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Title: Radiological Considerations in Using Pu-242 as a Substitute for Pu-239

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Radiological Considerations in Using Pu-242 as a Substitute for Pu-239

Jenelle Mann

Radiological Engineer

Managed by Triad National Security, LLC for the U.S. Department of Energy's NNSA

Motivation

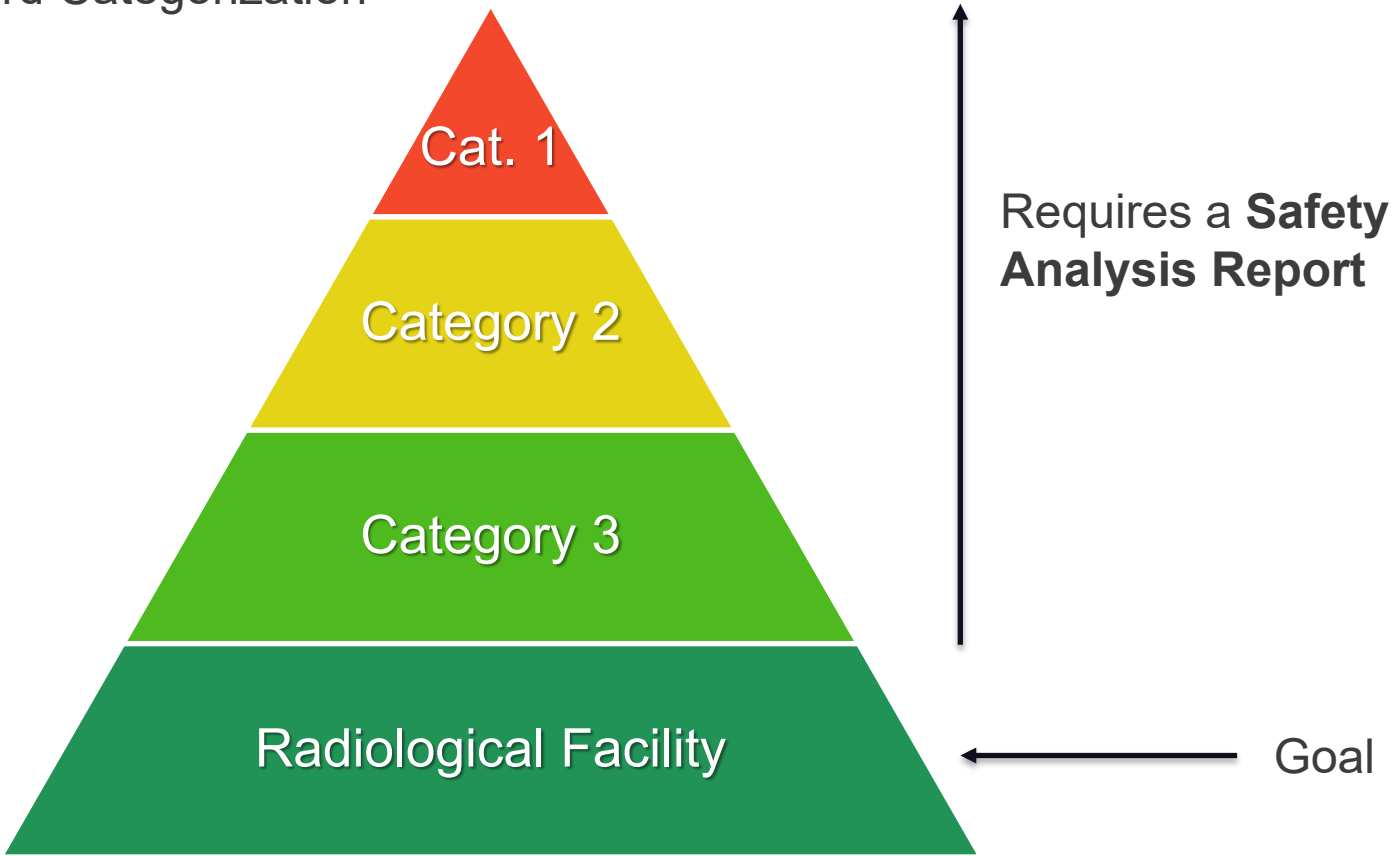
- Laboratory retrofit
  - Replace current fumehoods and install new gloveboxes
  - Primary operations
    - Recovery of uranium from a solution to a stable oxide
    - Extraction of **plutonium** from graphite and/or salt targets
      - Pu-242 specifically

- Majority of the worked performed at LANL (and other national laboratories) is performed with Pu-238 and Pu-239

- White Paper on the Differences between the Radiological Hazards of Pu-239 and Pu-242
  - Luke Hetrick, Gerald Murrell

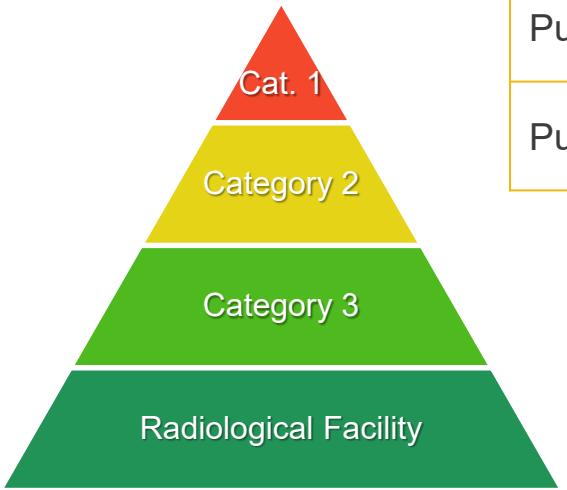
Why Pu-242?

- NNSA Hazard Categorization



Why Pu-242?

- NNSA Hazard Categorization



Radioisotope	Radiological Facility	Category 3 Facility	Category 2 Facility
Pu-238	< 1.53E-01 g	1.53E-01 g to 1.03E+01 g	> 1.03E+01 g
Pu-239	< 3.86E+01 g	3.86E+01 g to 2.61E+03 g	> 2.61E+03 g
Pu-242	< 6.49E+02 g	6.49E+02 g to 2.29E+04 g	> 2.29E+04 g

Discussed Considerations

- Background
- Decay Scheme
- External Dose
  - Shielding Consideration
- Internal Dose
  - Inhalation
  - Ingestion
- Categorization Revisited
- Ventilation
- Criticality
- Detection (HPAL)
- Internal Dosimetry

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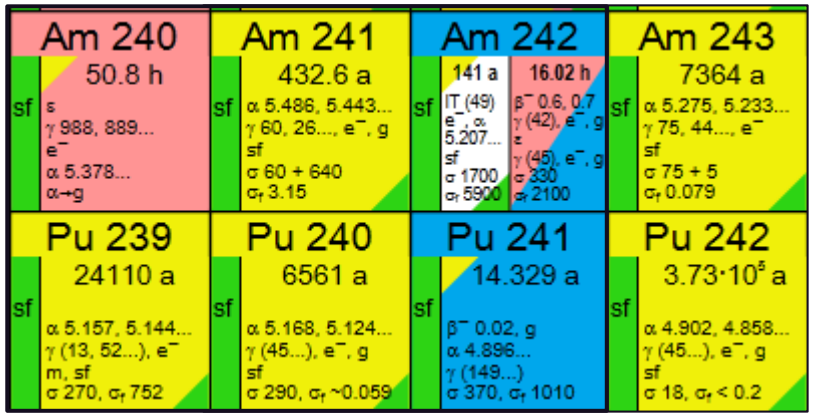
Example Plutonium Uses at LANL

- Proton Radiography (PRad) at LANSCE **Pu-239**
- Heat Sources for NASA **Pu-238**
  - Aqueous recovery
  - Fuel fabrication
  - Small item fabrication
  - Inspection of heat sources
- Computer Numerical Controlled (CNC) Machining and Lathe **Pu-239**
- Radiography using a Linatron **Pu-239**

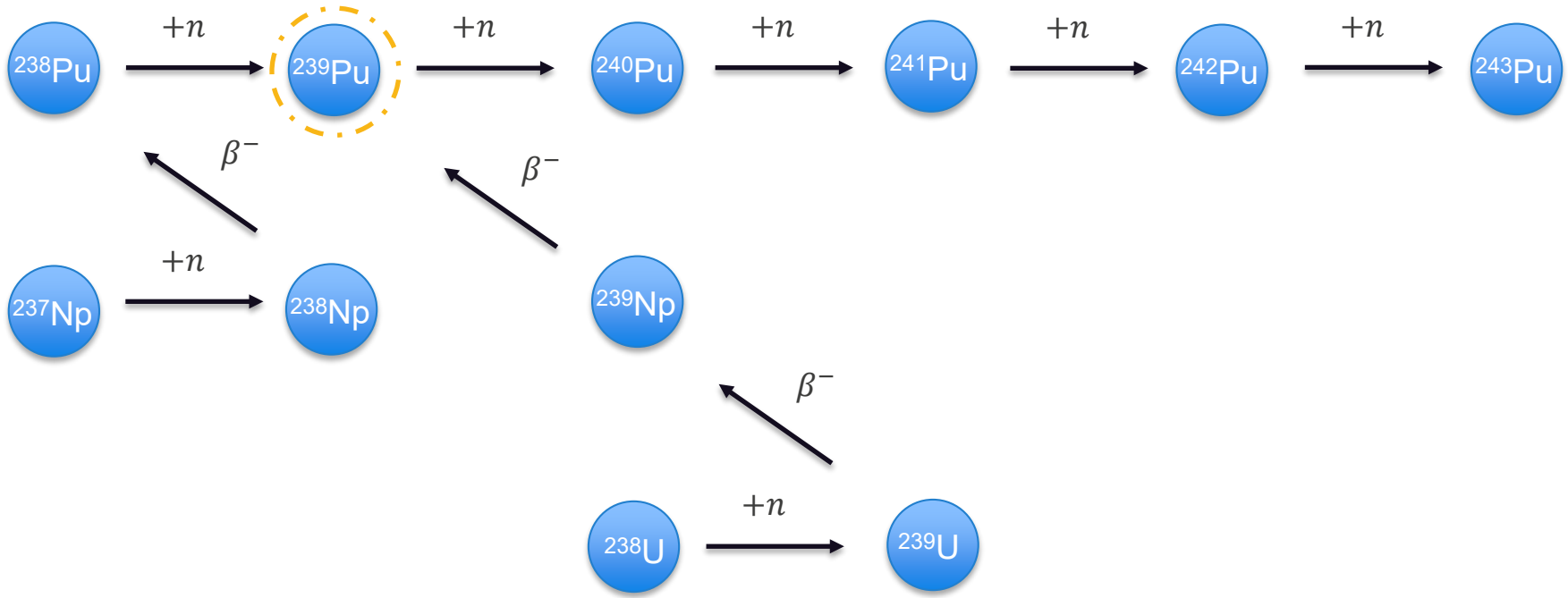
Background

	Half Life (yrs)	Specific Activity (Ci/g)	Decay Type
Pu-238	87.74	1.7×10^{1}	α, β, γ
Pu-239	24119	6.2×10^{-2}	α, β, γ
Pu-242	376300	3.9×10^{-3}	α, β, γ, SF

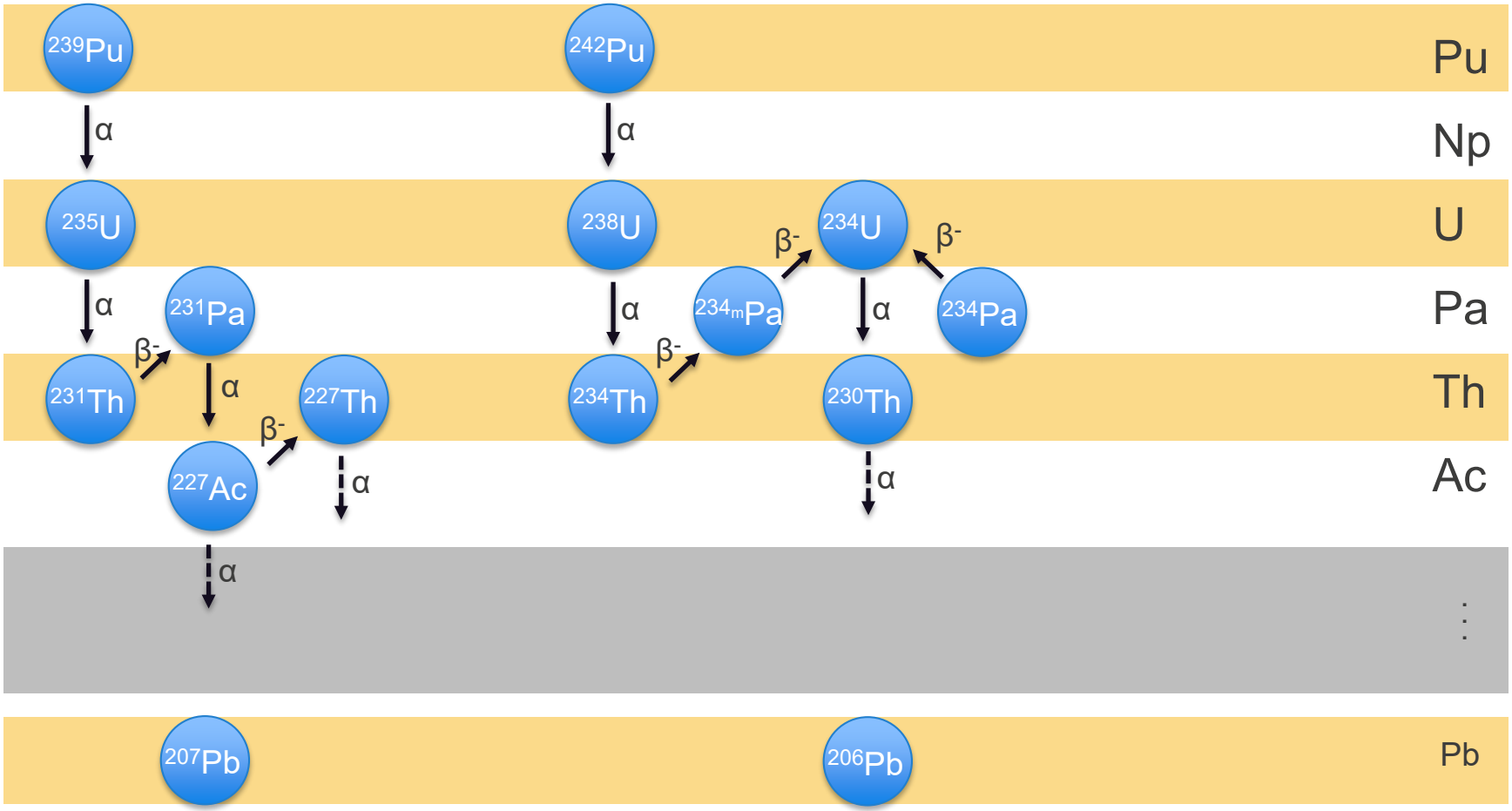


Background

- Production of Pu-239 and Pu-242



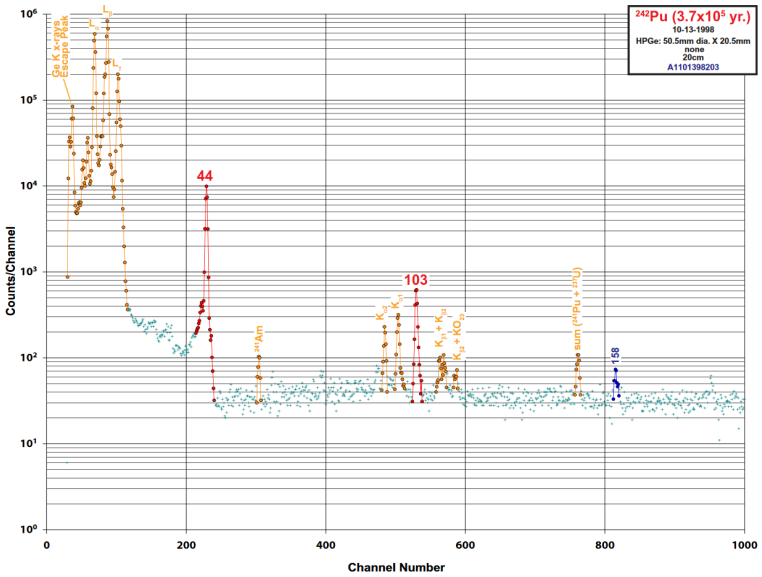
Decay Scheme

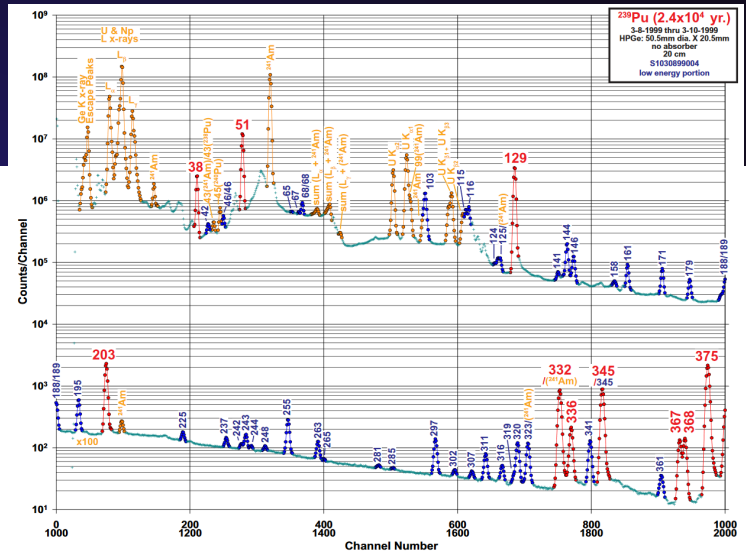


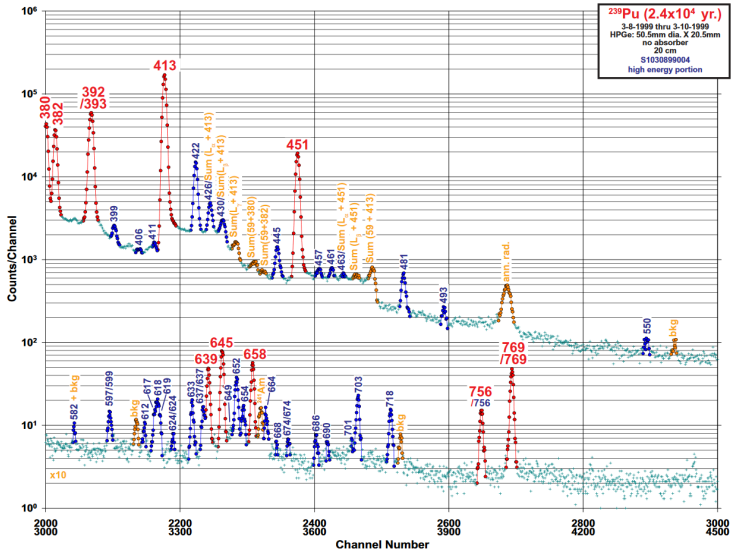
External Dose – Photons

- Emission Rate - OrigenARP

Photon Emission		
Pu-239	8.25E+07 γ/s-g	1.33E+09 γ/s-Ci
Pu-242	9.97E+06 γ/s-g	2.56E+09 γ/s-CI







External Dose – Photons

	Point Source Air Kerma Coefficient (rad-m^2/Ci-hr)	Specific Gamma Ray Constant (rem/hr) at 1 m from 1 Ci
Pu-239	1.48E-02	3.014E-02
Pu-242	2.84E-02	6.231E-02

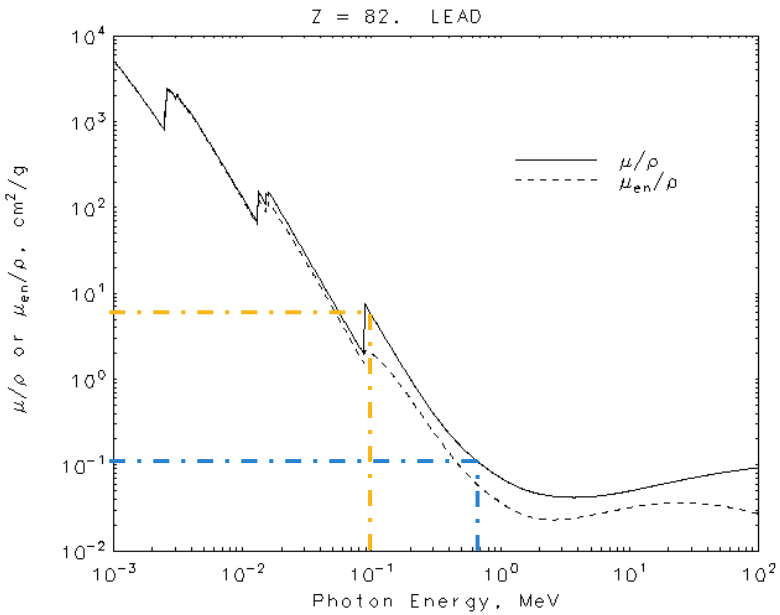
ICRP-107,
"Nuclear Decay Data for Dosimetric Calculations"

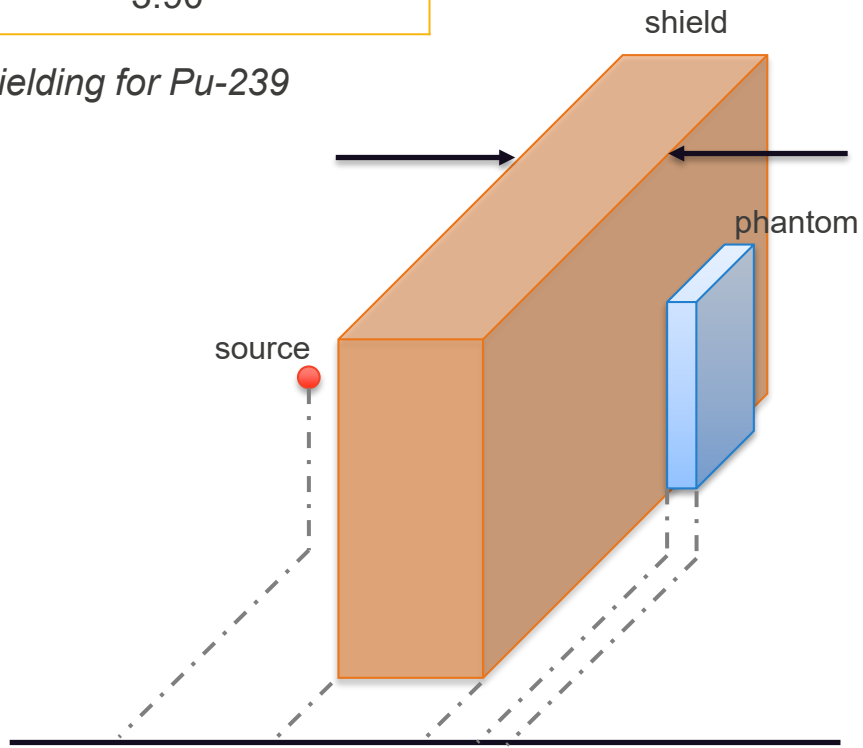
ORNL/RSIC-45, "Specific Gamma-Ray Constants for Nuclides Important to Dosimetry and Radiological Assessment"

External Dose – Photons, Shielding Considerations

Material	HVL (cm) for 103 keV	HVL (cm) for 769 keV
Lead	0.011	1.77
Concrete	0.543	3.90

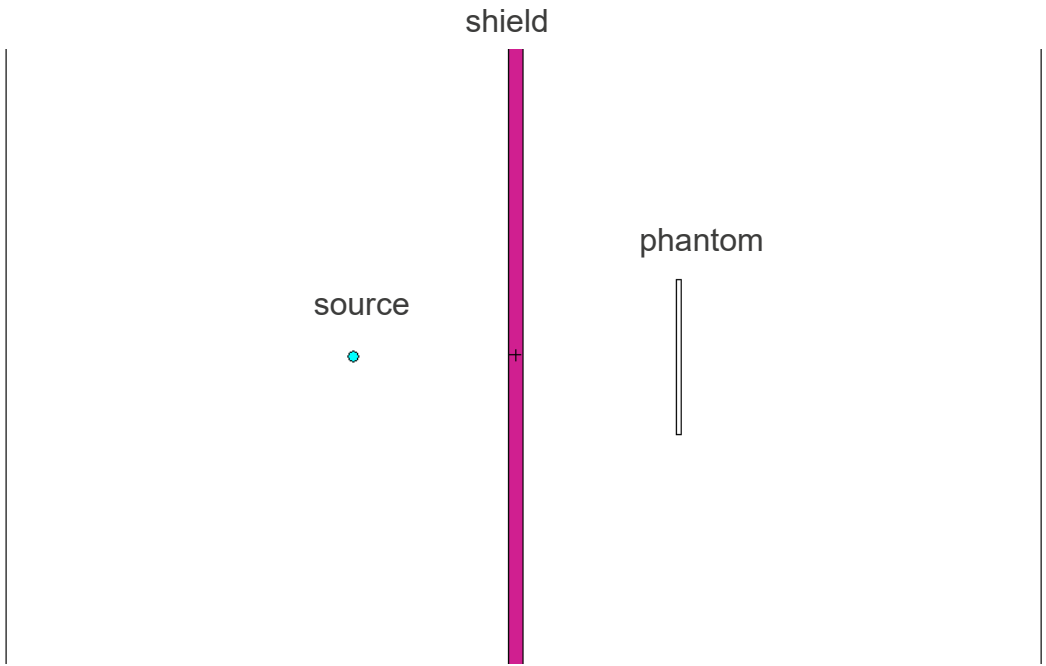
For same fluence rate, would need more shielding for Pu-239





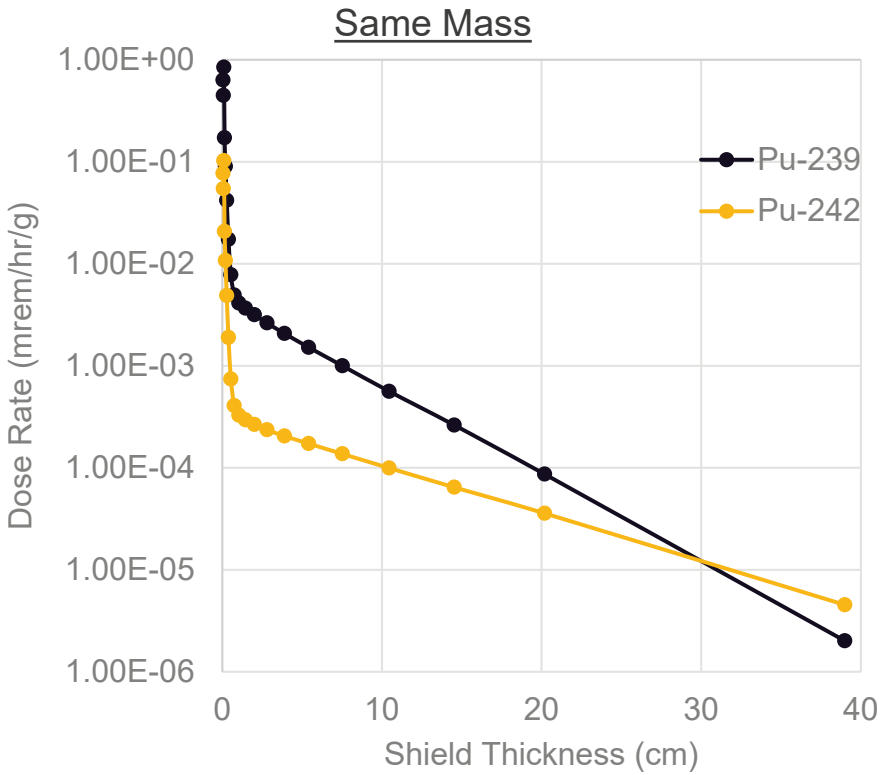
External Dose – Photons, Shielding Considerations

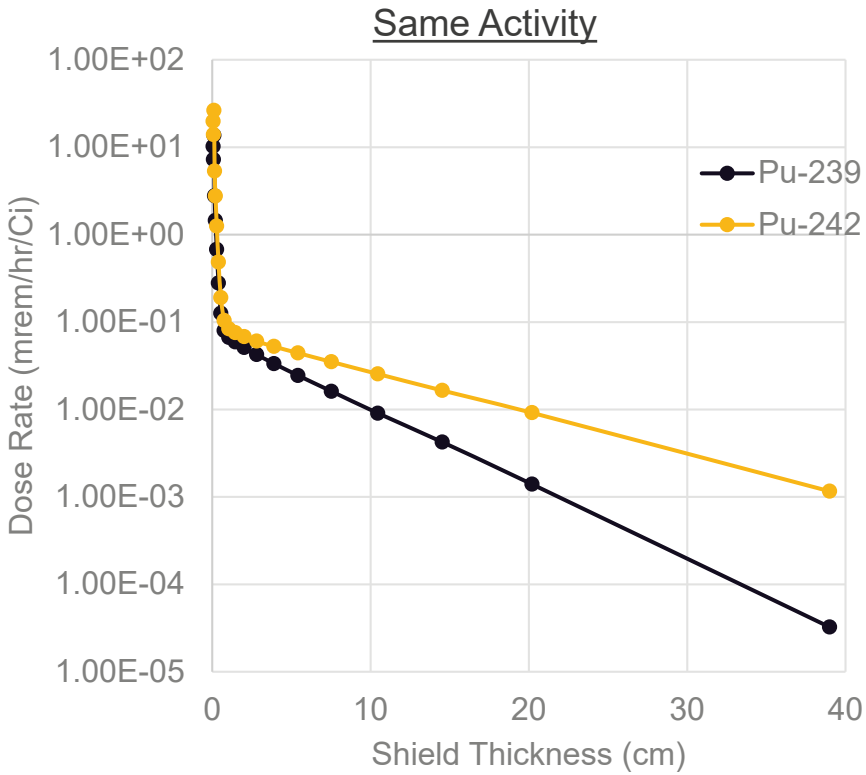
- Compared pure Pu-239 and Pu-242
  - Spectrum from OrigenArp
  - Modeled in MCNP6.2
- Dose to air phantom
  - Fluence to Dose Conversion Factors from ICRP-74
    - Photons: Table A.1
- Shield:
  - Concrete: 0.0543 cm – 39 cm



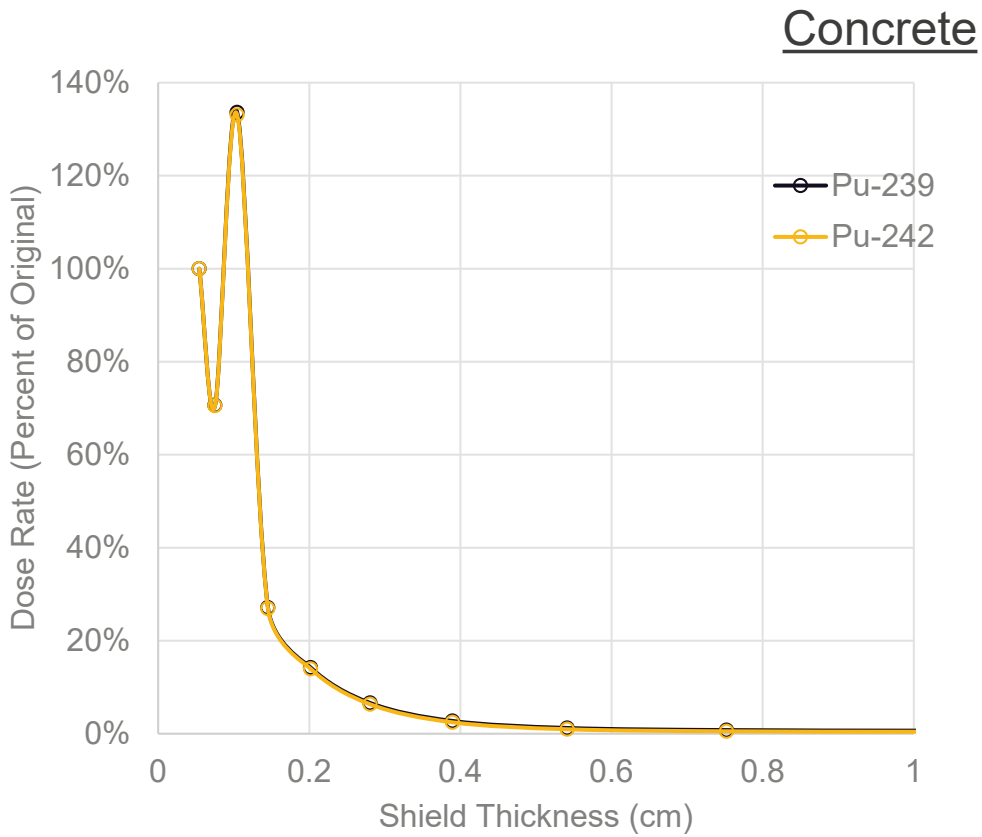
External Dose – Photons, Shielding Considerations

Concrete





External Dose – Photons, Shielding Considerations



Shield Thickness (cm)	Pu-239	Pu-242
0.0543	100.00%	100.00%
0.0752	70.62%	70.53%
0.105	133.59%	133.06%
0.145	27.14%	26.91%
0.202	14.22%	13.96%
0.280	6.61%	6.33%
0.390	2.74%	2.46%
0.541	1.23%	0.96%
0.752	0.78%	0.53%
1.05	0.65%	0.42%
1.45	0.58%	0.38%
2.02	0.50%	0.34%
2.81	0.42%	0.31%
3.90	0.33%	0.27%
5.42	0.24%	0.22%

External Dose – Neutrons

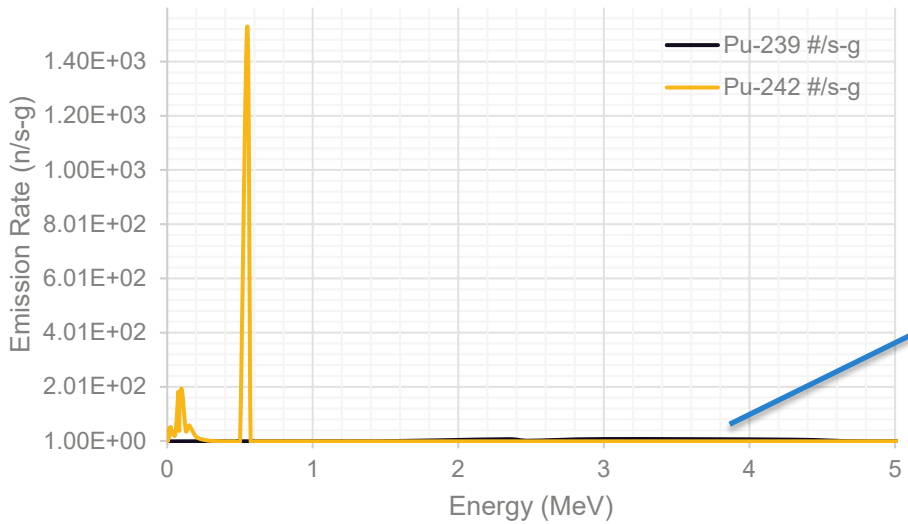
- Emission Rate - OrigenARP

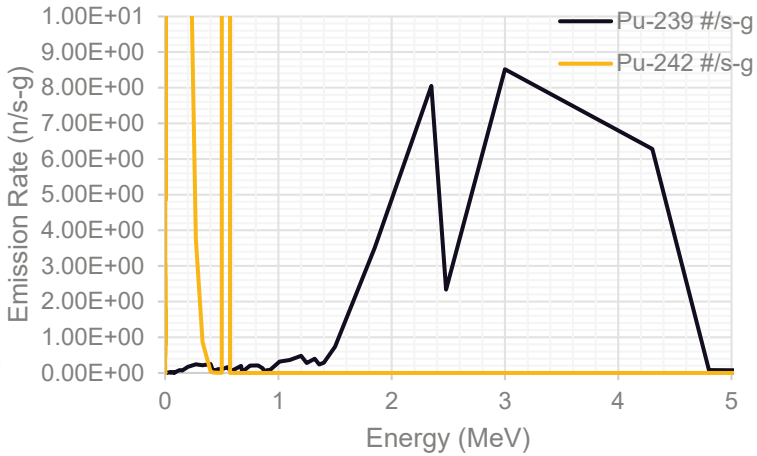
Neutron Emission		
Pu-239	3.47E+01 n/s-g	5.60E+02 n/s-Ci
Pu-242	1.53E+03 n/s-g	3.92E+05 n/s-Ci

Pu-239, SF branching ratio is 3E-10%

v.

Pu-242, SF branching ratio is 5.5E-4%



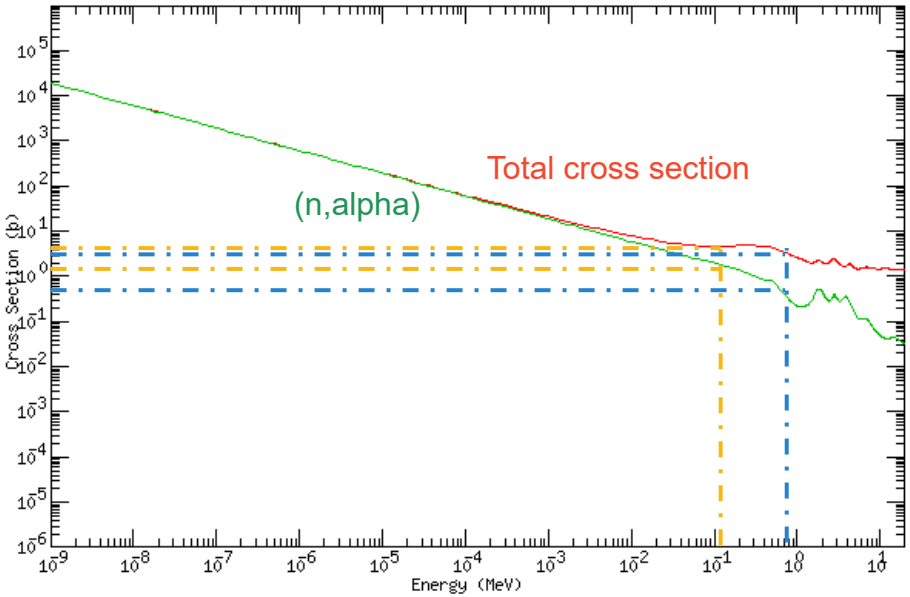


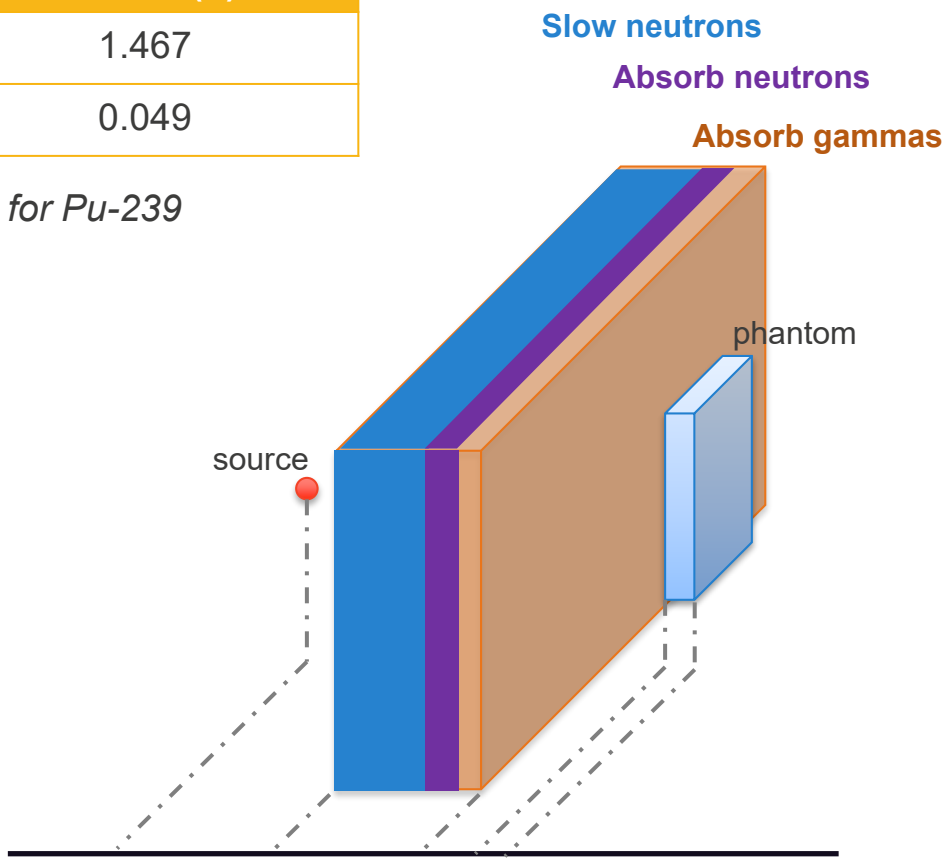
Pu-239 produces neutrons at higher energies

External Dose – Neutrons, Shielding Considerations

Cross Section Type	Thermal (b)	14 MeV (b)
Total	3840	1.467
(n,alpha)	3837	0.049

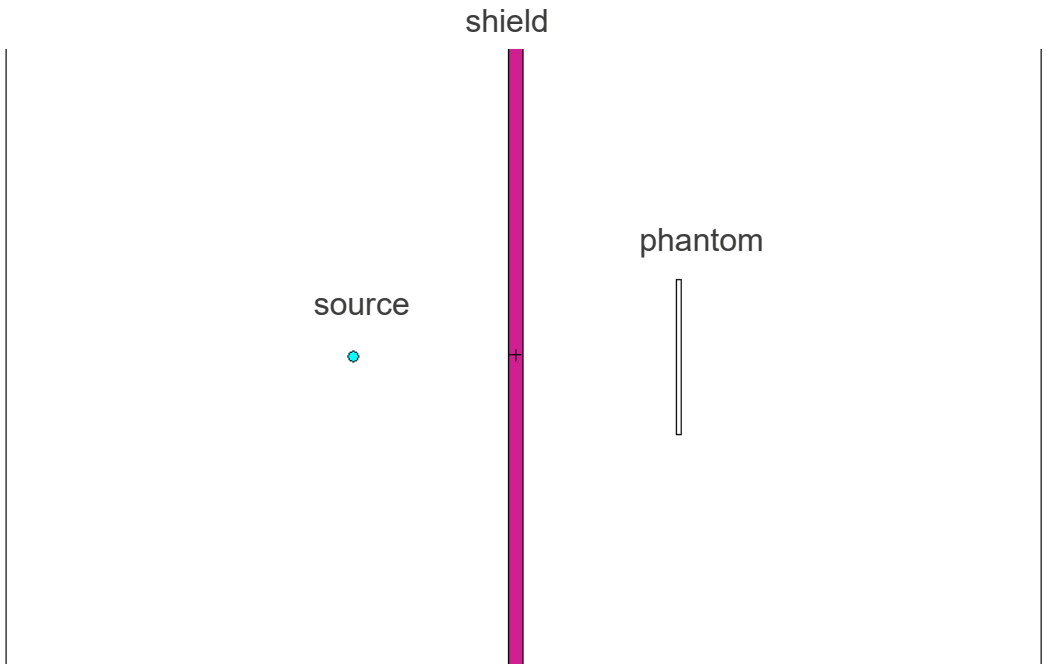
For same fluence rate, would need more moderation for Pu-239





External Dose – Neutrons, Shielding Considerations

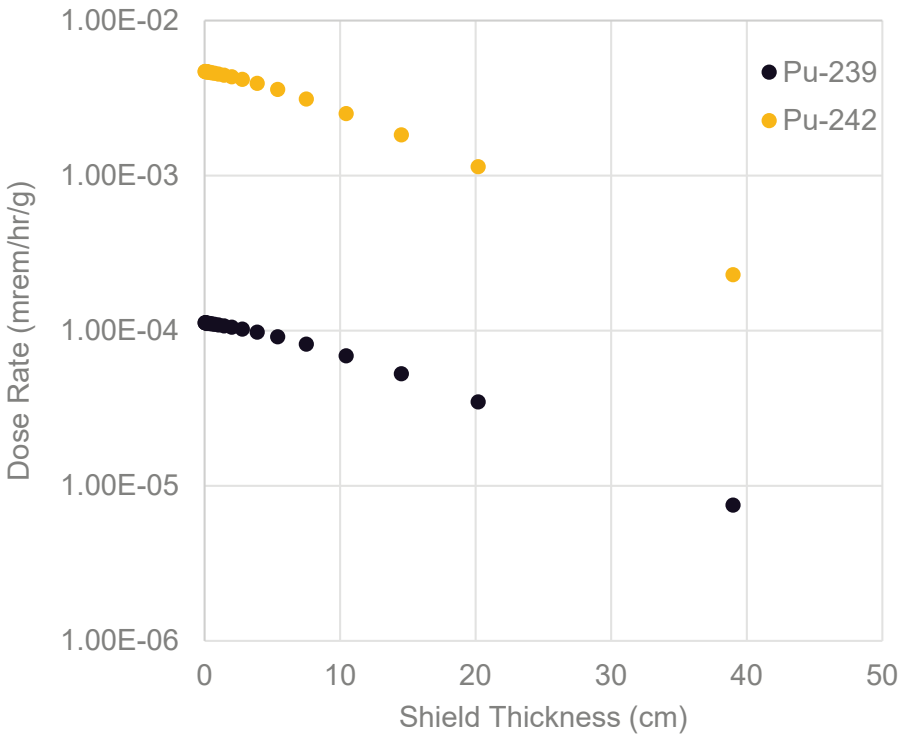
- Compared pure Pu-239 and Pu-242
  - Spectrum from OrigenArp
  - MCNP6.2
- Dose to air phantom
  - Fluence to Dose Conversion Factors from ICRP-74
    - Neutrons: Table A.42
- Shield:
  - Concrete: 0.0543 cm – 39 cm



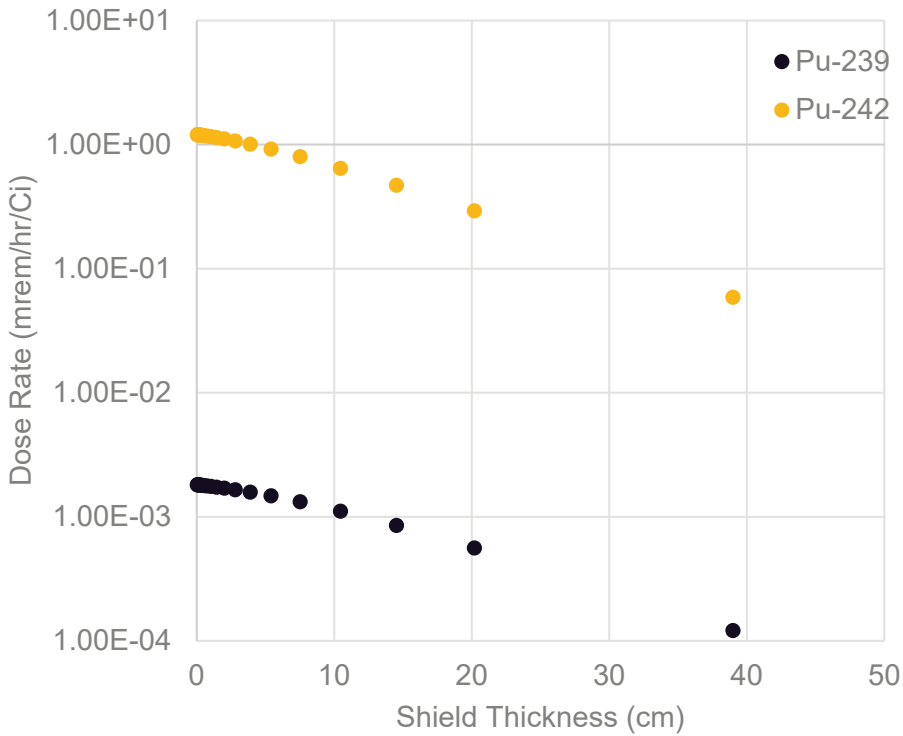
External Dose – Neutrons, Shielding Considerations

Concrete

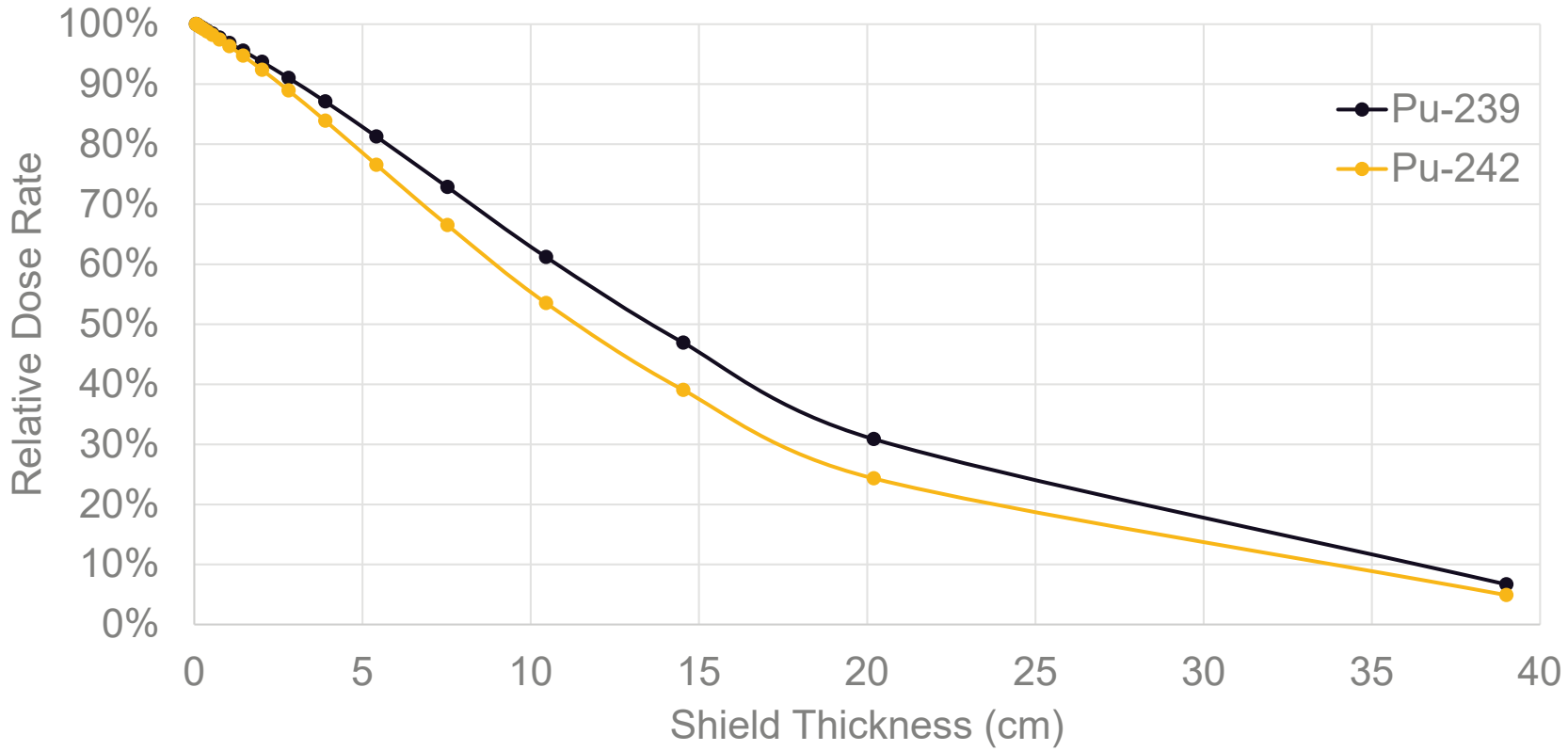
Same Mass



Same Activity



External Dose – Neutrons, Shielding Considerations



Internal Dose

Radioisotope	10 CFR 835 DAC (uCi/mL)	10 CFR 20 DAC (uCi/mL) Class W	10 CFR 20 DAC (uCi/mL) Class Y
Pu-239	5.00E-12	3E-12	7E-12
Pu-242	5.00E-12	3E-12	7E-12

all compounds except PuO_2

PuO_2

	ICRP 68		ICRP 72	
Radioisotope	Inhalation (rem-Ci^{-1})	Ingestion (rem-Ci^{-1})	Inhalation (rem-Ci^{-1})	Ingestion (rem-Ci^{-1})
Pu-239	1.18E+08	9.25E+05	4.44E+08	9.25E+05
Pu-242	1.15E+08	8.88E+05	4.07E+08	8.88E+05
	102.6%		*109.1%*	

Internal Dose

By Mass

Mass (g)	Pu-239, Dose (rem)	Pu-242, Dose (rem)
1E-09	7.32E-03	4.49E-04
1E-06	7.32E+00	4.49E-01
1E-03	7.32E+03	4.49E+02
1E+00	7.32E+06	4.49E+05

By Activity

Activity (Ci)	Pu-239, Dose (rem)	Pu-242, Dose (rem)
1E+01	1.18E-02	1.15E-02
1E+04	1.18E+01	1.15E+01
1E+07	1.18E+04	1.15E+04
1E+10	1.18E+07	1.15E+07

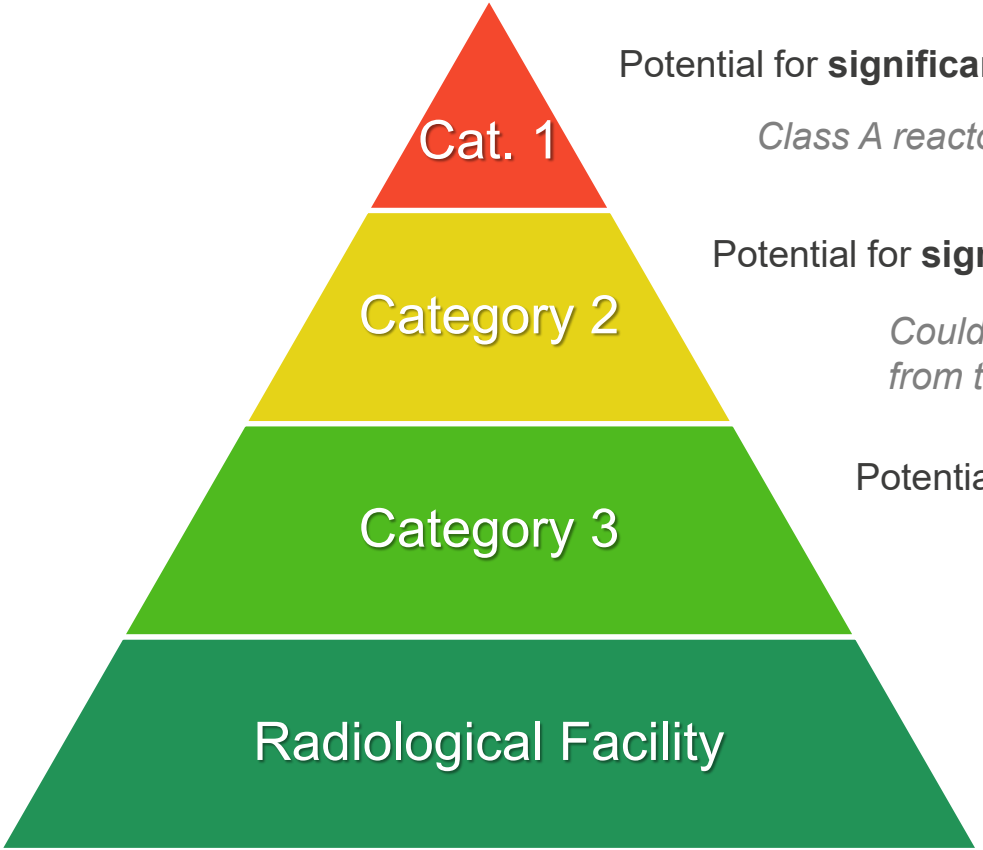
ICRP 68

Radioisotope	Inhalation (rem-Ci^{-1})	Ingestion (rem-Ci^{-1})
Pu-239	1.18E+08	9.25E+05
Pu-242	1.15E+08	8.88E+05

External and Internal Dose Summary

- Main difference between external and internal doses from Pu-239 and Pu-242 are due to specific activity not due to hazards
  - Both radionuclides have similar decay progeny and decay modes
- Pu-242 has a much higher neutron dose rate for equal mass and activity
- Pu-242 has a lower photon dose rate for equal mass, but has a higher photon dose rate for equal activity
- For large amounts of material and similar fluence rates:
  - Need more neutron moderation for Pu-239
  - Need thicker photon shielding for Pu-242

Hazard Categorization – Revisited



Potential for **significant off-site** consequences

Class A reactors (steady-state power greater than 20 MWt)

Potential for **significant on-site** consequences

Could produce doses of 1 rem in the range of 100 m from the facility (emergency off site evac)

Potential for only **significant localized** consequences

Facilities that cannot have a significant radiological impact outside of the facility

Hazard Categorization – Category 2 Threshold

10 CFR 30, Schedule C Threshold Quantities – Result in a dose of 1 rem at 100 m
Modified DOE Equation

$$Q = \frac{1\ \text{rem}}{RF \cdot SA \cdot \frac{X}{Q} \cdot (CEDE + RR + CSDE)}$$

	SA (Ci/g)
Pu-239	6.2×10^{-2}
Pu-242	3.9×10^{-3}

ICRP 68

	Inhalation ($rem\text{-}Ci^{-1}$)	Ingestion ($rem\text{-}Ci^{-1}$)
Pu-239	4.44E+08	9.25E+05
Pu-242	4.07E+08	8.88E+05

Q = Threshold Quantity (g)

RF = Airborne Release Fraction over an Entire Facility

1. *Gases* — *1.0*
2. *Highly Volatile/Combustible* — *0.5*
3. *Semi-Volatile* — *1E-2*
4. *Solid/Powder/Liquid* — *1E-3*

SA = Specific Activity

X/Q = Accounting for dilution of release at a point given the meteorological conditions *1E-4 sec/m³*

$CEDE$ = Committed Effective Dose Equivalent for a Given Radionuclide

RR = Respiration Rate *3.5E-4 m³/s*

$CSDE$ = Cloudshine Dose Equivalent

Hazard Categorization – Category 3 Threshold

EPA 400-R-92-001, EPA in Manual of Protective Action Guides and Protective Actions for Nuclear Incidents – Value for protection of workers for planned reentry into a facility after an incident (10 rem at 30 m).

$$Q = \frac{10\ \text{rem}}{RF \cdot SA \cdot \frac{X}{Q} \cdot (CEDE + RR + CSDE)}$$

	SA (Ci/g)
Pu-239	6.2×10^{-2}
Pu-242	3.9×10^{-3}

ICRP 72

	Inhalation (rem-Ci^{-1})	Ingestion (rem-Ci^{-1})
Pu-239	1.18E+08	9.25E+05
Pu-242	1.15E+08	8.88E+05

Q = Threshold Quantity (g)

RF = Airborne Release Fraction over an Entire Facility

1. *Gases* *1.0*
2. *Highly Volatile/Combustible* *0.5*
3. *Semi-Volatile* *1E-2* ★
4. *Solid/Powder/Liquid* *1E-3*

SA = Specific Activity

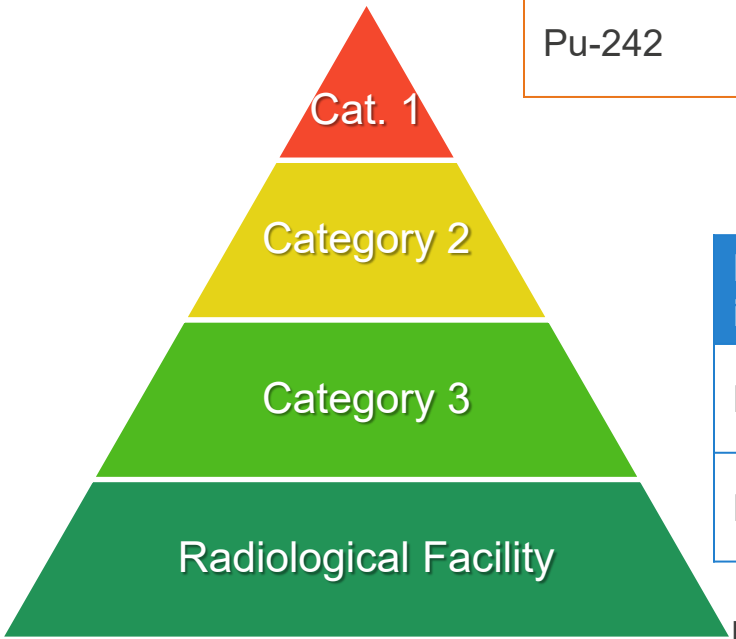
X/Q = Accounting for dilution of release at a point given the meteorological conditions *1E-4 sec/m³* ★

$CEDE$ = Committed Effective Dose Equivalent for a Given Radionuclide

RR = Respiration Rate *7.2E-2 m³/s* ★

$CSDE$ = Cloudshine Dose Equivalent

NNSA Categorizations



Activity (Ci)

Radioisotope	Radiological Facility	Category 3 Facility	Category 2 Facility
Pu-239	< 2.40E+00 Ci	2.40E+00 Ci to 1.62E+02 Ci	> 1.62E+02 Ci
Pu-242	< 2.56E+00 Ci	2.56E+00 Ci to 1.69E+02 Ci	> 1.69E+02 Ci

106.7% *104.3%*

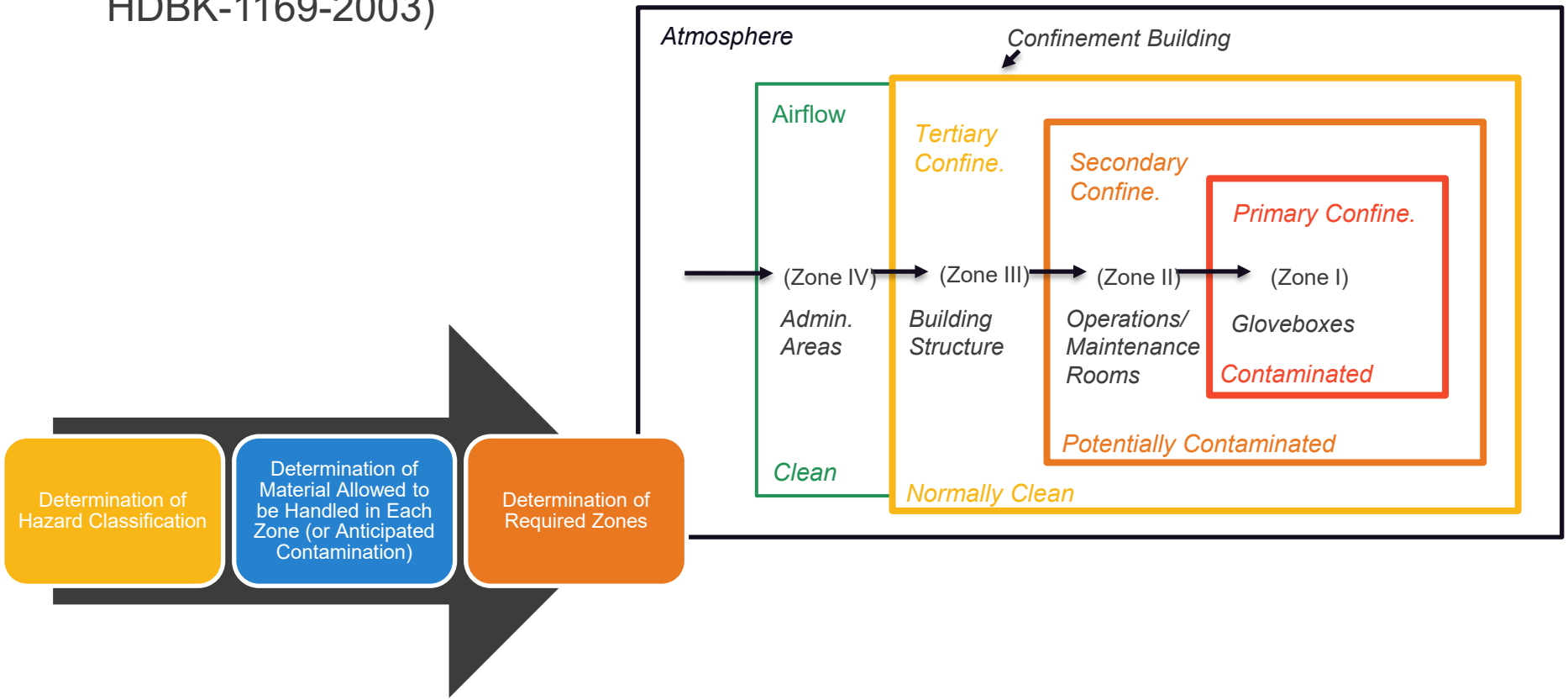
Mass (g)

Radio-isotope	Radiological Facility	Category 3 Facility	Category 2 Facility
Pu-239	< 3.86E+01 g	3.86E+01 g to 2.61E+03 g	> 2.61E+03 g
Pu-242	< 6.49E+02 g	6.49E+02 g to 2.29E+04 g	> 2.29E+04 g

Main difference = difference in the Specific Activity

Ventilation

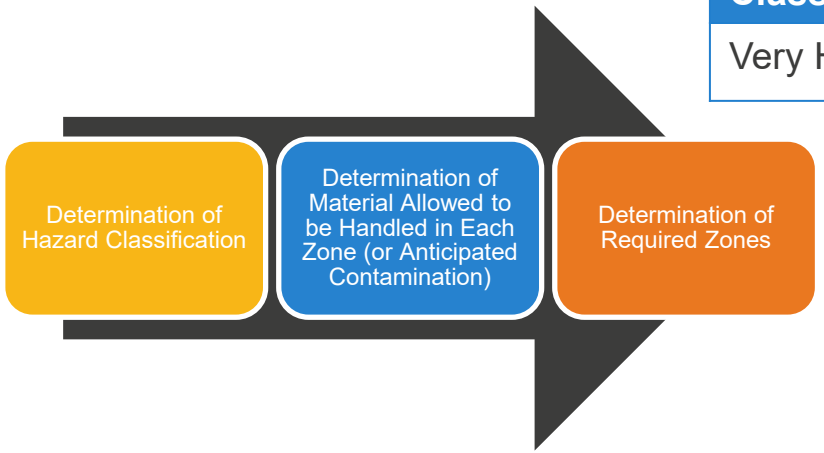
- Determination of Confinement Zones (Nuclear Air Cleaning Handbook, DOE-HDBK-1169-2003)



Ventilation

Hazard Class	Relative Hazard	DAC, Air (uCi/mL)
1	Very High	<1E-10
2	High	1E-10 to 1E-08
3	Moderate	1E-08 to 1E-06
4	Negligible	<1E-06

For Both Pu-239 and Pu-242, DAC is 5.00E-12 uCi/mL



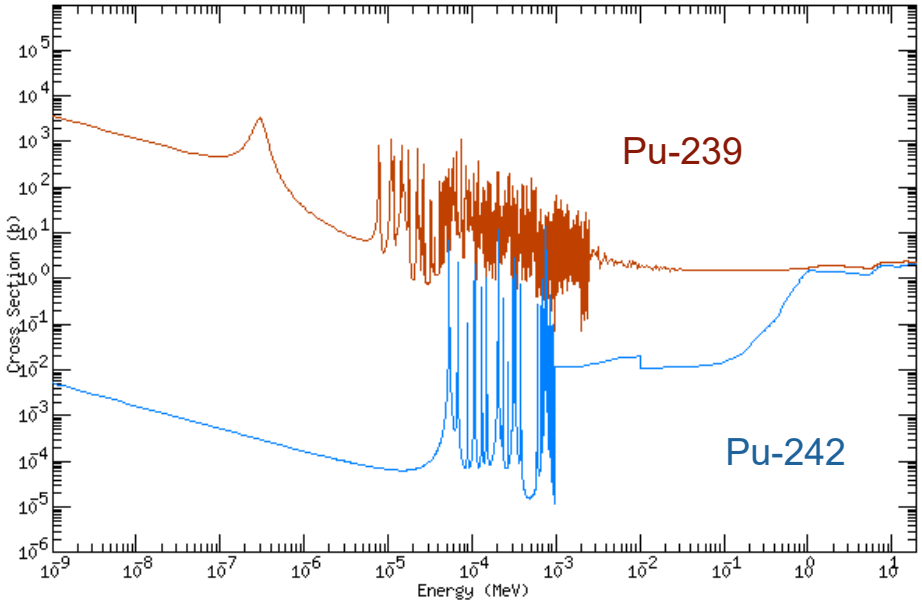
Hazard Class	Primary Confinement	Secondary Confinement	Tertiary Confinement
Very High	> 100 mCi	1.0 uCi – 100 mCi	0 – 1.0 uCi

Nuclide	Primary Confinement	Secondary Confinement	Tertiary Confinement
Pu-239	> 1.61E+00 g	1.61E-05 – 1.61E+00 g	0 – 1.61E-05 g
Pu-242	> 2.56E+01 g	2.56E-04 to 2.56E+01 g	0 – 2.56E-04 g

Criticality

	Not Requiring Criticality Evaluation or Controls	Significant Quantity Threshold (Full Criticality Safety Evaluation)
Pu-239	≤ 1 g	450 g
≥ 75% Pu-242	≤ 10 g	5000 g *

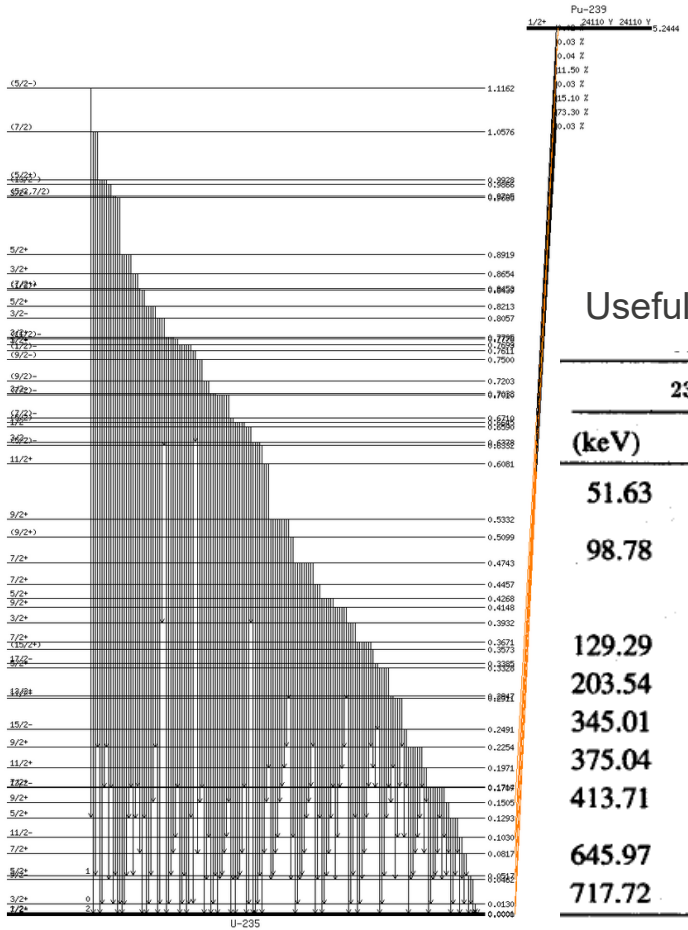
** Note: ANSI standard actually is 50,000 g*



at 0.0253 eV = 747.4 b

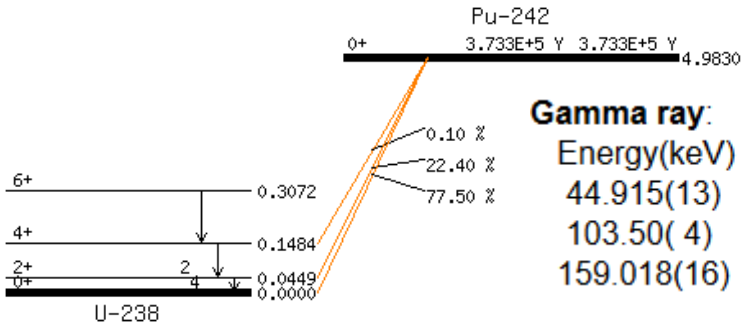
at 0.0253 eV = 2.557 mb

Detection



Useful Gammas

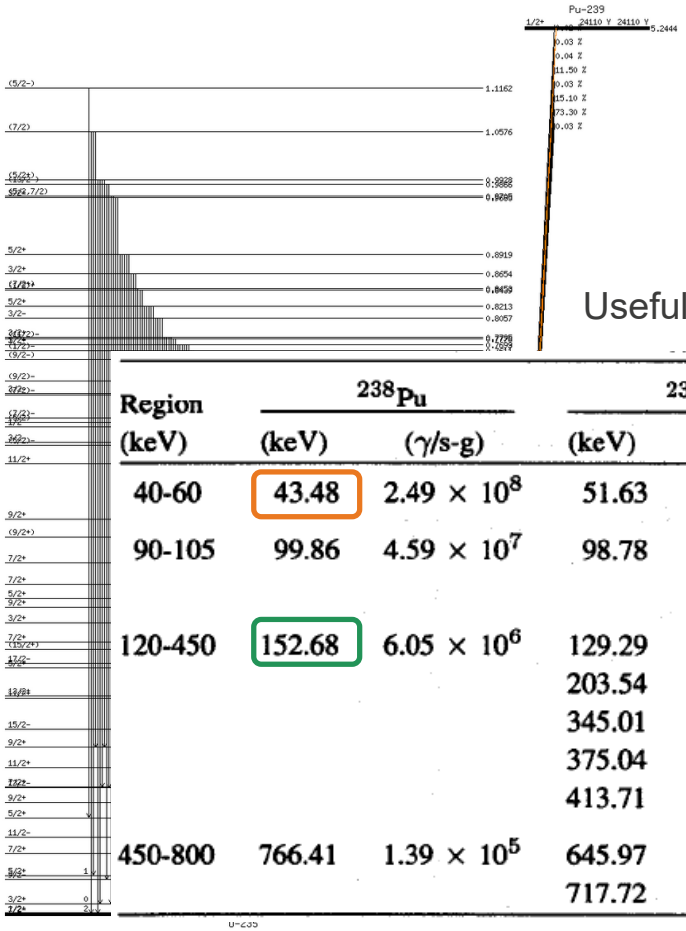
^{239}Pu	
(keV)	(γ/s-g)
51.63	6.19×10^5
98.78	2.80×10^4
129.29	1.44×10^5
203.54	1.28×10^4
345.01	1.28×10^4
375.04	3.60×10^4
413.71	3.42×10^4
645.97	3.42×10^2
717.72	6.29×10^1



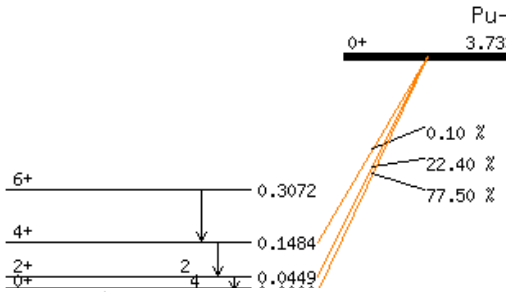
Gamma ray:

Energy(keV)	Intensity(rel)
44.915(13)	0.0384(8)
103.50(4)	0.0253(12)
159.018(16)	2.20E-4(8)

Detection



Useful Gammas



Gamma ray:

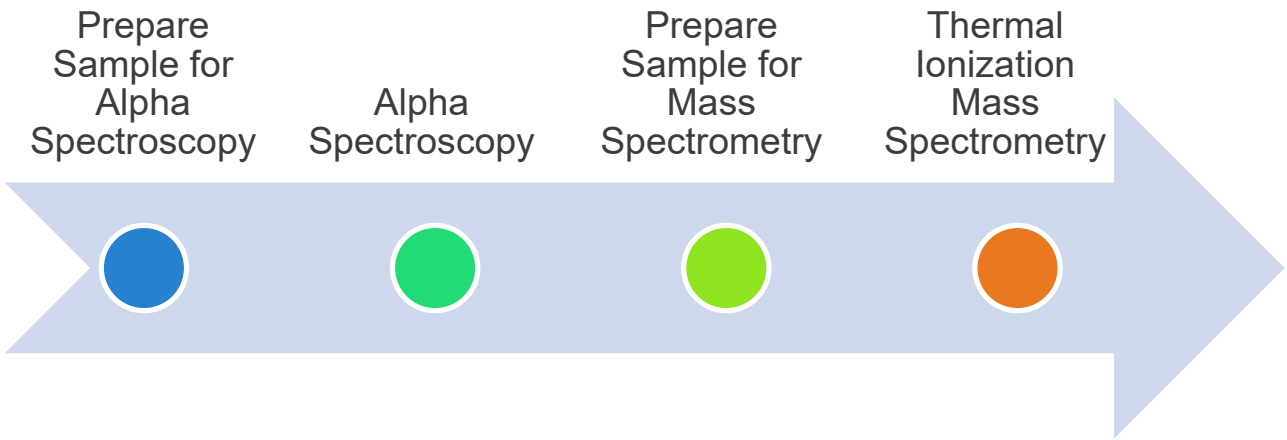
Energy(keV)	Intensity(rel)
44.915(13)	0.0384(8)
103.50(4)	0.0253(12)
159.018(16)	2.20E-4(8)

Region	^{238}Pu		^{239}Pu		^{240}Pu		^{241}Pu		^{241}Am	
(keV)	(keV)	(γ/s-g)	(keV)	(γ/s-g)	(keV)	(γ/s-g)	(keV)	(γ/s-g)	(keV)	(γ/s-g)
40-60	43.48	2.49×10^8	51.63	6.19×10^5	45.23	3.80×10^6	- - -		59.54	4.54×10^{10}
90-105	99.86	4.59×10^7	98.78	2.80×10^4	104.24	5.86×10^5	103.68	3.86×10^6	98.95	2.57×10^7
									102.97	2.47×10^7
120-450	152.68	6.05×10^6	129.29	1.44×10^5	160.28	3.38×10^4	148.57	7.15×10^6	125.29	5.16×10^6
			203.54	1.28×10^4			164.58[a]	1.73×10^6	335.40	6.28×10^5
			345.01	1.28×10^4			208.00[a]	2.04×10^7		
			375.04	3.60×10^4			332.35[a]	1.14×10^6		
			413.71	3.42×10^4			370.93[a]	1.04×10^5		
450-800	766.41	1.39×10^5	645.97	3.42×10^2	642.48	1.05×10^3	- - -		662.42	4.61×10^5
			717.72	6.29×10^1					721.99	2.48×10^5

Internal Dosimetry

- LANL does not currently have a procedure for potential intakes of Pu-242
  - Pu-242 is used as a tracer for chemical operations

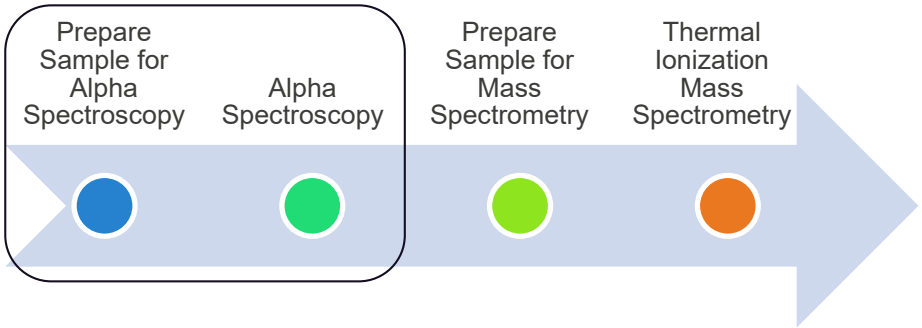
Analysis Method	Radionuclide	Detection Limit	Sample Load	Turnaround
Alpha Spectroscopy	Pu-238, Pu-239/Pu-240	0.01 pCi/aliquot	2000	Standard: 14 days
Mass Spectrometry	Pu-239, Pu-240, Pu-242	0.001 pCi/aliquot	1500	Standard: 35 days



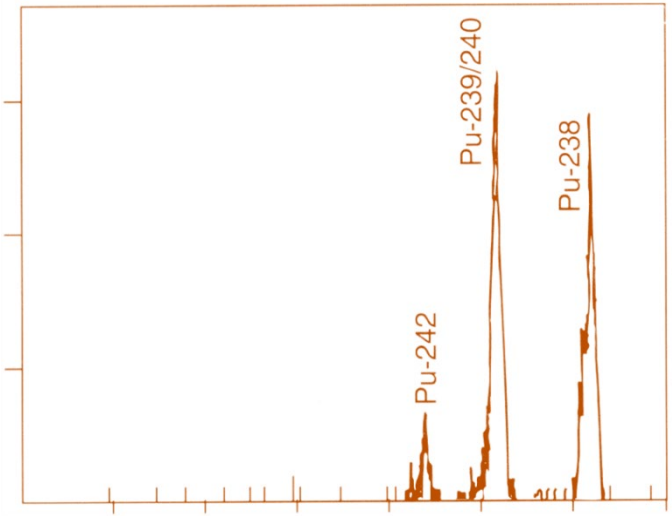
Internal Dosimetry

- Alpha spectroscopy is necessary, because of high U-238 background

- To prepare sample:
  - Chemical separation
  - Addition of Pu-242 tracer
  - Electroplate sample on stainless steel plates

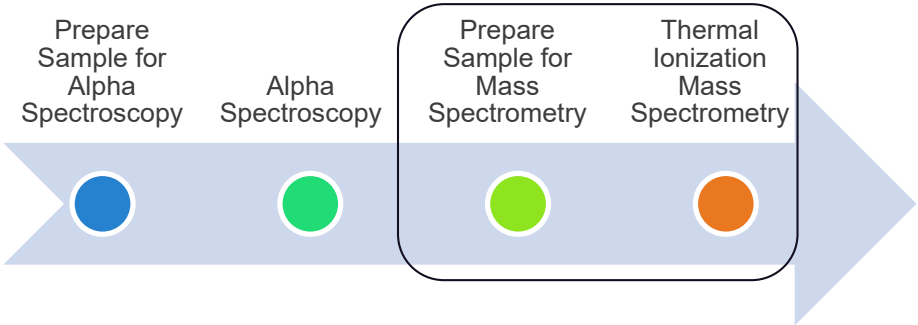


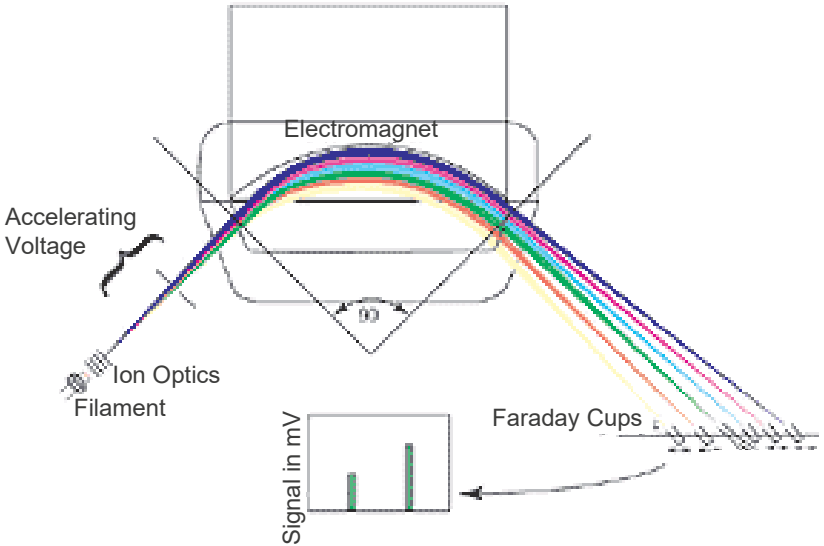
Nuclide	Main Alpha
Pu-238	5.499 MeV (70.91%) 5.456 MeV (29.89%)
Pu-239	5.144 MeV (17.11%) 5.106 MeV (11.94%)
Pu-240	5.168 MeV (72.8%) 5.124 MeV (27.1%)
Am-241	5.486 MeV (84.8% 5.443 MeV (13.1%)
Pu-242	4.902 MeV (76.5%) 4.848 MeV (23.4%)



Internal Dosimetry

- To prepare sample:
  - Need to extract sample from the stainless steel plates
  - Additional separation chemistry performed
  - Co-electroplated with platinum onto a rhenium filament



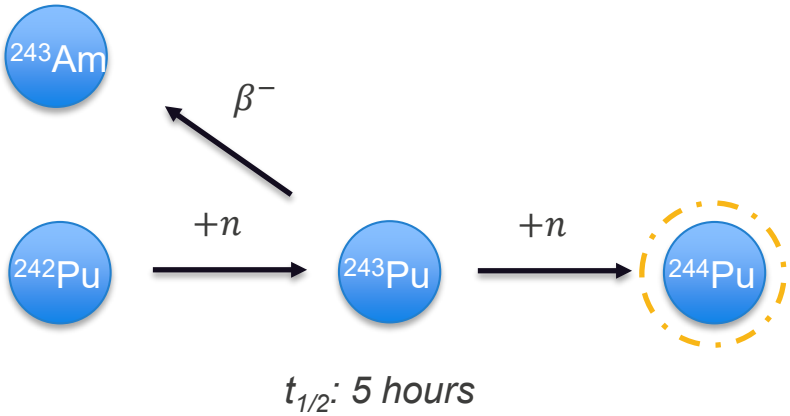


Internal Dosimetry

- Necessary changes for Pu-242:
  - Use Pu-244 as a tracer
    - Use of 2 samples:
      - One through traditional Pu processing
      - One with Pu-244
  - Ideally would have 3 to 6 month lead time
    - Verify chemistry and instrument response
    - Time to update the TIMS system & data processing system
    - Time for dosimetry to update dosimetry calculations
- Other complications
  - Cross contamination
    - Most equipment is only used once
    - Can handle 6-7 orders of magnitude
    - Ideally, would have separate laboratory space
  - Possible creep on TIMS machine

Internal Dosimetry

- Pu-244 complications:
  - Limited inventory of Pu-244
    - Ideally, batch sampling with ~20 employees
    - Up to 15 years inventory
  - Not DOELAP accredited for Pu-244



Summary of Considerations

- What do we care about – activity or mass of material?
  - Pu-242 has a much lower specific activity
    - More mass allowed in confinement zones
    - More material before CSE required
    - More material allowed before considered Cat 3 facility
  - If want same activity, not much difference

- Dose Rates
  - Pu-242 has a much higher neutron dose rate for equal mass and activity
  - Pu-242 has a lower photon dose rate for equal mass, but higher for equal activity

- Pu-242 is harder to distinguish using gamma spectroscopy

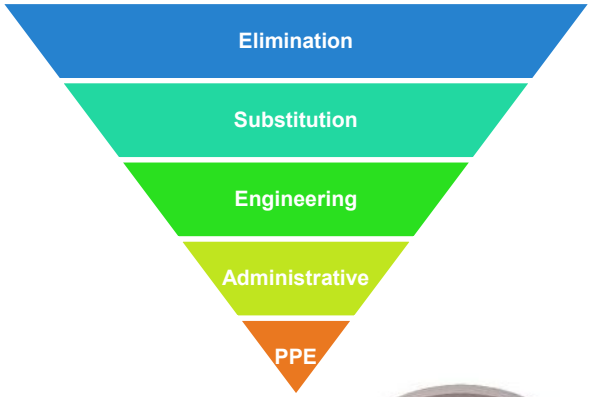
- Might need to develop new internal dosimetry procedures

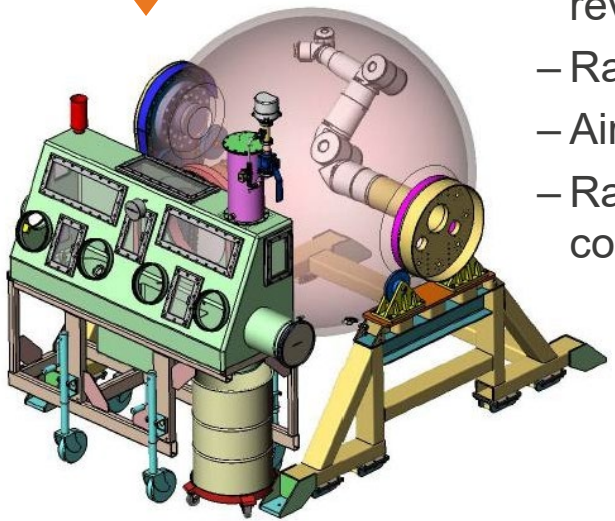
Radiological Engineering

Radiological Engineering focuses on the engineering design aspect of radiation protection for new or modifications to existing facilities, equipment, or operations.

- Program Drivers
  - 10CFR835
    - Chapter 12 of Attachment A of P121
    - Chapter 11 of the Engineering Standards Manual

Functions of Radiological Engineering



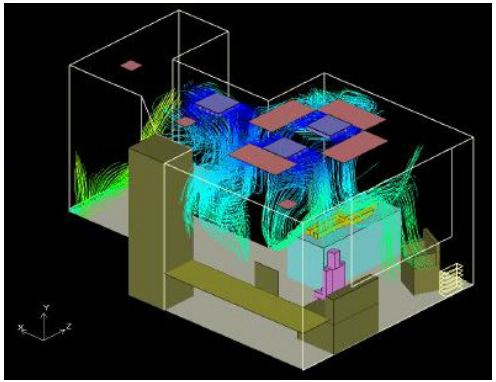


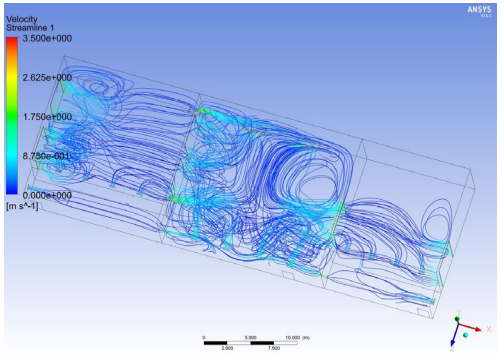
- Supports mission by providing:
  - Engineering design reviews
  - Radiation dose estimates
  - Airflow modeling (CFD)
  - Radiological project coordination
- Enables success by specifying:
  - HEPA filtration
  - Shielding (MCNP)
  - ALARA optimization
  - General radiological design aspects

Airflow Modeling

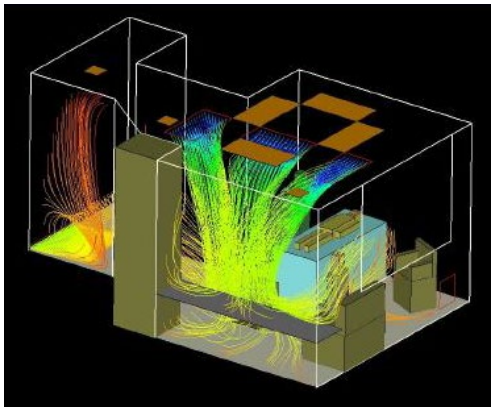
- Goal: Airborne and Contamination Control
- Specifications for ventilation
  - Cascading airflow
  - Containment Systems/ HEPA filtration
- Computation fluid dynamic codes
  - ANSYS Fluent
- Smoke testing

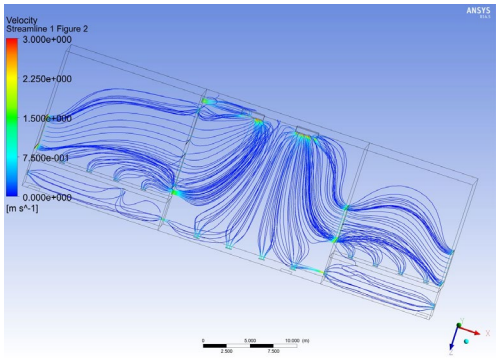
Before



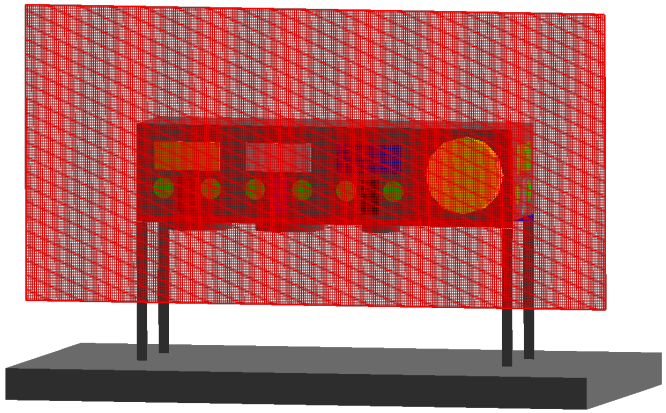


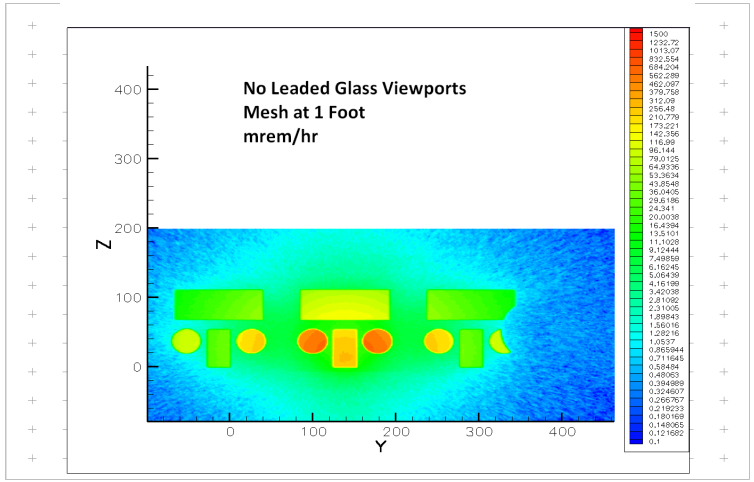
After





Detailed Shielding Analysis



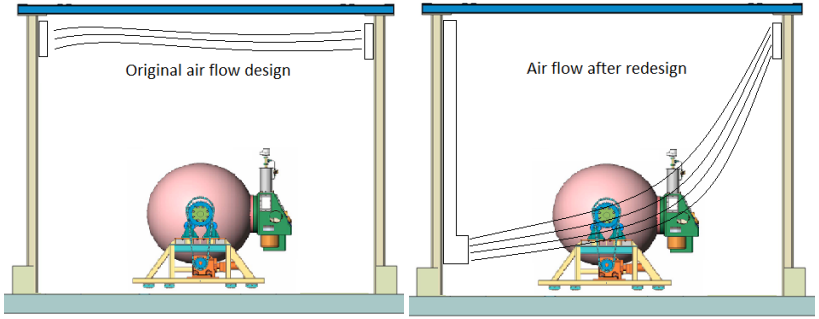


- Goal: Dose Reduction
- Computer Calculation
  - MicroShield
  - MCNP
  - Atilla
- Simulations allow for:
  - Detailed shielding analysis
  - Visualization tools to help make detailed decisions
  - Incorporation of ALARA optimization to balance cost versus benefits
- Time/motion studies

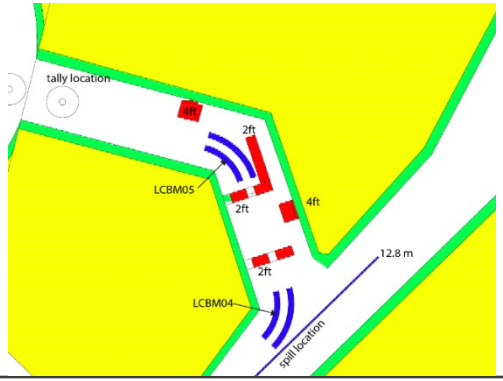
Key Projects

TA-35 TFF Boutique Pu Lab

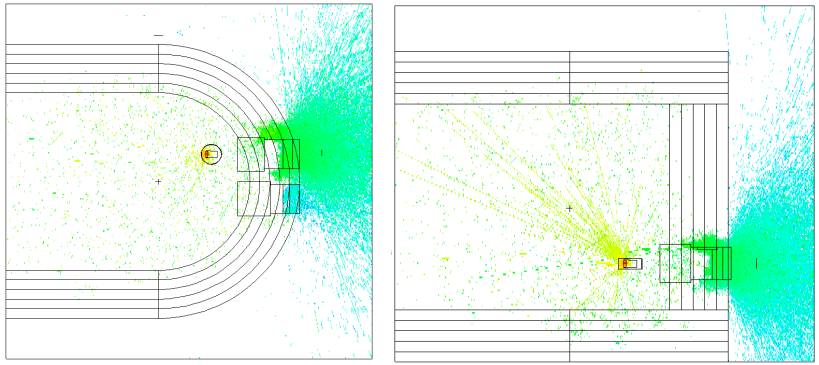
CMR CVD Ventilation Design



TA-53 Line C Shield Wall



DARHT Axis I Bullnose Penetration



Radiological Engineering

FOD(s)	Lead(s)	Backup
TA-55 & Weapons Programs	Jenelle M. & Luke H.	
LANSCE	John B.	Jordan D.
STO, D&D, and Small Science	Jordan D.	John B.