Delayed Neutron Spectroscopy for the Characterization of Special Nuclear Material

TRTR Meeting, San Diego

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Today's Talk

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Obituary notice: LNT dead at 89 years, a life in the spotlight

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ABSTRACT

Considerable recent findings have revealed that the linear dose response for cancer risk assessment has not only outlived its utility in predicting risk but is based on a flawed scientific foundation. The present article characterizes this demise of a key concept of environmental risk assessment, in the framework of a figurative obituary of a long-lived concept that has poorly served society. This obituary is intended to illustrate an integrated mix of poignant and improper historical judgments that led to both the acceptance and ultimately the demise of this once intellectually facile and nearly universally accepted concept.

Today's Talk

- Delayed neutrons. Historical context.
- Brief review of delayed neutrons.
- Estimating % composition of mixed U-235 and Pu-239.
- New methods, better sensitivity.
- UCI's delayed neutron spectrometer.
- Some of our results.
- Summary and future prospects.

History

- 1950s: Keepin *et al. Phys. Rev.* **104** (1957) 1044. Six group data for U, Pu, Th.
- 1960s: Amiel, *Analytical Chemistry* **34** (1962) 1863. ^{235}U enrichment, Th content.
- 1970s: Routine use by USGS to survey uranium ore for ^{235}U content.
- 1980s: Rudstam, *Nuclear Science and Engineering* **80** (1982) 238. Many nuclides!
- 2013: Kapsimalis *et al. J. Radioanal. Nucl. Chem.* (2013) 1721. Estimate ^{235}U/^{239}Pu content by analysis of neutron counts *vs.* time.

Advantages of DNS

- Specific to fission, except for a couple of interferences like $^{17}\mathrm{O}(n,p)^{17}\mathrm{N}$.
- Sub-microgram sensitivity.
- Mostly free from activation γ-rays.
- Linear relationship to fissile material content over a wide range.
- Applicable with little or no sample preparation; non-destructive.
- Results available within minutes in favorable cases.

Drawbacks of DNS

- Relatively high neutron flux, as from a reactor, is needed for routine work.
- No energy measurement of the emitted neutrons is made.
- It is difficult to tell *what kind* of fissile material you've got.
- The sample is usually mechanically shuttled in and out of the high-flux region, and occasionally a capsule may rupture or crack, resulting in long delays.

IAEA Eight-Group Fractions

- Relative fractions of the delayed neutron precursors for the three fissile nuclides:

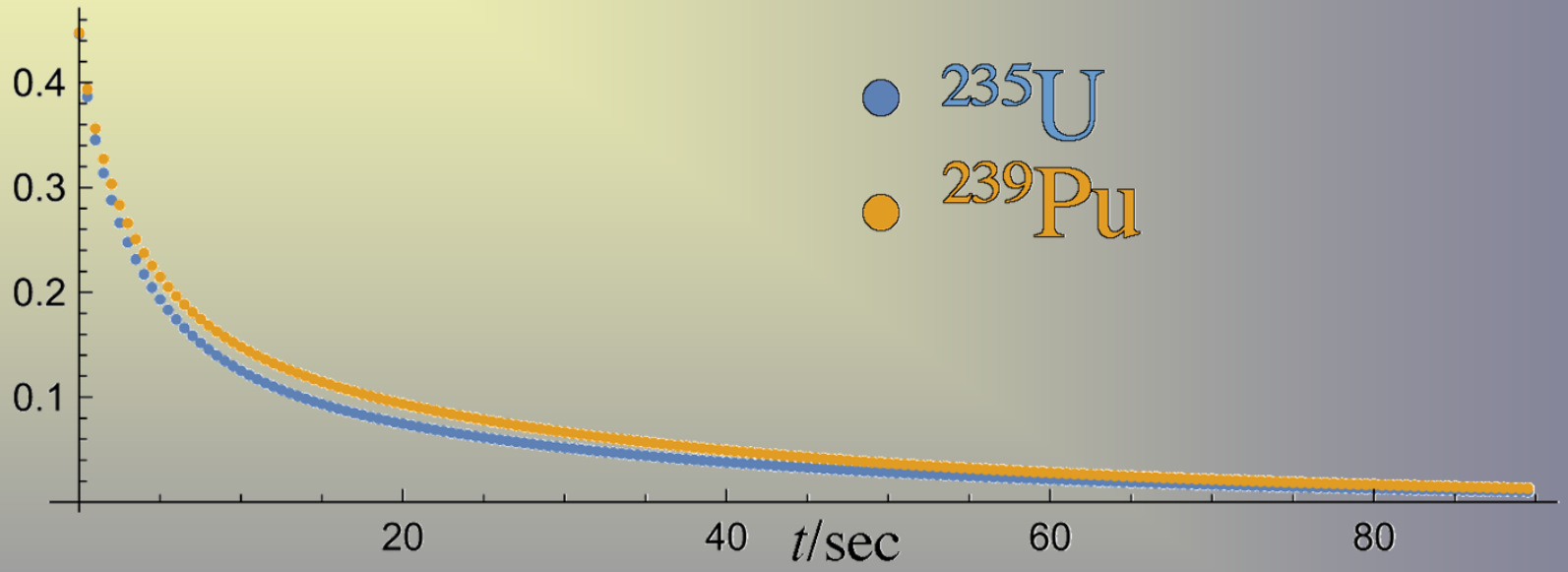
Group	Half-life/s	U-235	Pu-239	U-233
1	55.6	**0.0328**	**0.0319**	**0.0797**
2	24.5	**0.154**	**0.237**	0.167
3	16.3	0.0914	0.0826	0.150
4	5.21	0.197	0.182	0.200
5	2.37	**0.331**	**0.294**	0.298
6	1.04	0.0903	0.0816	0.0388
7	0.424	0.0812	0.0722	0.0560
8	0.195	0.0229	0.0185	0.0105

Basis Function Method

- As the group populations are slightly different, the plot of delayed neutrons detected versus time should differ.
- This is the clever basis function method of Kapsimalis *et al.* where the observed decay curve of a mixture is cast as a linear combination of the curve for ^{235}U and that for ^{239}Pu.
- In practice, the basis functions are measured, not calculated, but calculation of mock signals/mixtures is informative.

Basis Function Method

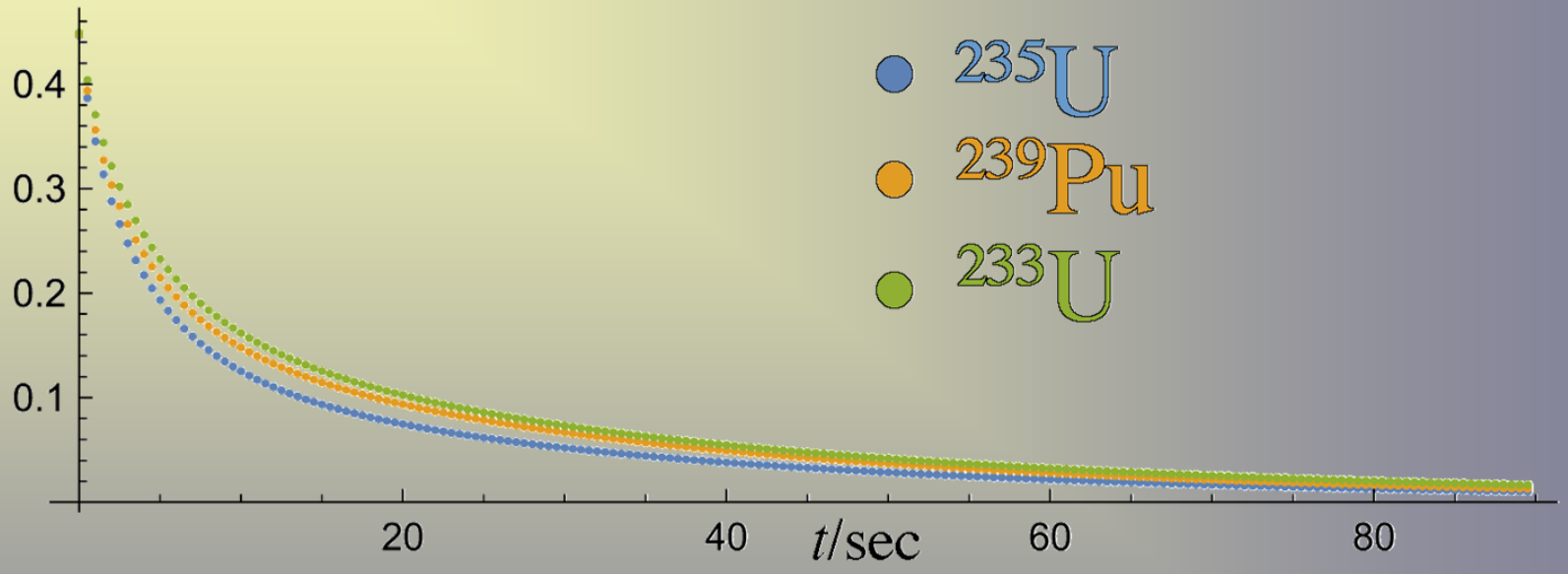
- Noiseless basis functions are still quite similar (90 sec irradiation).



- SVD analysis of the (N×2)•(2×1) = N×1 matrix problem reveals a condition number of about 30.

Basis Function Method

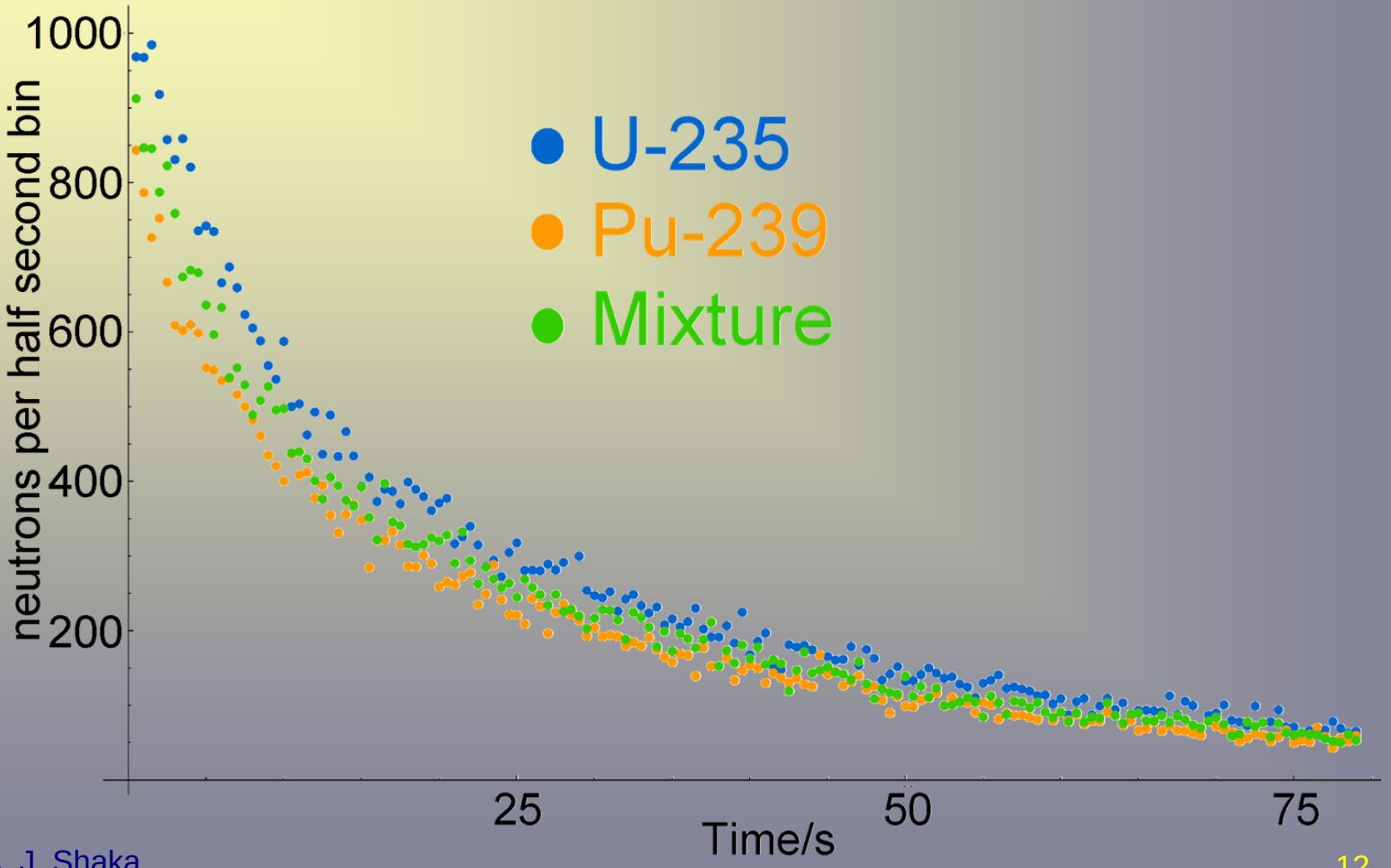
- More than two components makes the problem pretty ill-conditioned.



- SVD analysis of the $(N\times3)\bullet(3\times1) = N\times1$ matrix problem reveals a condition number of about 10^3.

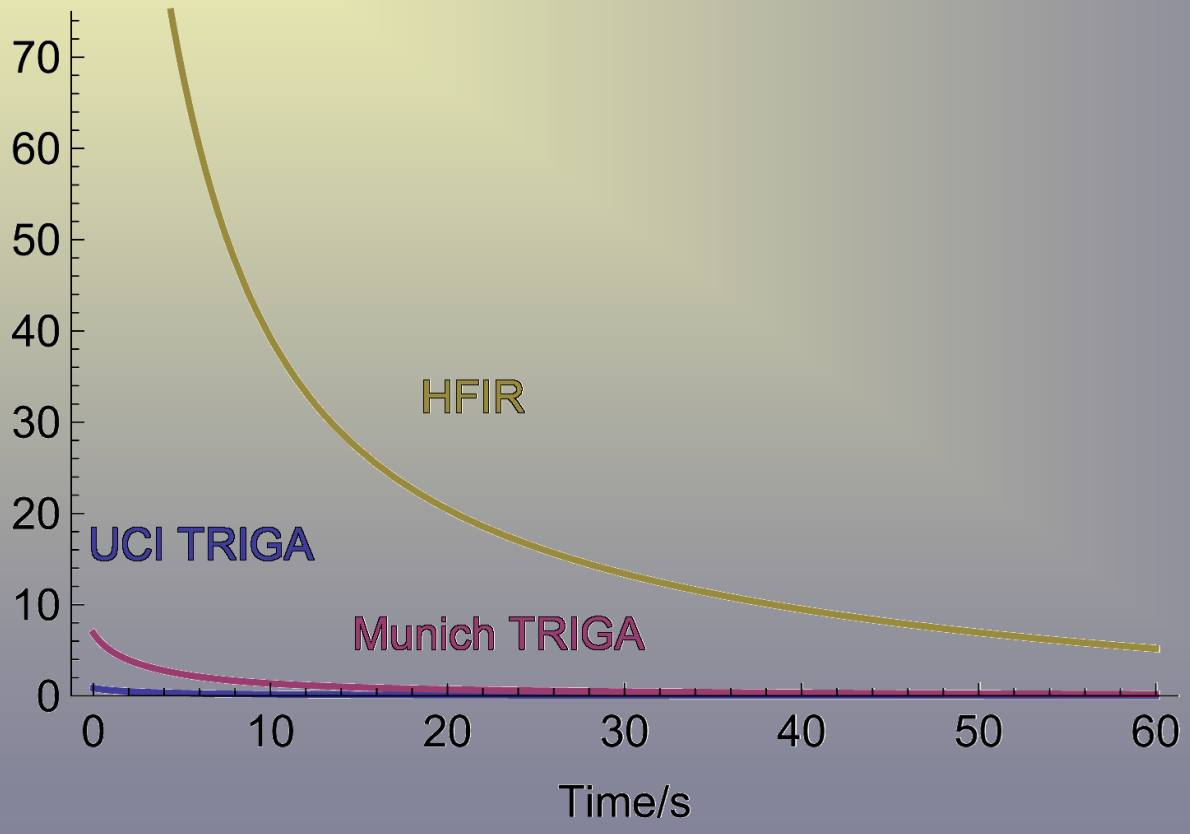
Basis Function Method

- Here's how it looks (for us), $1.6\ \mu g$ samples.



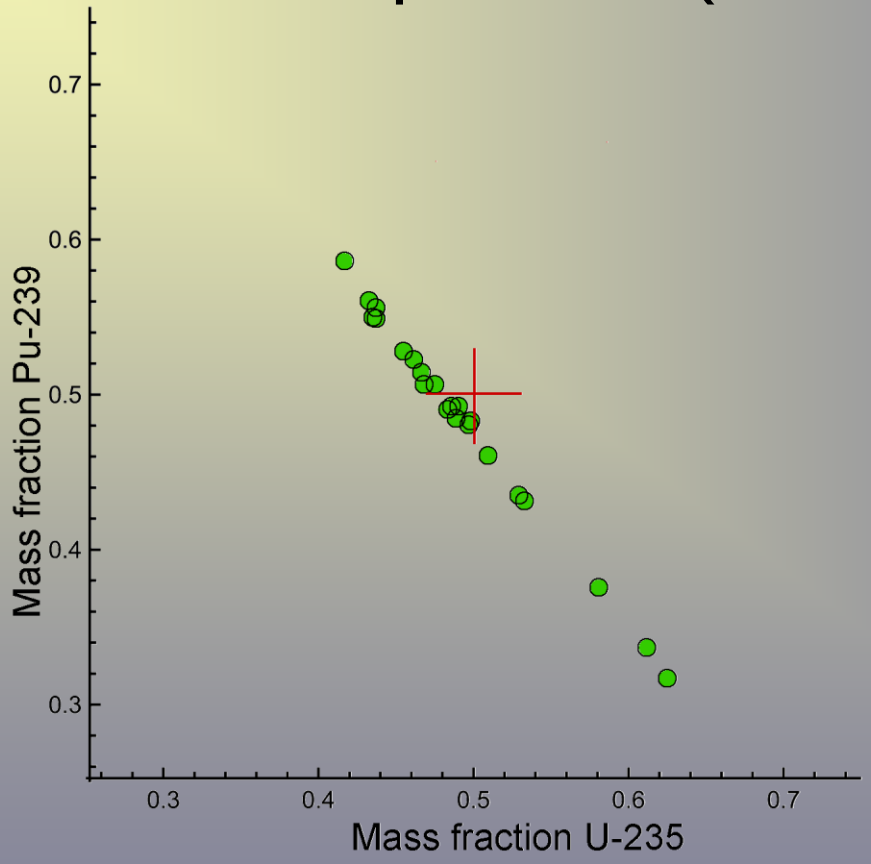
Basis Function Method

- We concluded that even composition to within 10% would be problematic.
- How did the ORNL team succeed?



Basis Function Method

- Triplicate measurements show the scatter we observed in composition ($3^3 = 27$).



Prior Work From Mainz

- Time series method known from 2002.

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Rapid determination of uranium and plutonium content in mixtures through measurement of the intensity–time curve of delayed neutrons

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Abstract

The delayed neutron emission of fissile isotopes after irradiation can be described by the six-groups theory. Due to the different half-lives and yields of the fission fragments, the intensity–time curve of delayed neutrons has an individual shape for different fissile isotopes. According to this characteristic, a new rapid analysis method has been developed to determine the concentration of different isotopes in a mixture, such as ^{235}U and ^{239}Pu, without any chemical separation. The intensity–time curve of delayed neutrons is measured by using the multichannel scanning (MCS) method with five ^{3}He-detectors installed on a fast transfer system at the research reactor Munich (FRM). This method was validated by about 200 measurements with different isotope contents even below the microgram region.

PACS: 23.90.+w; 28.41.Bm; 82.55.+e

Keywords: Delayed neutron; Fission material; Composition analysis

Prior Work From Mainz

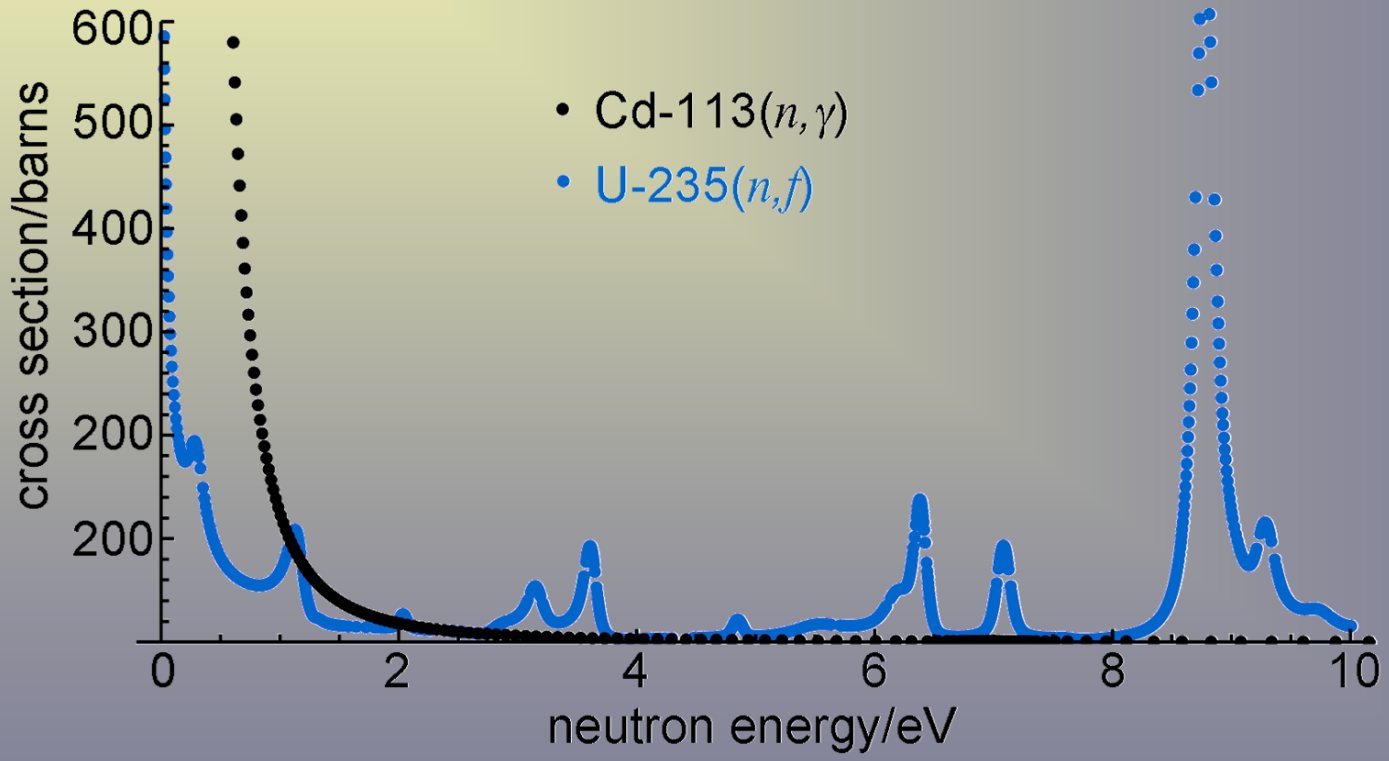
- Time series method known from 2002.

Uranium and thorium can be determined in a mixture by using a Cd-shielding without chemical separation [6–8]. This is based on the different neutron energy thresholds for fission and two irradiations, one with and one without Cd-shielding, are needed.

For a mixture of ^{235}U and ^{239}Pu, such a threshold of neutron energy does not exist. Both undergo fission when they interact with thermal neutrons. The method with Cd-shielding as in the determination of U-Th cannot be applied. A purely instrumental method with the total counting of delayed neutrons seems generally impossible, if the chemical separation is not performed...

Neutron Filtering

- ^{113}Cd is commonly used to attenuate thermal neutrons.
- However, it has a *differential effect on the rate of fission, too*.



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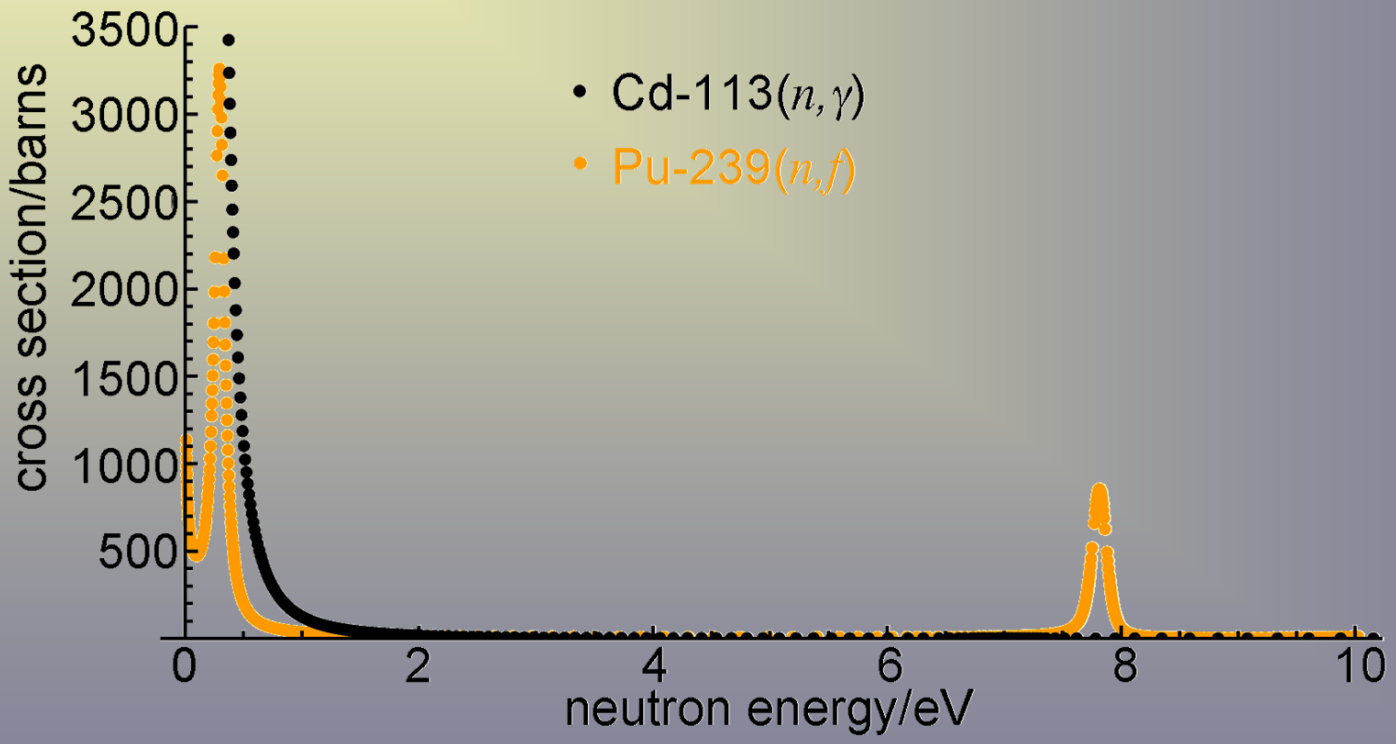

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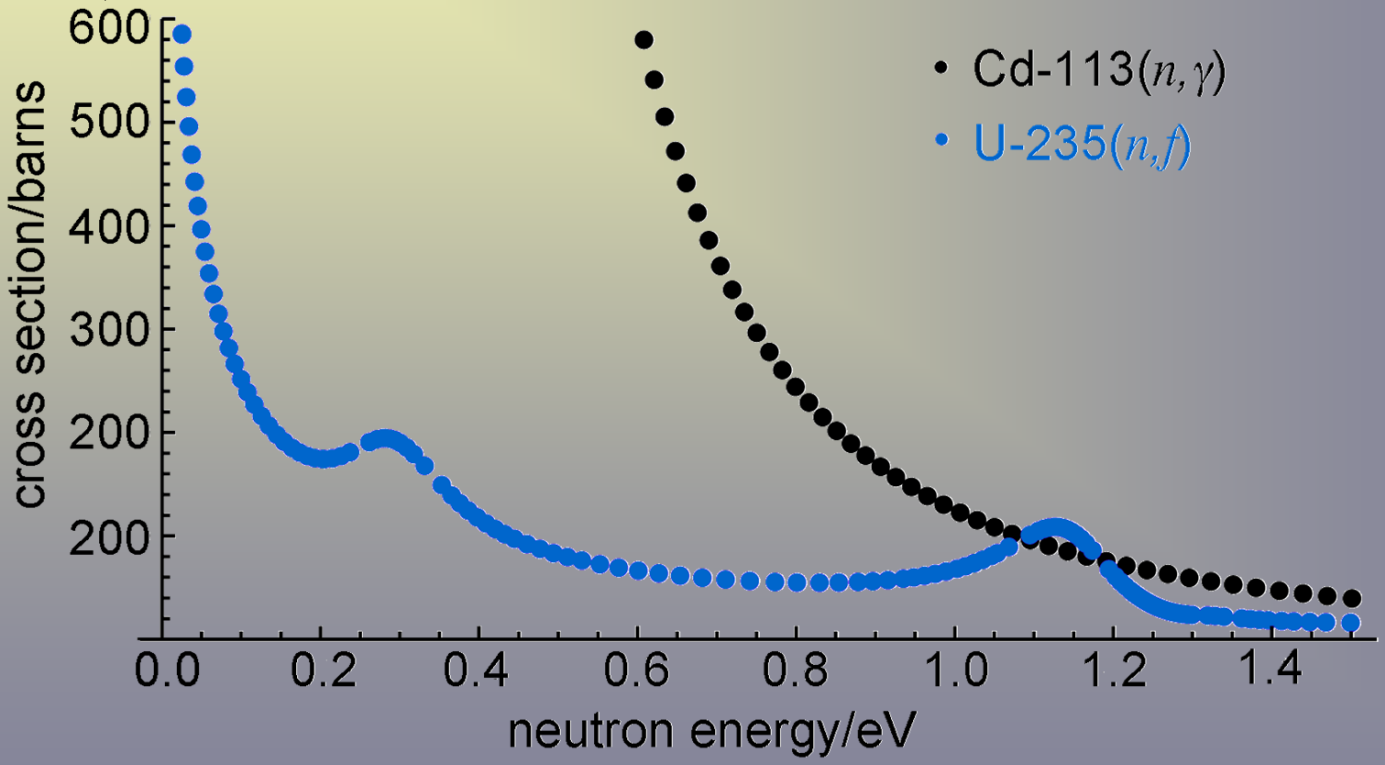

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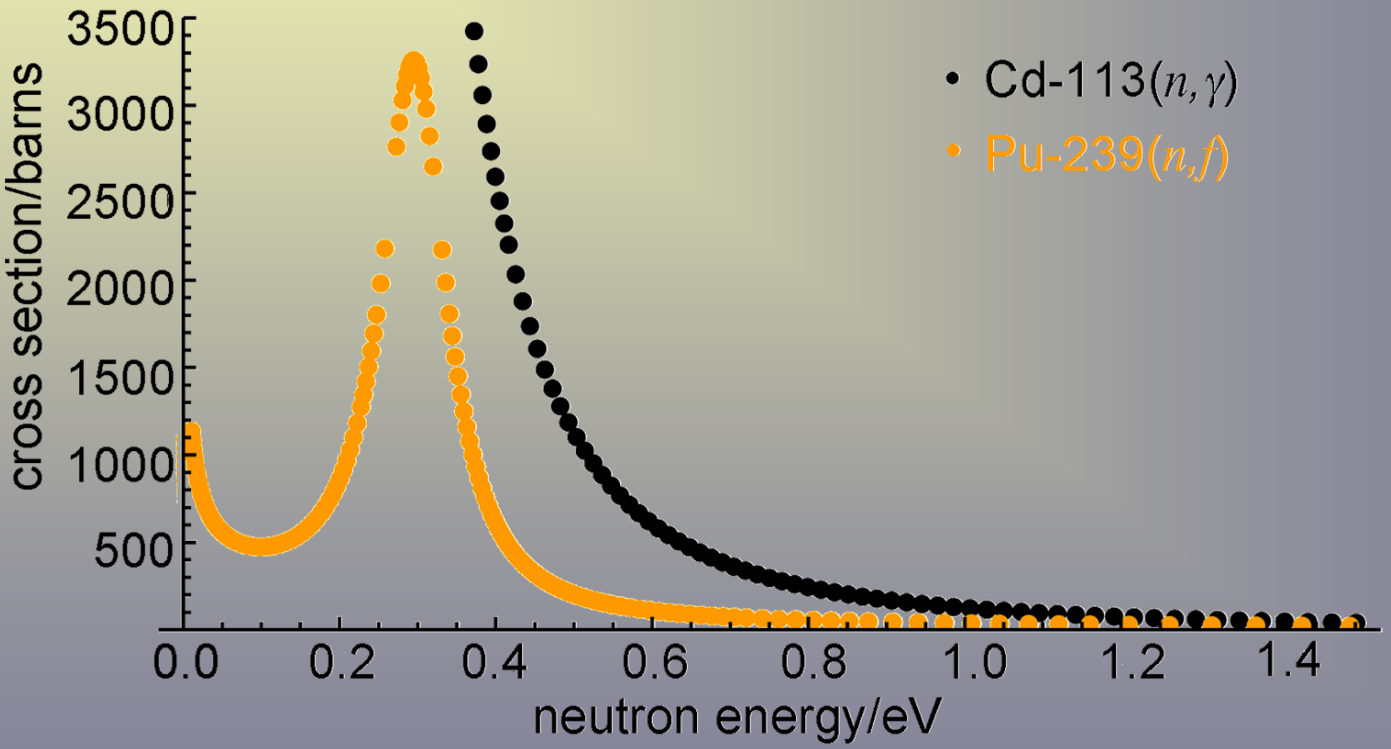


Neutron Filtering

- $^{113}\mathrm{Cd}$ cuts both rates of fission way down.
- But it cuts $^{239}\mathrm{Pu}$ rather more.
- If the reduction is r_U and r_{Pu} and the counts we get in the two conditions (bare) and Cd-shielded are c_b and c_s for the mixture, then

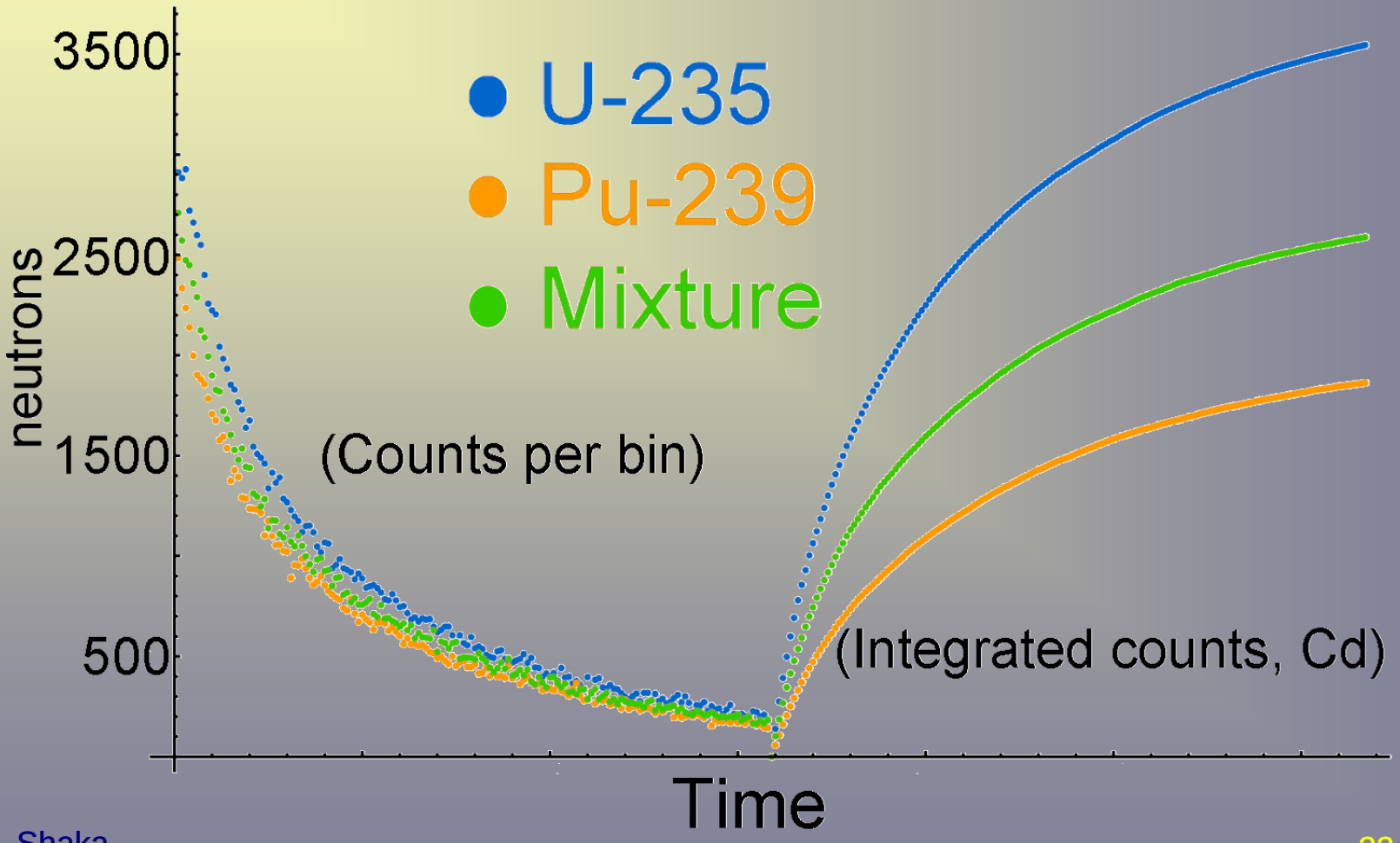
$$c_b = xc_{bU} + yc_{bPu}$$

$$c_s = xr_Uc_{bU} + yr_{Pu}c_{bPu}$$

- After measuring the Cd-knockdown for uranium, plutonium (standards) and the mixture, we simply solve two equations and two unknowns.
- The system is well conditioned if r_U and r_{Pu} differ.

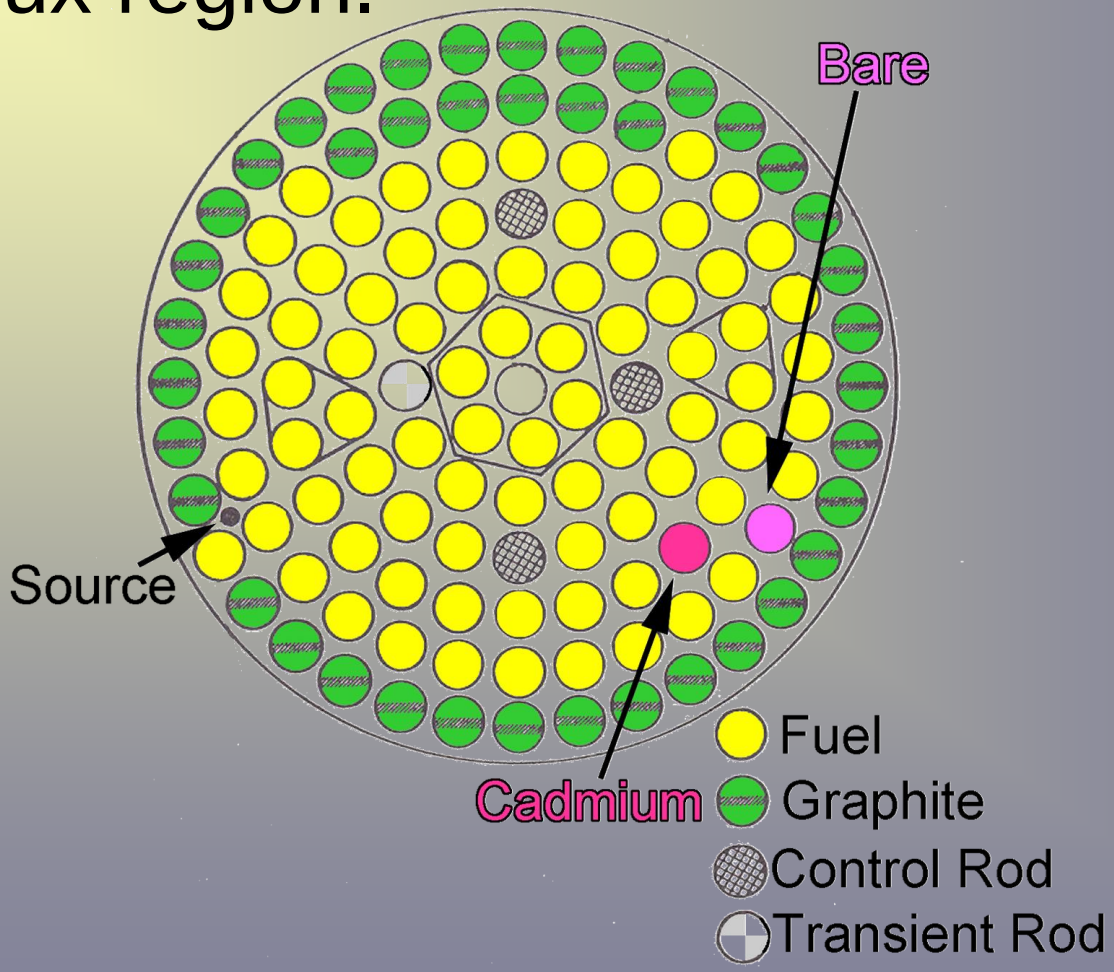
Neutron Filtering

- Changing the rate of production of delayed neutron precursors is easier to discern.



Neutron Filtering

- In our core the Cd terminus is in a slightly higher flux region.

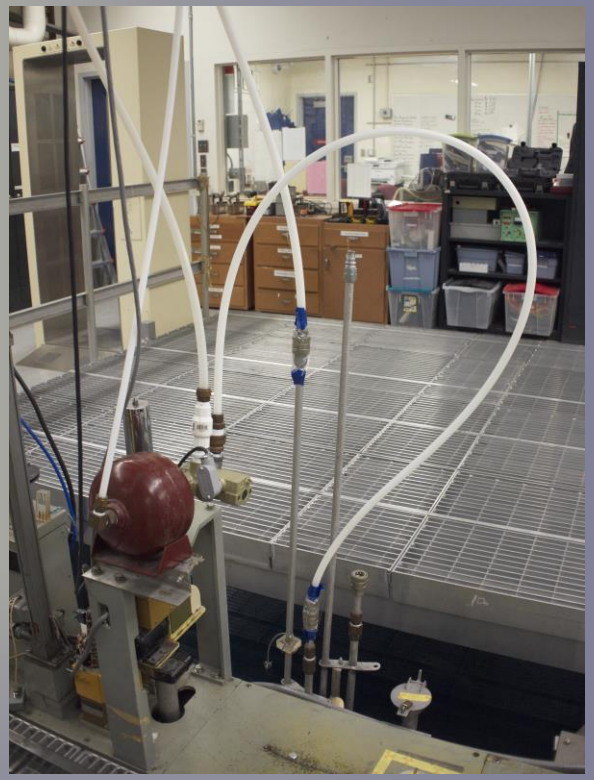


Neutron Filtering

- In practice we see reductions of about 0.24 to 0.25 for $^{235}\mathrm{U}$ and 0.16 to 0.18 for $^{239}\mathrm{Pu}$ depending on power levels, cooling, other samples running, *etc.*
- We can simply integrate up all the neutrons; their temporal signature is irrelevant for this method.
- Once we have enough neutrons in the 0.16 to 0.24 range, we can estimate the accuracy of the method.
- We run the standards at the initiation and at the conclusion of each experiment.
- How high-tech is this?

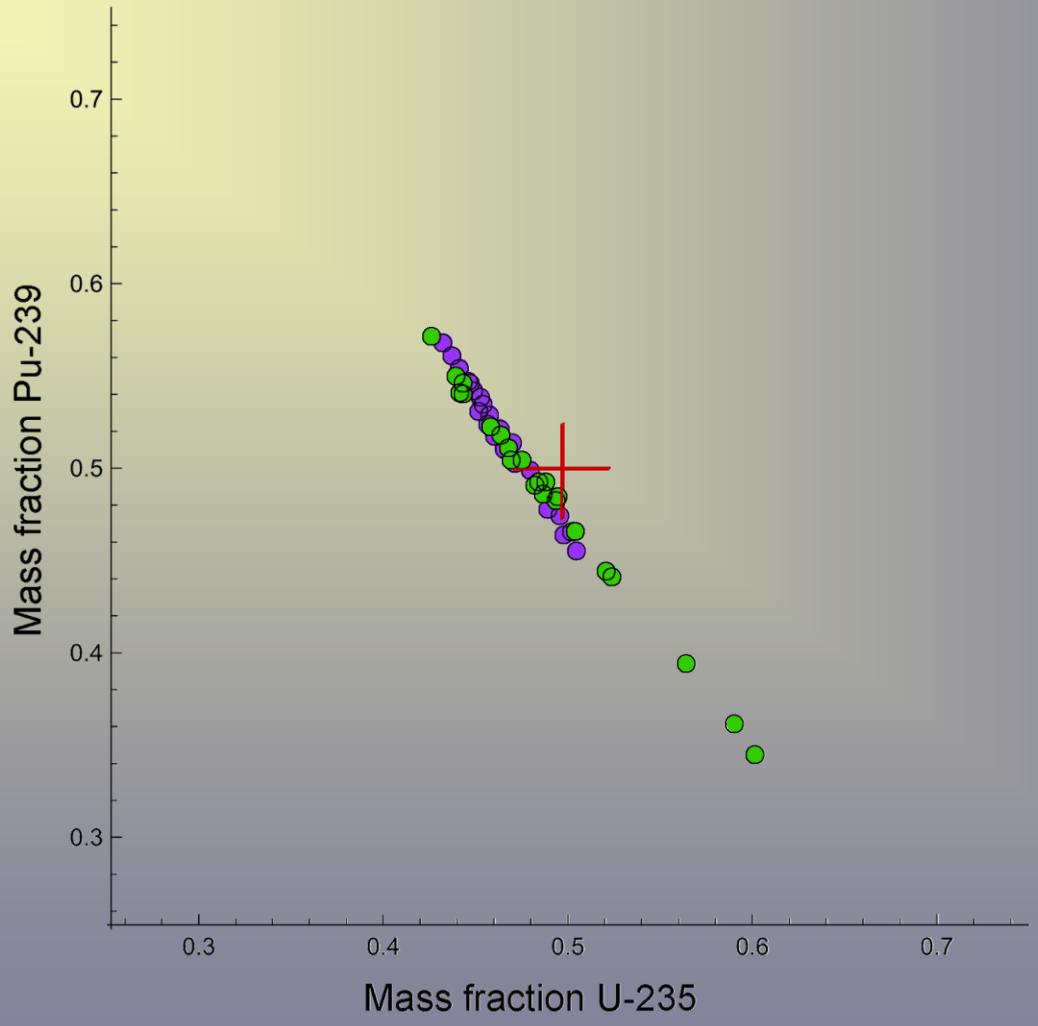
UCI Delayed Neutron System

- Lower flux, mediocre efficiency, and mechanical reliability have been issues for us.



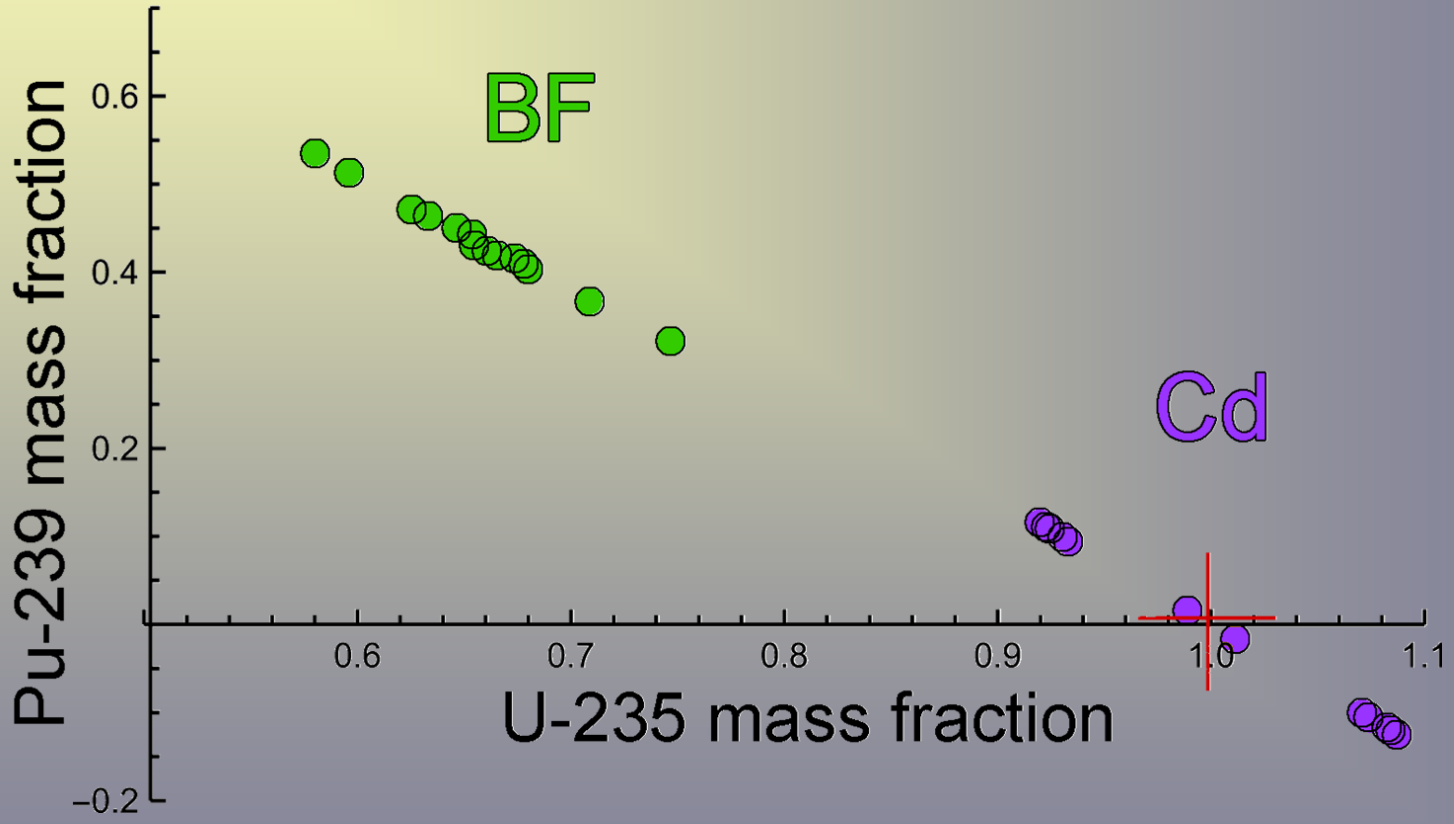
Comparison

- With Cd measurement, scatter is less.



Comparison

- But for “pure” samples there is rather more difference.



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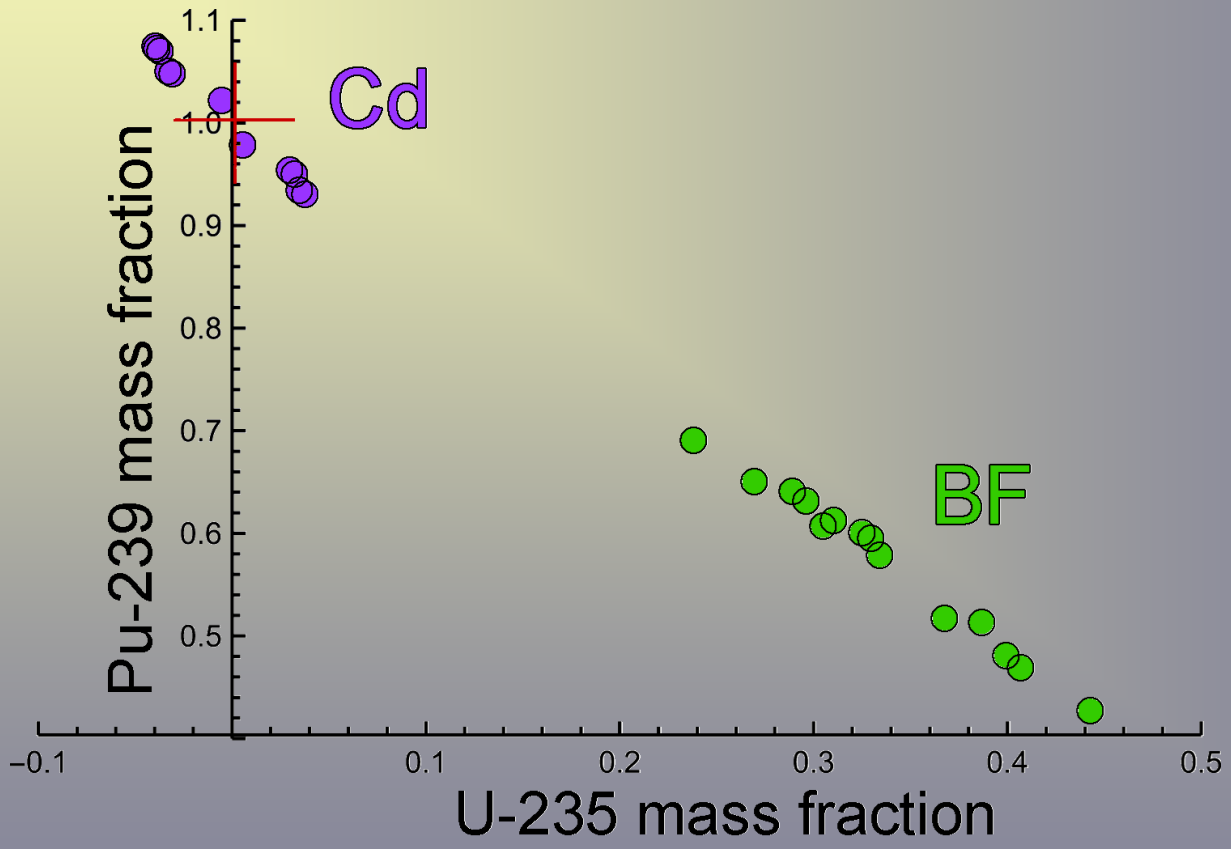

Summary

- Even with very modest flux, detector efficiency, *etc.*, it is possible to estimate U/Pu ratios if one is determined.
- It might even be possible with a portable neutron generator.
- The Cd method seems fairly tolerant of experimental conditions and variations of them.
- We had to emerge from “The Swamp” but we seem to be on dry land now.
- We have enjoyed all the twists and turns!

Acknowledgments

- Dr. Bruce Mincher (INL) supplied us with a small amount of plutonium and verified its authenticity.
- The Beall Foundation supported some aspects of the work that might lead to applications to used fuel recycling or MOX fuel.
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- Thanks for listening!