Nuclear Forensics

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Outline

Seaborg’s Plutonium ?

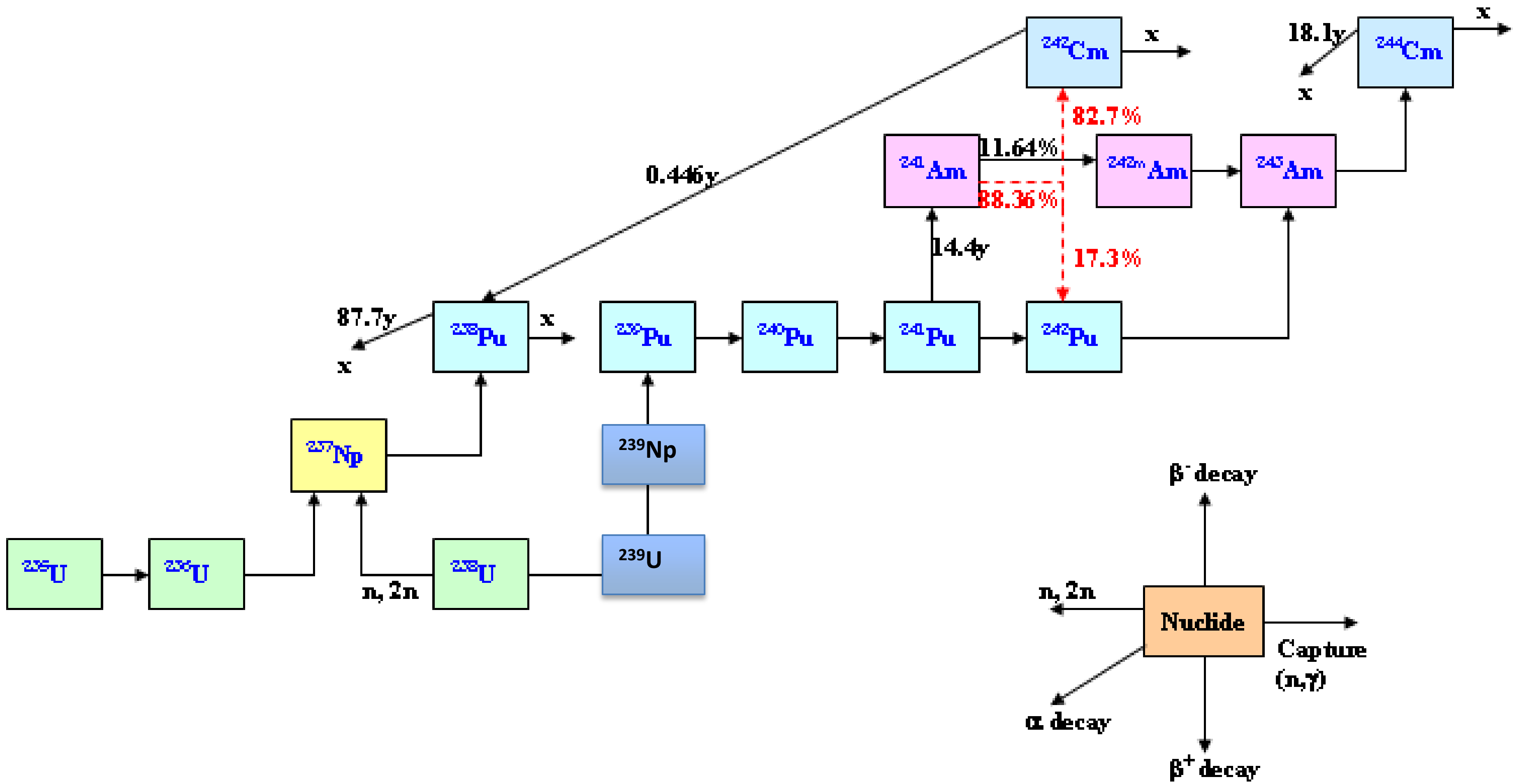
Explosion Yield of the Trinity Device

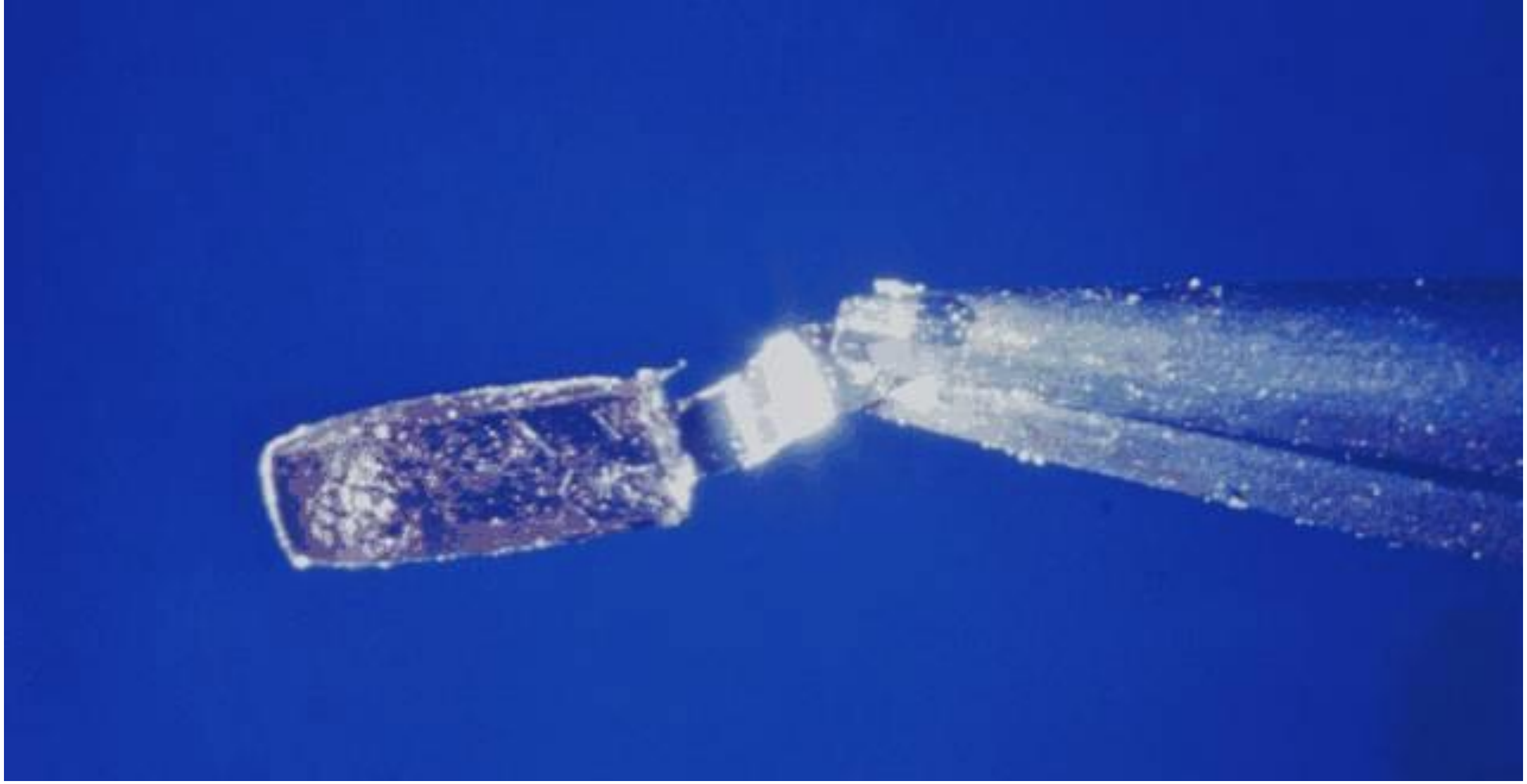
Anti-neutrino Monitoring of Nuclear Reactors

Neutron Capture Cross Section of ^{88}Zr

Seaborg's Plutonium ?

Production of Plutonium Isotopes



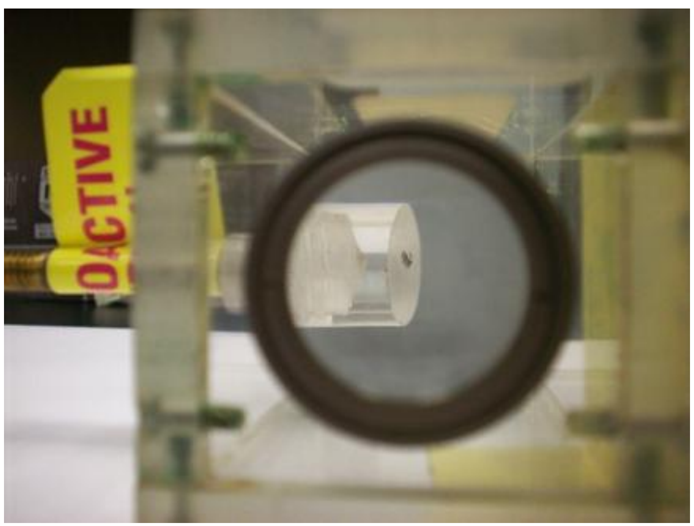


The first sample of ^{239}Pu containing 2.7-micrograms of oxide was weighed on September 10, 1942, at the University of Chicago's Metallurgical Laboratory. It is shown here as a deposit on a platinum foil held by forceps.

(a)

(b)

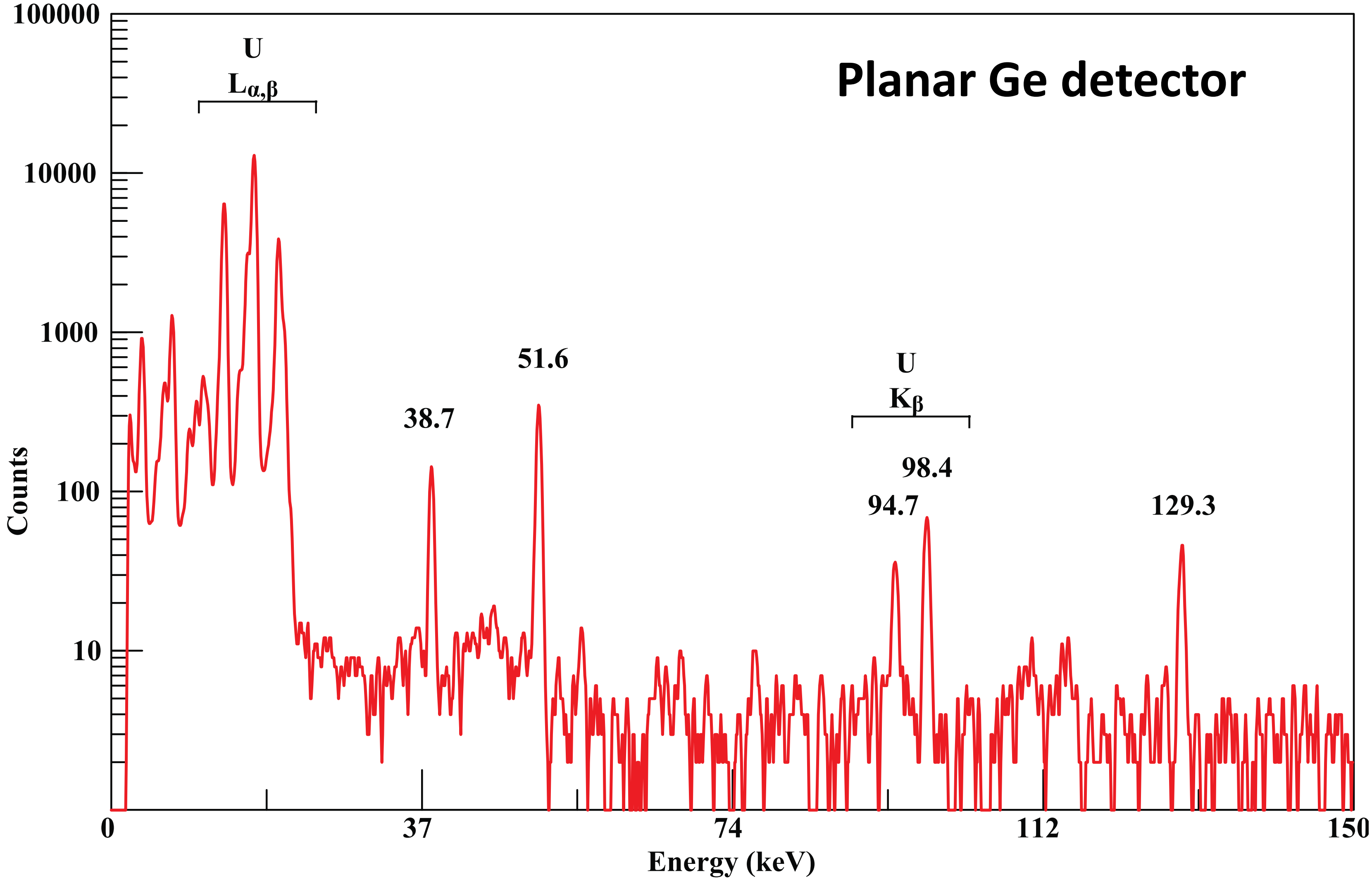
(c)



(a) outside of sample box with labels

(b) head-on view showing plastic rod with sample attached

(c) side view showing sample attached to plastic rod.

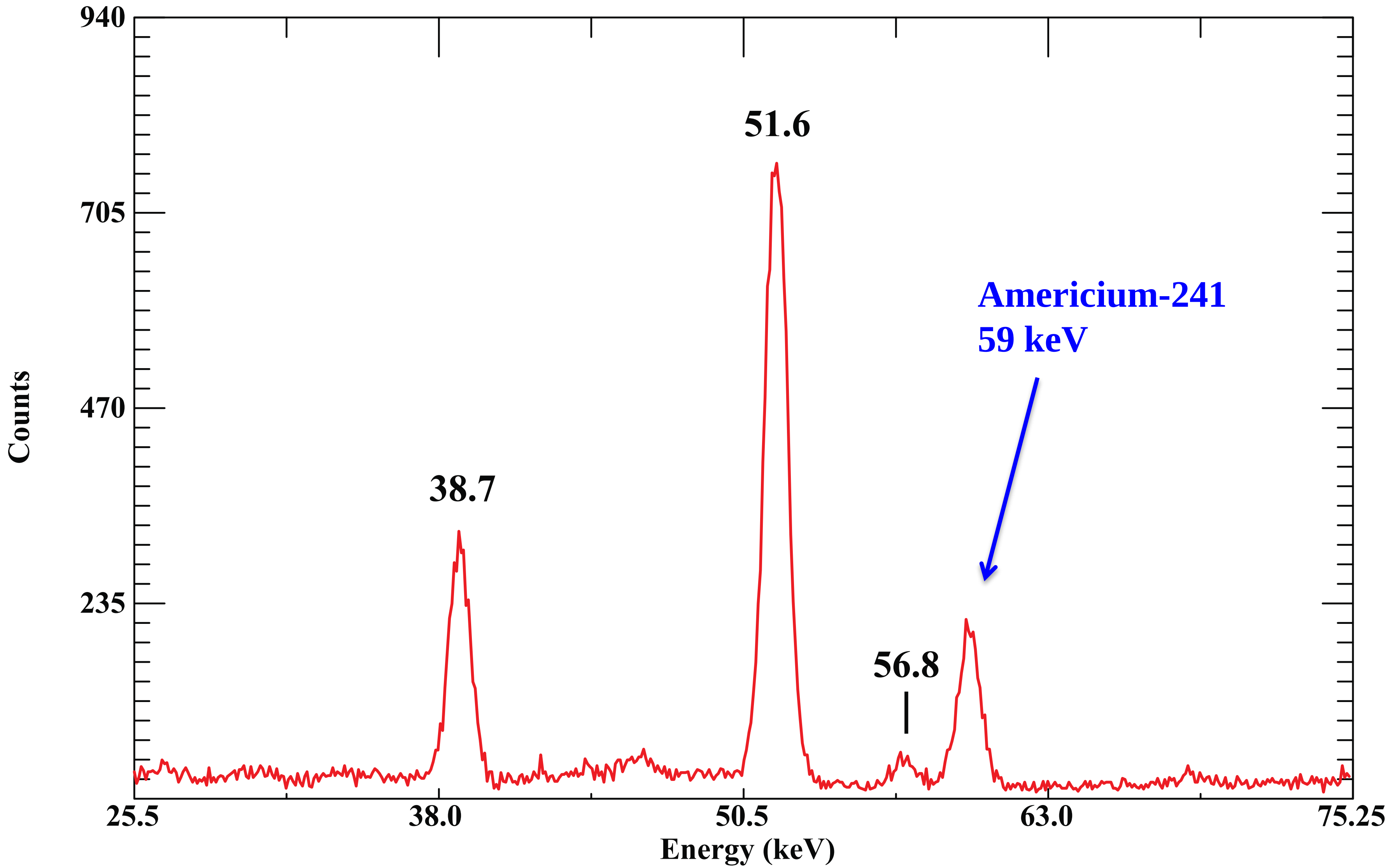


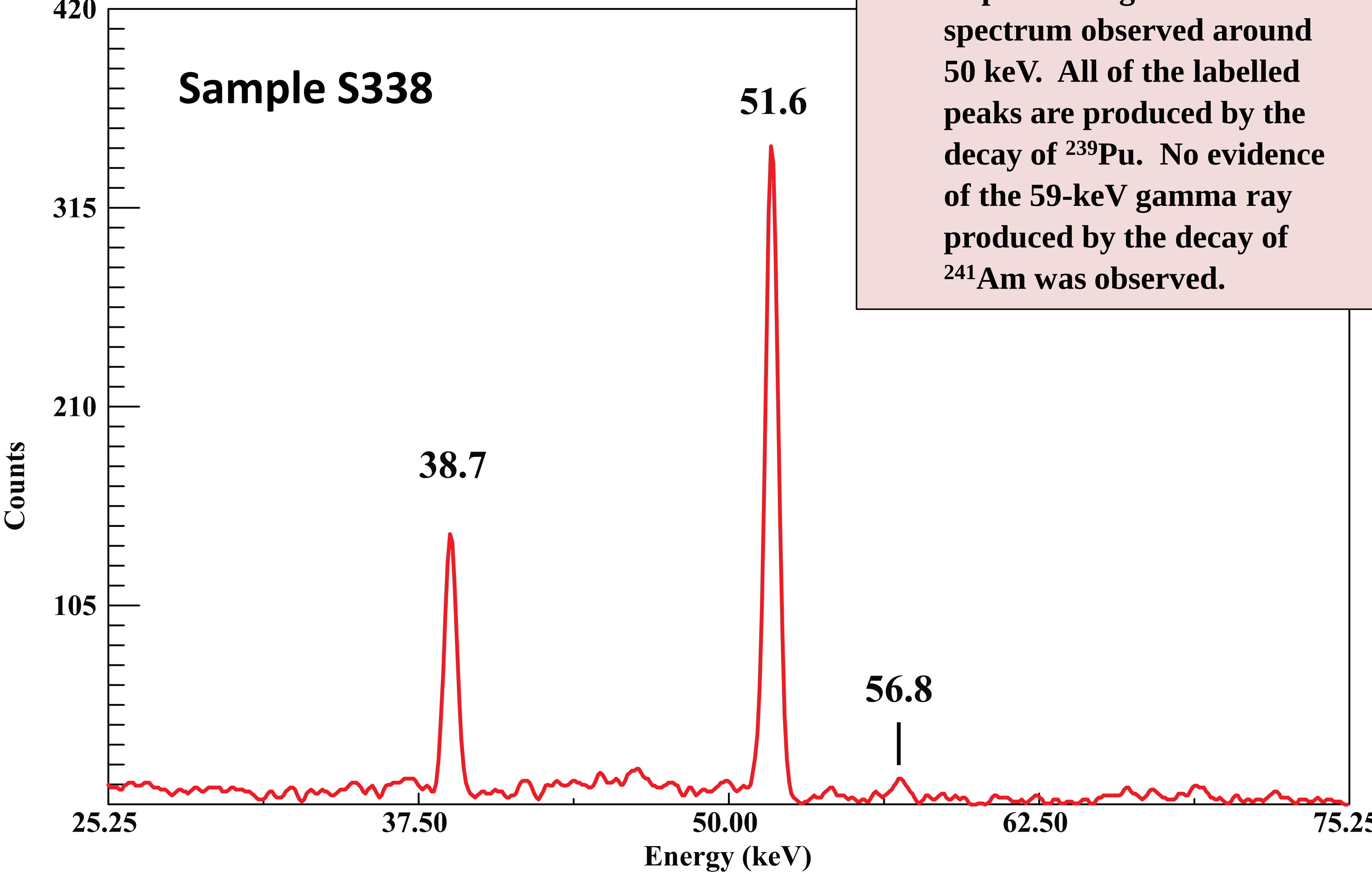
Background subtracted spectrum observed from Sample S338. All of the labelled peaks are x-rays and gamma rays produced by the decay of ^{239}Pu.

To determine the mass of ^{239}Pu contained in S338, we:

1. **Measured the efficiency of our detector using calibrated sources of ^{57}Co, ^{137}Cs, and ^{241}Am. These sources provide x-ray and gamma-ray lines at 26, 32, 36, 59, 122, and 136 keV.**
2. **Gamma-rays emitted from the S338 sample had to pass through the 0.63-cm thick wall of the plastic box in which it is contained. In order to account for the attenuation this produced, we placed a 0.63-cm thick block of polyethylene between our sources and the detector.**
3. **We extracted the peak areas of the 38.7, 51.6, and 129.3-keV lines from the spectrum obtained from S338 and then determined the sample mass from each line. Results were averaged to establish the mass of ^{239}Pu contained in sample S338 to be 2.0 $\pm$ 0.3 μg.**
4. **Seaborg stated that the first weighed sample contained 2.77 μg of PuO_2 with no uncertainty given. This would imply a ^{239}Pu mass of 2.44 μg.**
5. **Thus, the mass we determined is in reasonably good agreement with what Seaborg stated.**

Gamma spectrum using the same detector from "modern" Pu.





Expanded region of the spectrum observed around 50 keV. All of the labelled peaks are produced by the decay of ^{239}Pu. No evidence of the 59-keV gamma ray produced by the decay of ^{241}Am was observed.

In his reports, Seaborg states that 45 kg of uranium irradiated for 2 months with neutrons (produced by deuteron breakup) produced 200 μg of Pu.

From this one can infer a total neutron fluence

$$\Phi_n = 2\text{x}10^{15} \text{ neutrons/cm}^2$$

Note: This is about the same fluence a uranium nucleus in a modern commercial power plant sees in a few seconds !

→ expect N(^{239}Pu) : N(^{240}Pu) : N(^{241}Pu) = 1.00: 3x10^{-7} : 6x10^{-14}

After 72 years, almost all of the ^{241}Pu would have decayed to ^{241}Am, producing less than 2μBq of activity(far too small for us to see)

Thus, our failure to observed ^{241}Am is consistent with S338 being Seaborg's plutonium.

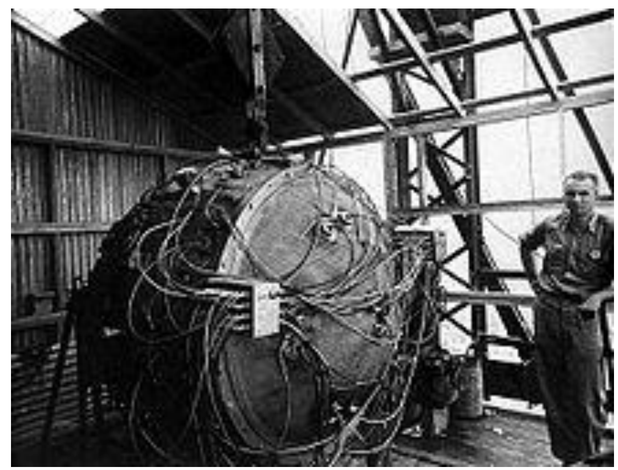
If it looks like a duck, swims like a duck, and quacks like a duck, then it probably is a duck.

A **mallard** correctly identified as a duck using the duck test

E. B. Norman, K. J. Thomas, K. E Telhami, Amer. Journ. Phys. 83 (2015) 843

Explosion Yield of the Trinity Device

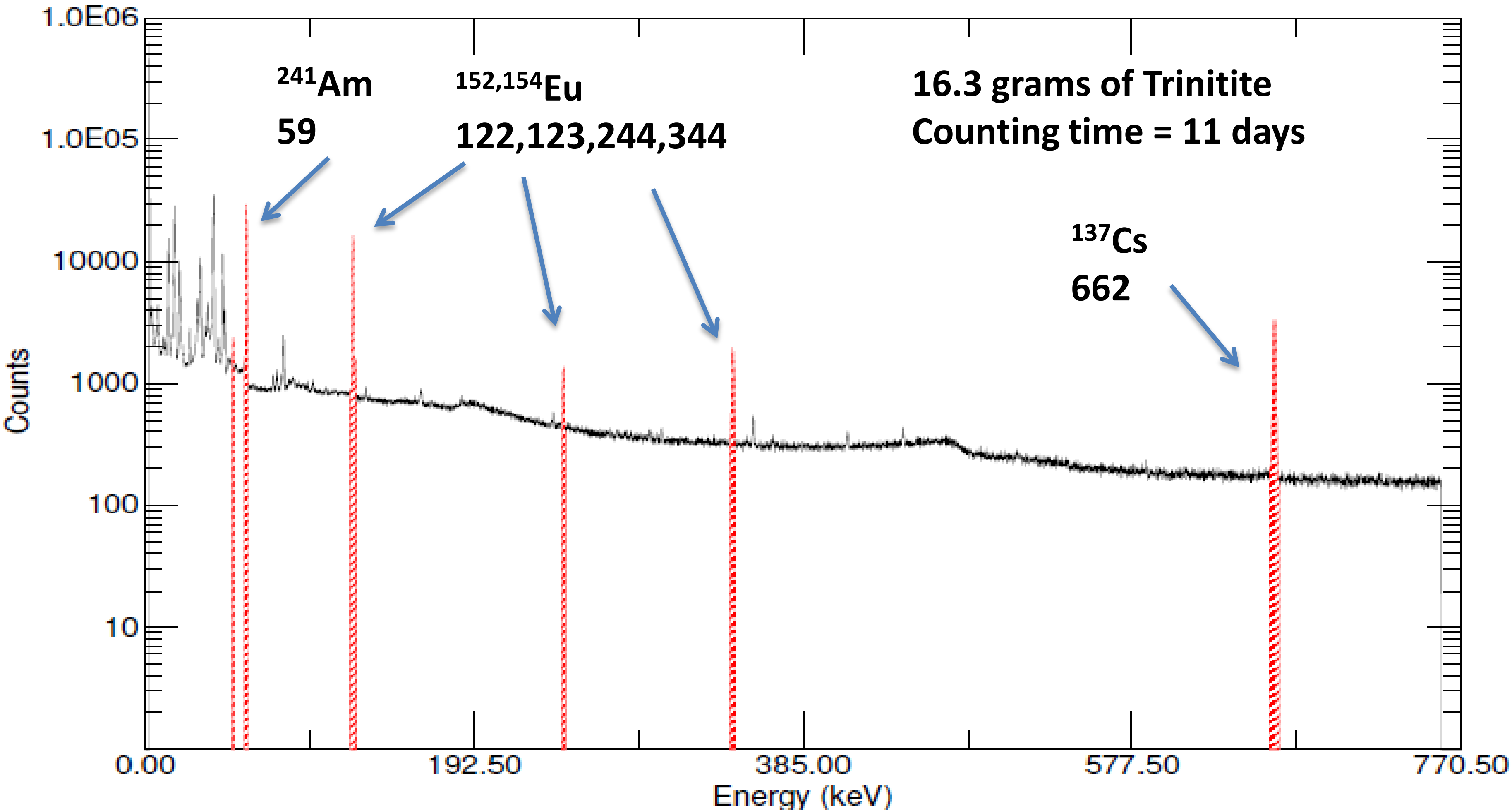
Trinity: First Nuclear Weapon Explosion



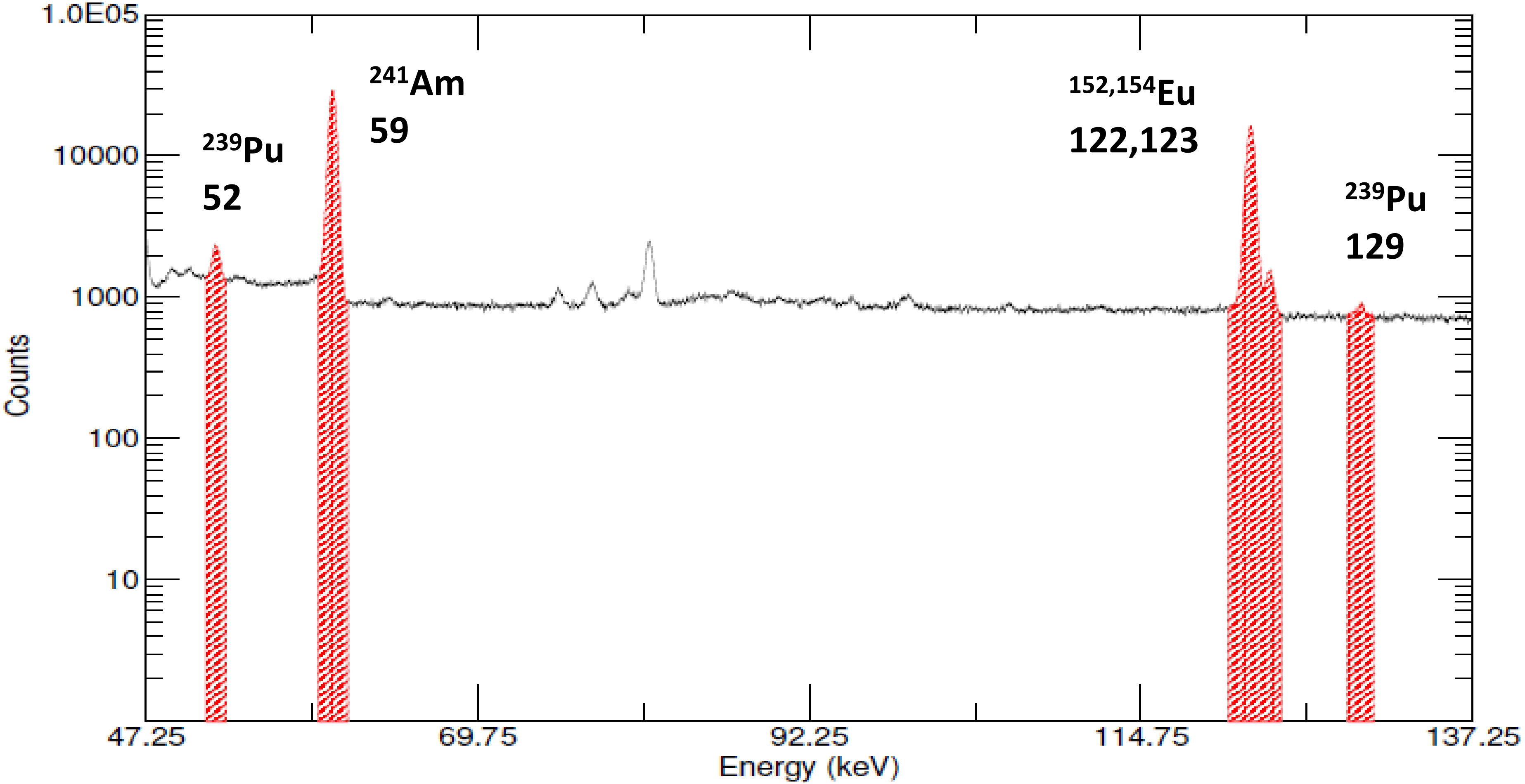
Trinitite Analysis

4 samples of Trinitite obtained from Mineralogical Research Co. (San Jose, CA)
9 – 16 grams each

Gamma-ray spectrum observed with planar Ge detector



Low Energy Portion of Spectrum



Simple Minded Analysis

Use observed counting rates of 52- and 129-kev lines from ^{239}Pu and 662-keV line from ^{137}Cs to determine yield of Trinity device

- Assume same probability for Cs and Pu to become incorporated in Trinitite
  - Due to high volatility of Cs, this should lead us to underestimate the yield
- Neglect fission yield from tamper (depleted uranium)
  - Will also cause us to underestimate yield

Interesting interplay of nuclear physics and chemistry involved in analysis

→ Our Measured Yields = 3.5 – 9.8 kT

Low, but correct order of magnitude
from simple minded analysis

Actual yield = 21 kT

Anti-neutrino Monitoring of Nuclear Reactors

Nuclear reactors produce enormous numbers of anti-neutrinos which can be used to monitor reactor operations

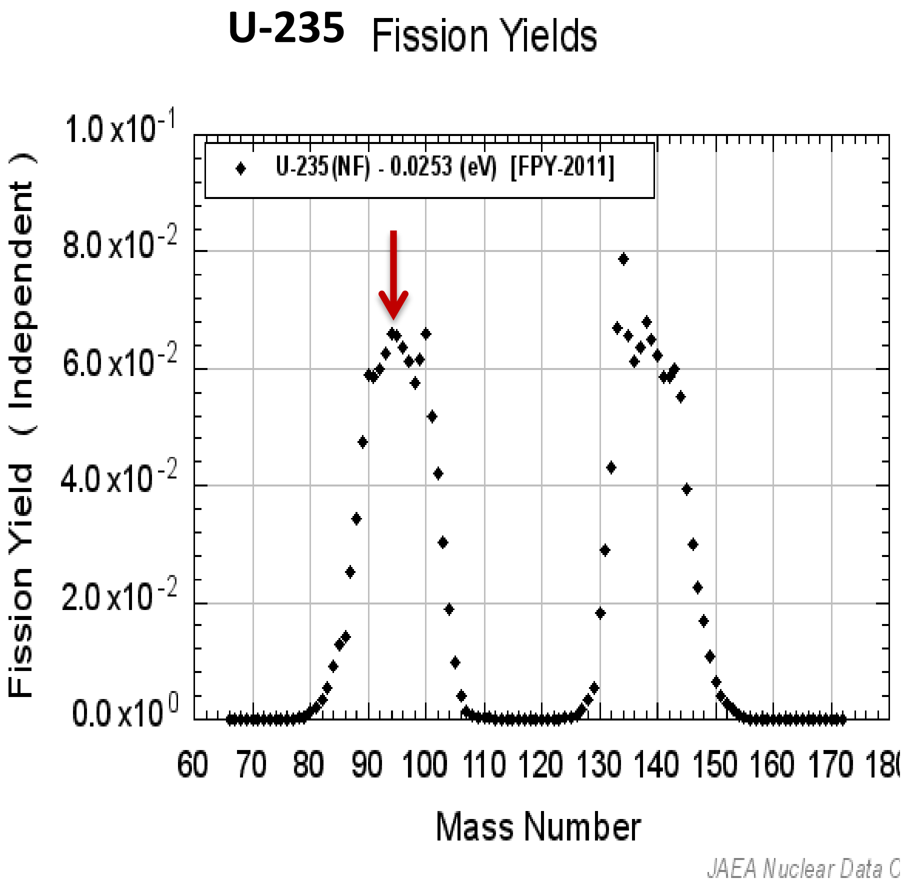
$^{235}U + n \rightarrow$ fission $\rightarrow$ $^{92}Rb + {}^{142}Cs + 2n$ (only one example)

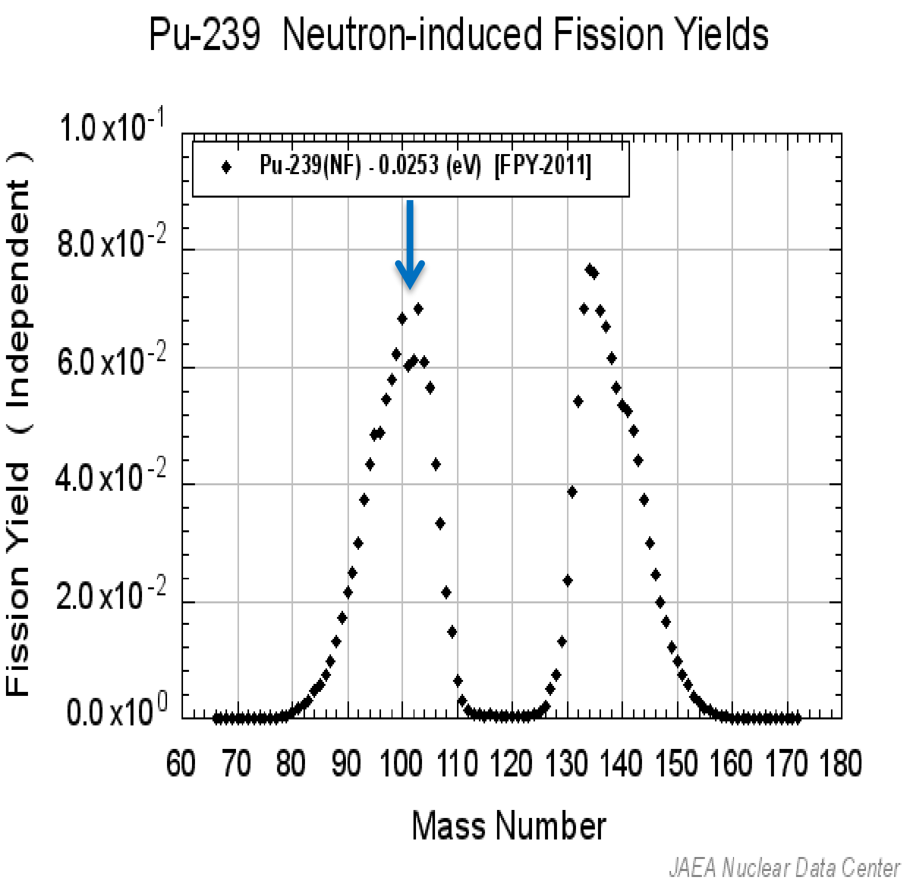
$$^{92}Rb \rightarrow {}^{92}Sr + e^{-} + \bar{\nu}_e$$

$$^{142}Cs \rightarrow {}^{142}Ba + e^{-} + \bar{\nu}_e$$

Mass distribution of fission products depends on the fissile material

U-235

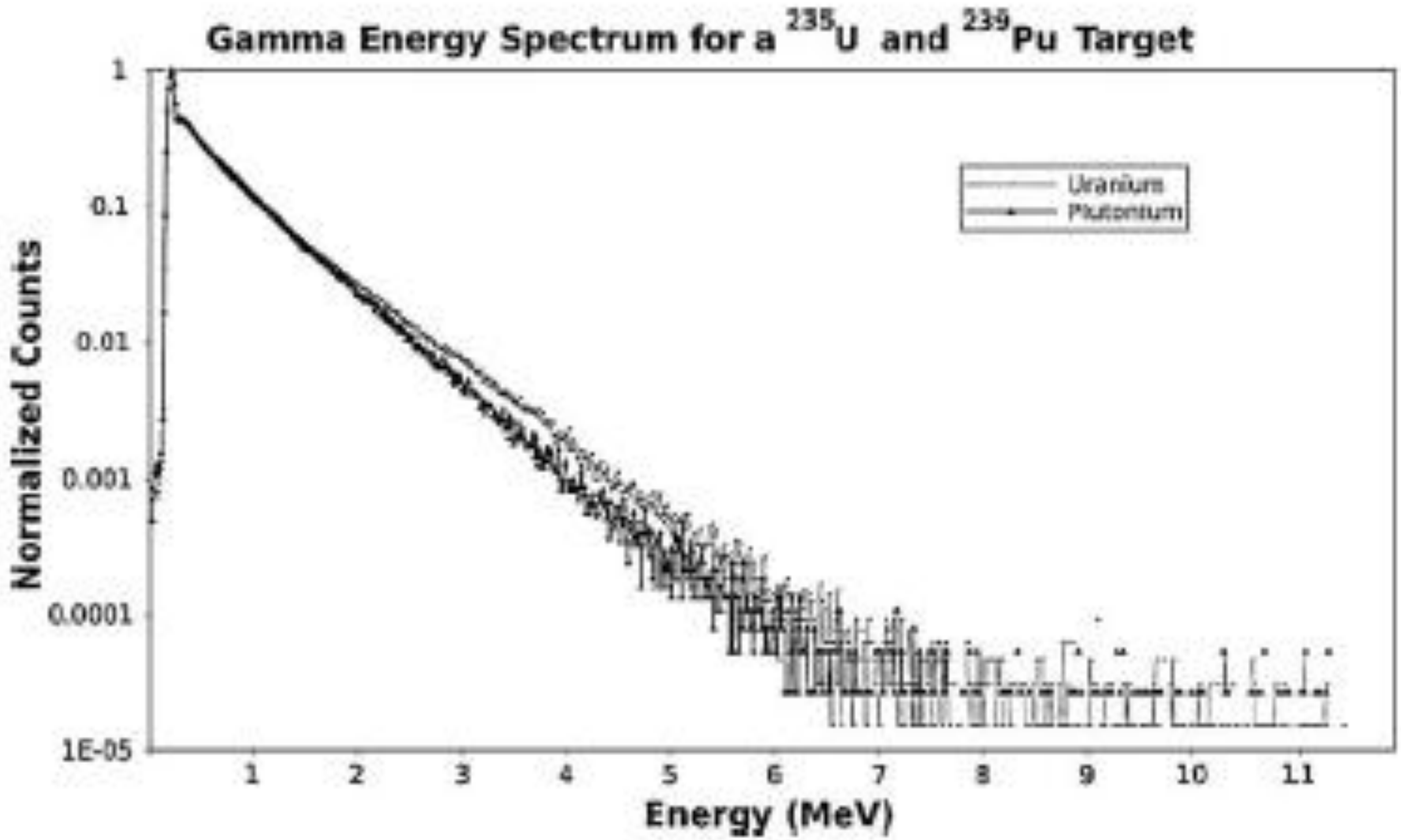




Mass distributions for thermal fission of ^{235}U

Mass distributions for thermal fission of ^{239}Pu

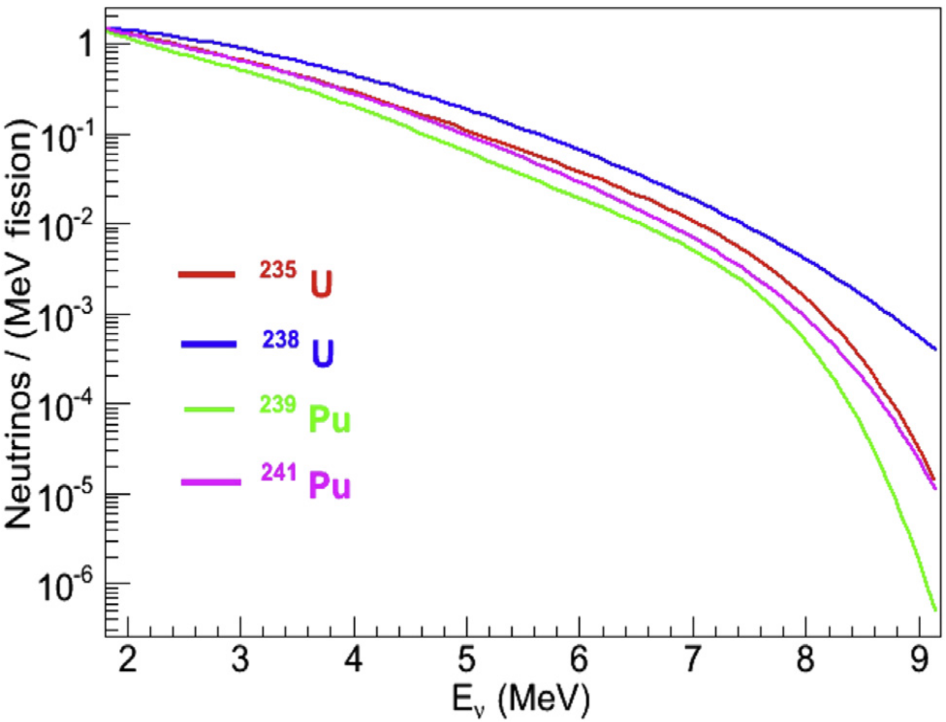
Beta-delayed gammas following fission observed through 0.75” Pb with plastic scintillator

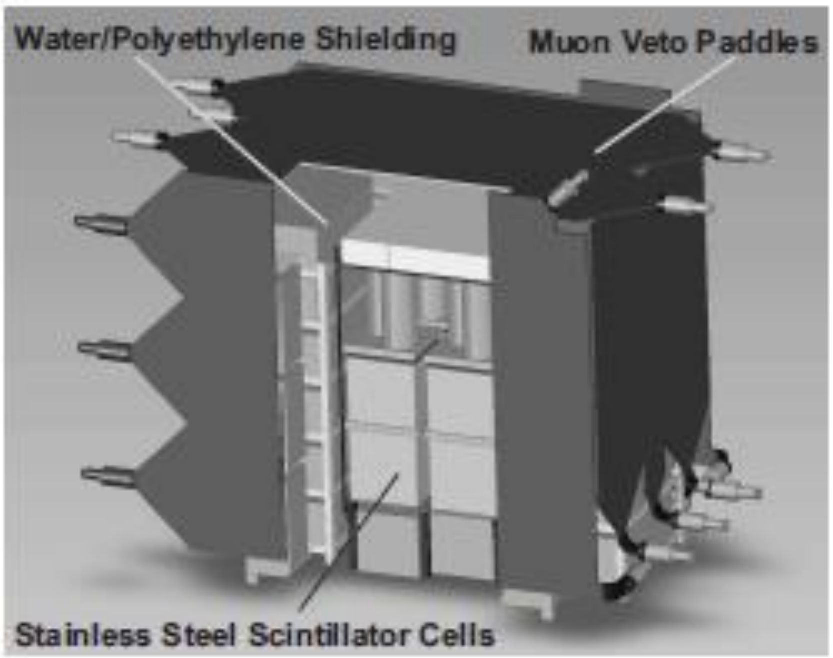


Observe more gammas > 3 MeV From ^{235}U than from ^{239}Pu

Remote monitoring of nuclear reactors

Reactor anti-neutrinos





$\overline{\nu}_e + p \rightarrow n + e^+$

Composition of fuel changes with time: ^{235}U is destroyed, ^{239}Pu is produced

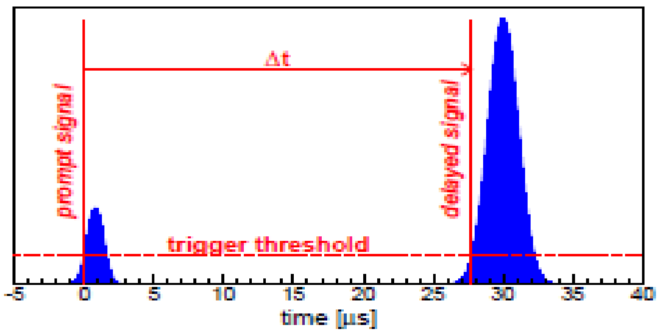
A. Bernstein *et al*.
SONGS reactor

Inverse Beta Decay

$$\boxed{\bar{\nu}_e + p \rightarrow e^+ + n}$$

- High cross section: $\sigma_{IBD} \approx 9.30 \cdot 10^{-42} \left(\frac{E_\nu}{10\,MeV}\right)^2 cm^2$
- Energy threshold: 1.8 MeV
- Coincident signal between positron and neutron, in space (*few cm*) and time (*several μs*)

$\Rightarrow$ *Clear event signature*



Prompt Event (p^+)

- Energy deposition of the kinetic energy of the e^+ (+1.022 MeV by annihilation)
- Energy strongly correlated to the neutrino energy

$\Rightarrow$ *Neutrino spectroscopy possible*

Delayed Event (n)

- Capture of the thermalized neutron on a nucleus A:

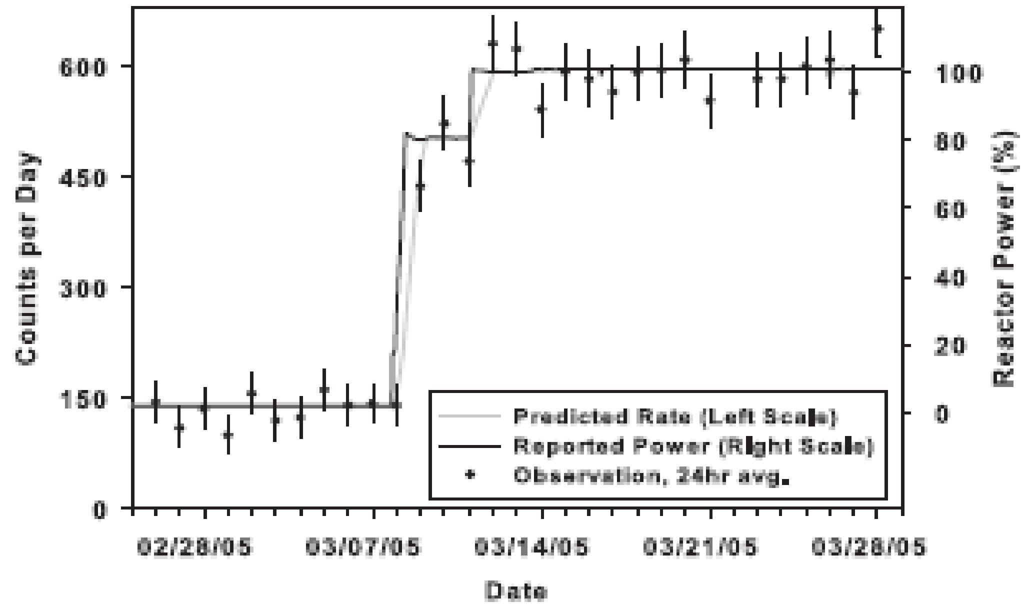
$$\boxed{n + A \rightarrow (A+1)^* \rightarrow (A+1) + \gamma}$$

- Subsequent γ-emission:

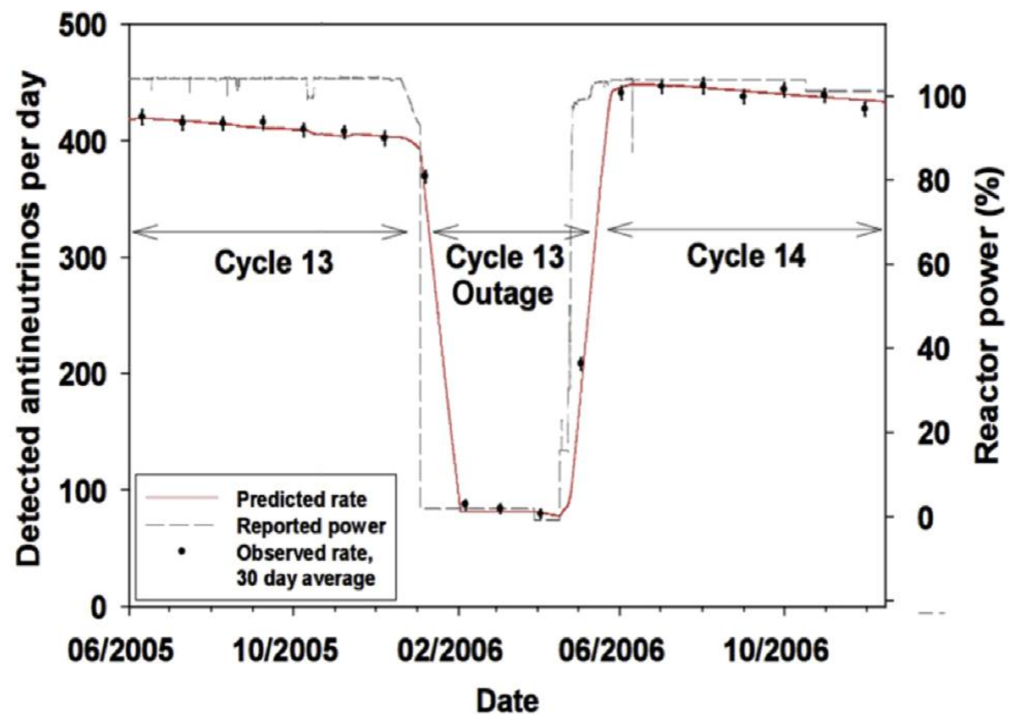
Hydrogen: $E_\gamma \approx 2.2$ MeV
Carbon: $E_\gamma \approx 4.9$ MeV
Gadolinium: $E_\gamma \approx 8$ MeV

M. Hofmann,
L. Oberauer,
S. Schonert

Observed neutrino counting rate before and after reactor turns on



Observed neutrino counting rate during two reactor fuel cycles



N S Bowden 2008 *J. Phys.: Conf. Ser.* ***136 022008***

The Surprisingly Large
Neutron Capture Cross Section of ^{88}Zr

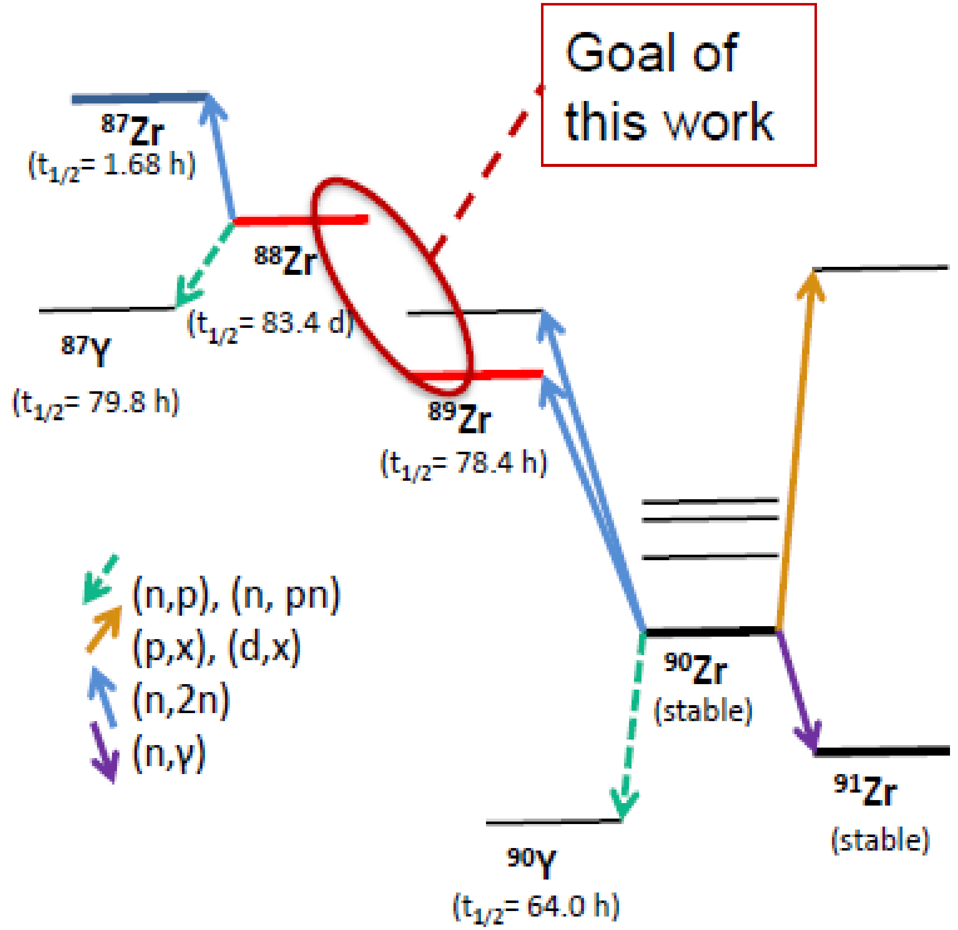
- Neutron-induced reactions are important for Stockpile Stewardship
  - **radchem diagnostics**: reaction networks involve many radioactive nuclei close to stability
  - **fission-product burn up**: key reactions far from stability influence resulting chain yields

- Nuclei heavier than iron are forged through neutron-capture reaction
  - **s process**: builds up elements along a path near stability over a timescale of thousands of years in asymptotic giant branch stars
  - **r process**: rapid neutron captures create very exotic nuclei in core-collapse supernova, neutron star mergers, or γ-ray bursts

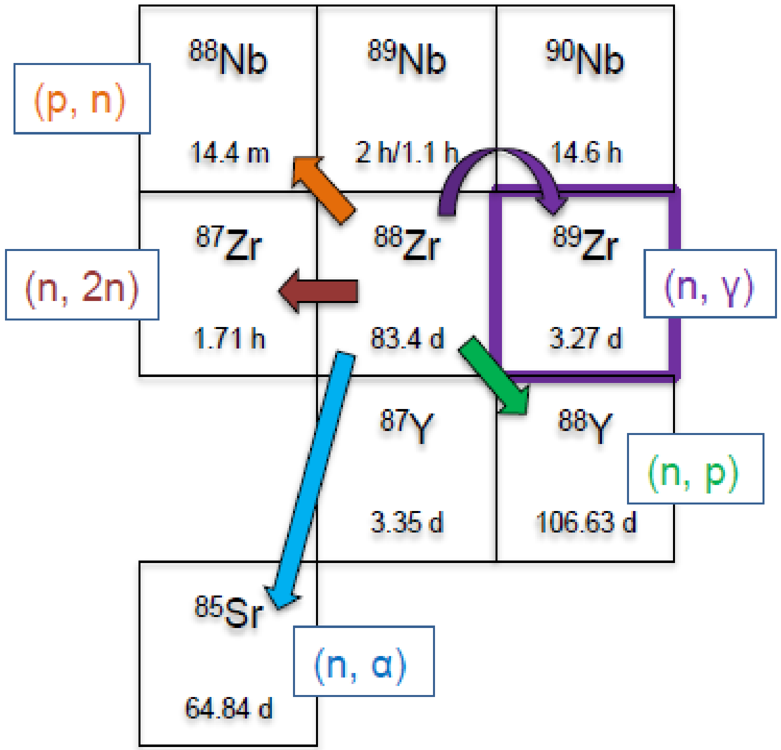
Measure missing cross section in Zr reaction network:

- Stockpile stewardship
  - Understanding reactions on radiochemical detector materials used in nuclear weapons

Zr reaction network with cross section data:

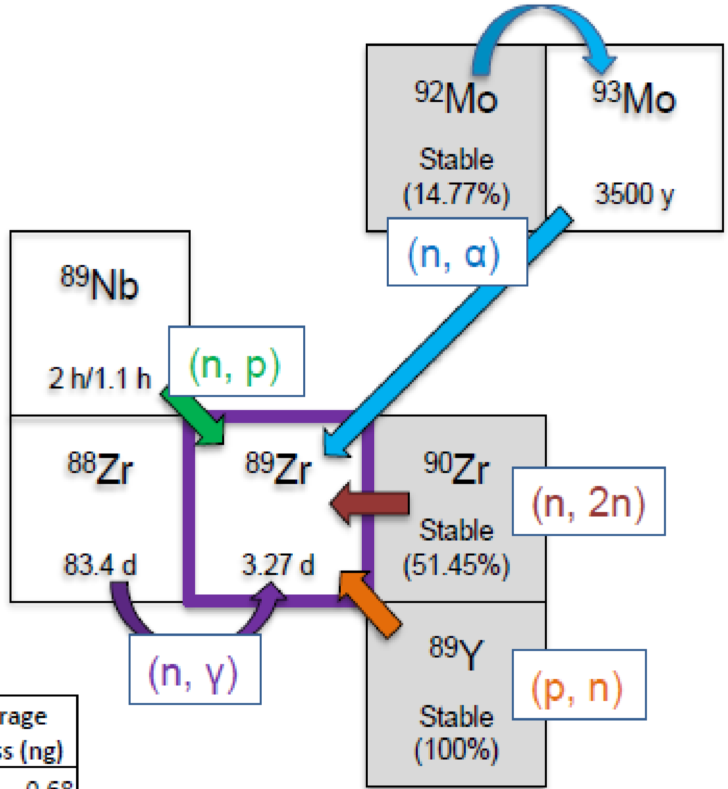


Destruction routes for ^{88}Zr in a reactor



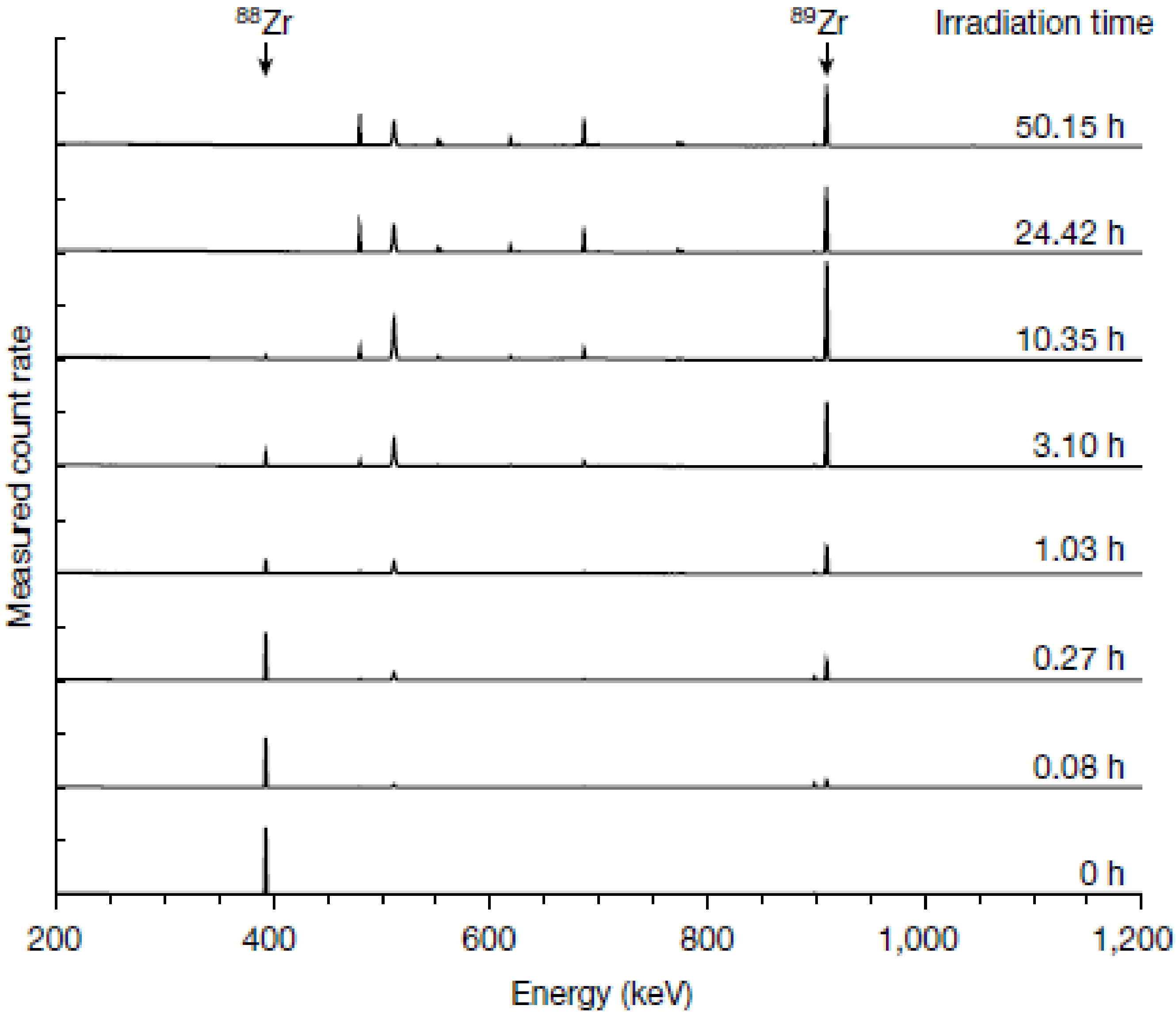
- ^{88}Zr(n,α)^{85}Sr
  - Will have to allow samples to decay to observe 514 keV ^{85}Sr
- ^{88}Zr(n,p)^{88}Y
  - $A(^{88}Y)_{meas} >> A(^{88}Y)_{decay}$
  - σ not well known but should be small
  - Will have to consider ^{88}Y(n,γ)^{89}Y which also has unknown σ
- ^{88}Zr(p,n)^{88}Nb
  - Need sufficient # few MeV protons
- ^{88}Zr(n,2n)^{87}Zr
  - Threshold > 12.5 MeV neutrons

Production routes for ^{89}Zr in a reactor

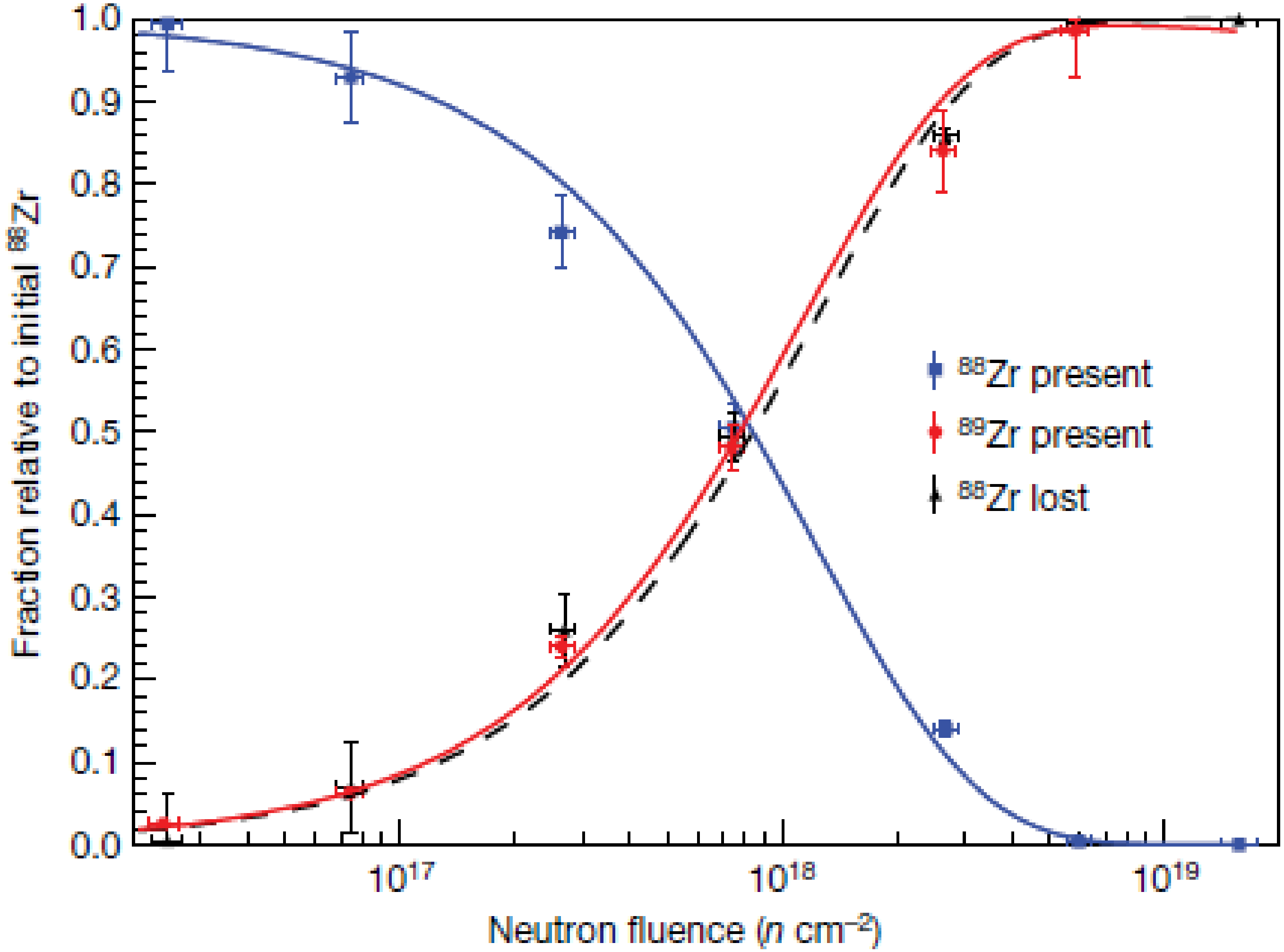


Element	Average Mass (ng)
Sr	0.68
Y	0.28
Zr	89.52
Nb	0.14
Mo	4.58

- ^{92}Mo(n,α)^{89}Zr
  - Mo isotopes have no been observed so far nor should primary beam make it to setup
- ^{89}Nb(n,p)^{89}Zr
  - No way to produce enough ^{89}Nb to get ^{89}Zr
- ^{89}Y(p,n)^{89}Zr
  - Need sufficient # protons of several MeV energy
  - Peak σ at ~16 MeV protons
- ^{90}Zr(n,2n)^{89}Zr
  - From fragmentation there shouldn't be ^{90}Zr in sample



Observed gamma ray spectra from ^{88}Zr targets as a function of irradiation time



Populations of 88Zr (blue squares) and 89Zr (red circles) as a function of neutron fluence)

We determine the thermal neutron capture cross section of ^{88}Zr to be

861,000 $\pm$ 69,000 barns

This is the second largest thermal neutron capture cross section ever measured. Only ^{135}Xe has a larger cross section.

J. A. Shusterman *et al.*
Nature 565 (2019) 328

Conclusion

Nuclear forensics plays an important role in many areas of science, engineering, and security.