU and Pu core**
  - Pu core selected due to higher worth of glory hole pieces



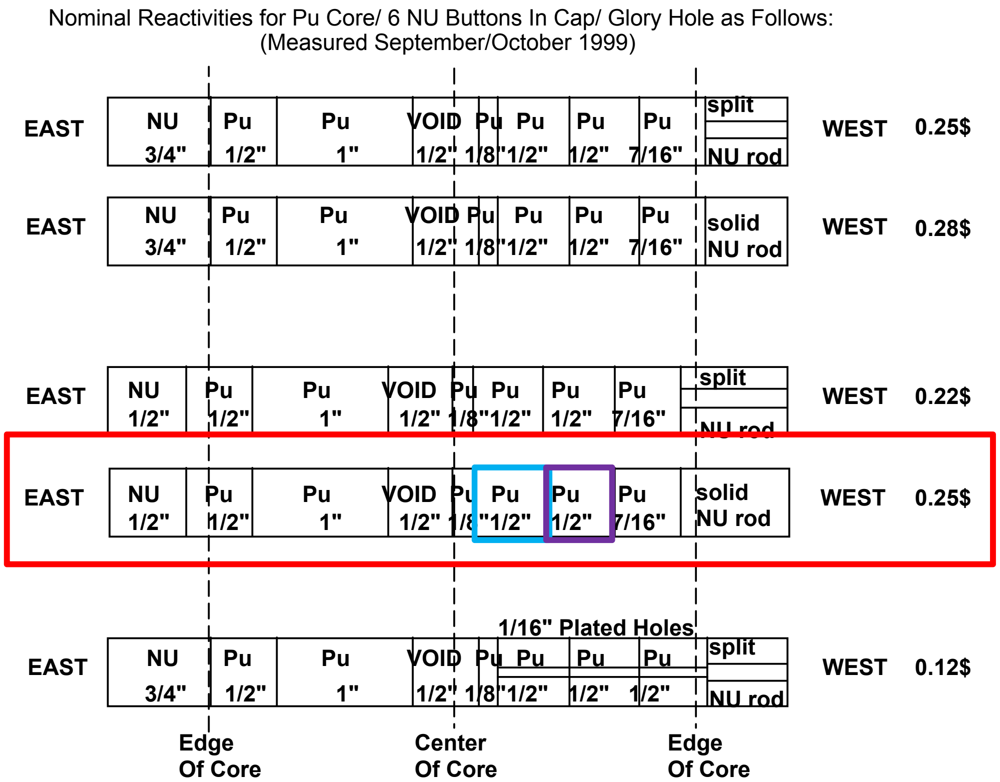
Elevation View



Pu Core: 3.57" Diameter
U Core: 4.77" Diameter
Rod Diameters are 13 mils less than the shown hole sizes

Final Design: Overview

- Glory hole loading with "new" and "old" Pu
- The Flattop Pu core and glory hole pieces were manufactured in ~1957 at Los Alamos
- Diameter of Flattop Pu core glory hole is 0.5". All pieces used fit tightly
- New pieces, 0.5" OD x 0.5" L, will be manufactured at Los Alamos
- New pieces will be included in the configuration and compared to reactivity results from old configuration
- Locations selected based on highest reactivity worth



Glory Hole Loadings and Associated Reactivity for Flat-top Pu core

Final Design: Isotopics

- **Manufactured in ~1957, δ-phase plutonium, stabilized in gallium**
- **Approximate density: 15.83 g/cm^3**
- **5 mil thick nickel cladding on all pieces**
  - New pieces will have 10 mil thick Invar cladding
- **Computationally decayed, using MISC, to 2016**
- **No burnup assumed**
- **No density change effects considered in decay, although acknowledged it decreased slightly from He-3 ingrowth**

Initial Isotopic Composition

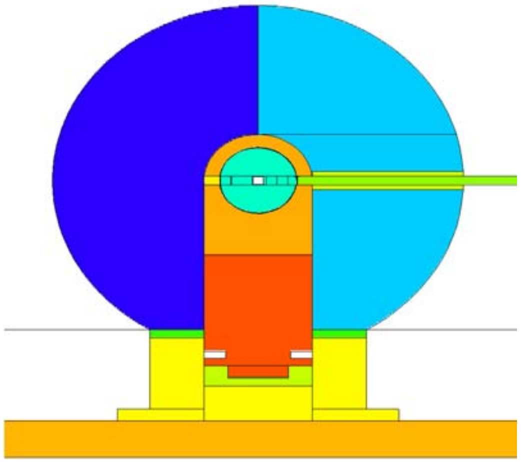
Nuclide	Weight Percent
^{239}Pu	93.8
^{240}Pu	4.8
^{241}Pu	0.3
^{69}Ga	0.6611
^{71}Ga	0.4389

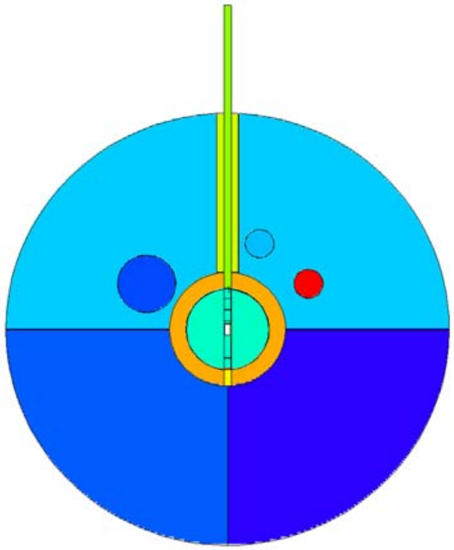
Approximate 2016 Decayed Isotopic Composition

Nuclide	Weight Fraction
^{69}Ga	6.611E-03
^{71}Ga	4.389E-03
^{207}Pb	4.889E-15
^{227}Ac	1.105E-14
^{229}Th	8.384E-14
^{231}Th	6.140E-15
^{232}Th	2.352E-10
^{231}Pa	4.165E-11
^{233}Pa	5.807E-12
^{233}U	1.237E-09
^{235}U	1.510E-03
^{236}U	2.834E-04
^{237}U	5.895E-12
^{237}Np	1.719E-04
^{239}Pu	9.365E-01
^{240}Pu	4.771E-02
^{241}Pu	1.890E-04
^{241}Am	2.636E-03

Final Design: MCNP®6 Simulations

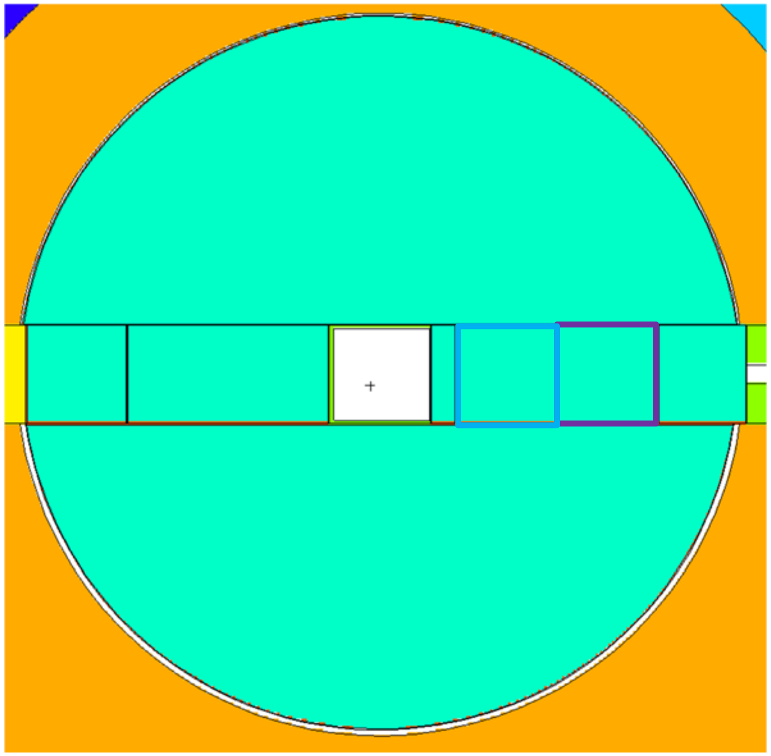
- **Detailed Flattop model for HEU core, adapted to the Pu core**
- **Pu core based on detailed engineering drawings**
  - Different than those used in previous benchmarks with Flattop
- **All blues, orange, and reds represent natural uranium; teal represents plutonium.**





Final Design: MCNP®6 Simulations

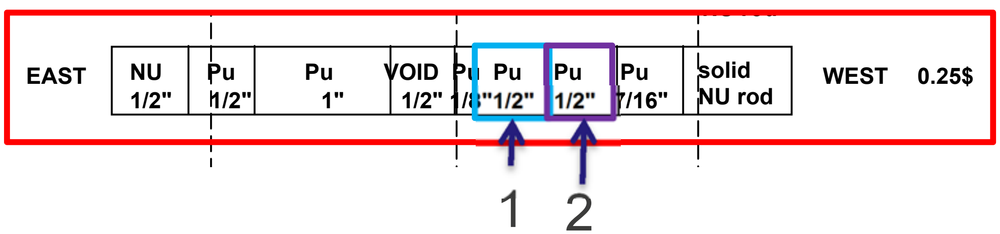
- Close up view of Pu core with the proposed glory hole loading
- Blue and purple boxes represent the pieces that will be replaced with new ones
- All blues, orange, and reds represent natural uranium; teal represents plutonium.
- HEU and blank pieces also considered in the same locations



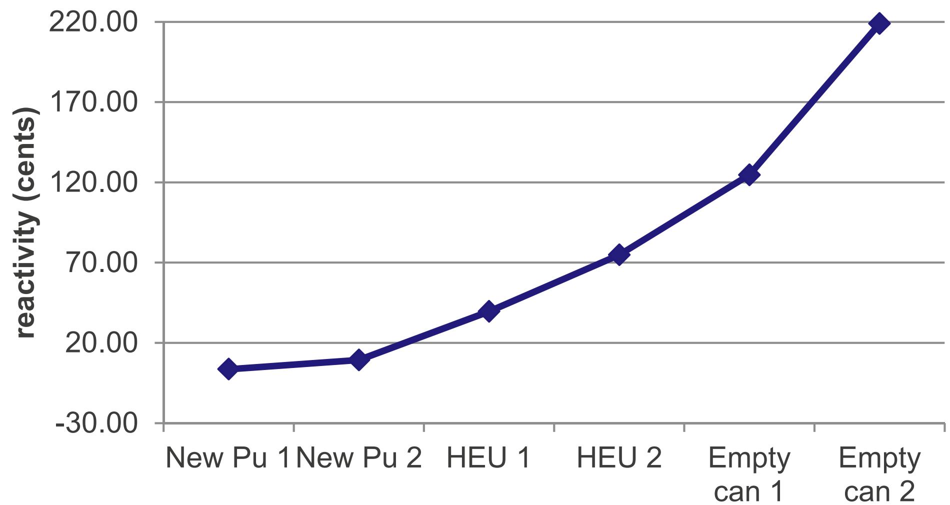
Final Design: MCNP®6 Simulations

MCNP6 keff and Reactivity Results for Expected Replacement Measurements

configuration	keff	delta_keff	reactivity (cents)	delta reactivity (cents)
Base (all old Pu)	1.00269	0	96.85	
New Pu 1	1.00259	0.00010	93.26	3.59
New Pu 2	1.00243	0.00026	87.51	9.34
HEU 1	1.00159	0.00110	57.31	39.54
HEU 2	1.00061	0.00208	22.01	74.84
Empty can 1	0.99923	0.00346	-27.82	124.67
Empty can 2	0.99663	0.00606	-122.07	218.92

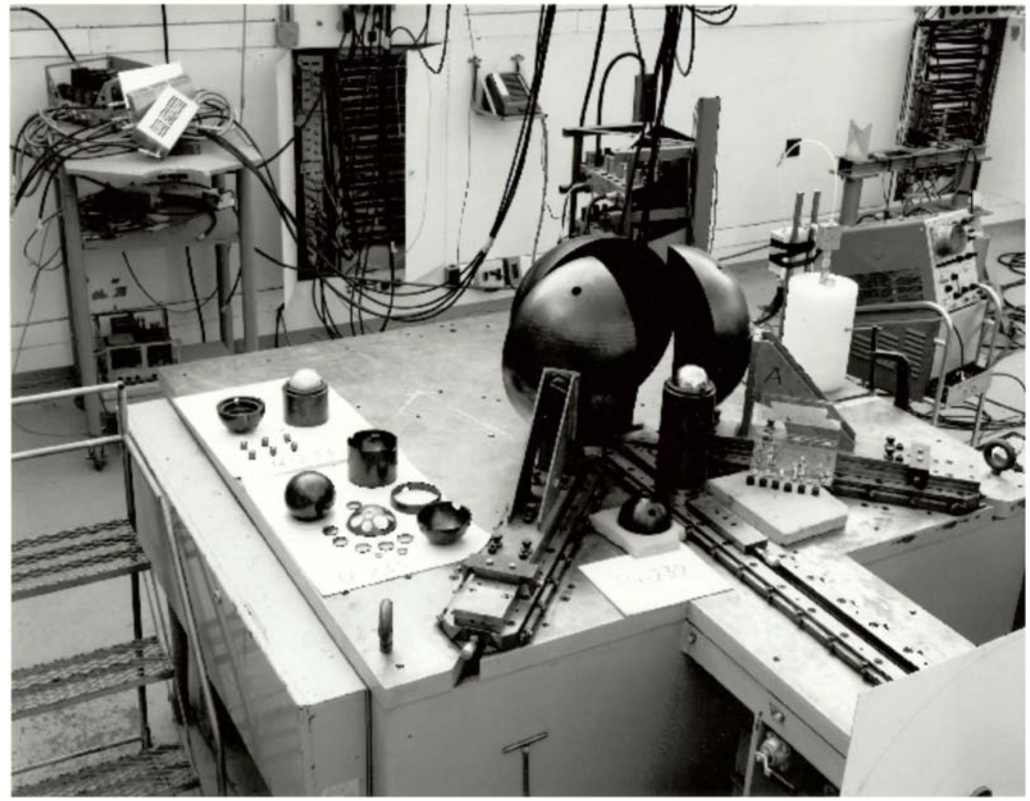


Delta Reactivity (cents) for All Considered Replacement Measurements



Summary

- Knowledge gaps exist on criticality behavior of plutonium as it ages
- An integral experiment has been designed to measure the reactivity worth of new and 50+ year old plutonium
- Experiment based on proven concept with Np-237 in Flattop



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