LA-UR- 11-06427

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Title: Quantifying Fissile Content in Spent Nuclear Fuel Using 252Cf Interrogation with Prompt Neutron Detection

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Intended for: Presentation at the Global 2011 conference Makuhari, Japan, Dec 11-15, 2011

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Quantifying Fissile Content in Spent Nuclear Fuel Using ^{252}Cf Interrogation with Prompt Neutron Detection

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Abstract

The majority of the plutonium on earth is stored in spent nuclear fuel assemblies (FAs). Presently, there is no means for directly measuring the mass of the plutonium in these assemblies by nondestructive assay (NDA). Researchers at LANL have been coordinating a multi-laboratory effort to quantify the capability of 14 NDA techniques for the purpose of combining a subset of these techniques into a system that can directly measure the isotopic Pu mass. ^{252}Cf Interrogation with Prompt Neutron (CIPN) detection is one of the 14 proposed NDA techniques, and it shows promise of quantifying fissile content in spent FAs. CIPN is a relatively low-cost and portable instrument, and it looks like a modified fork detector combined with an active interrogation source. Fission chambers (FCs) were chosen as the neutron detectors because of their insensitivity to γ radiation. The CIPN assay comprises two measurements, a background count and an active count, without and with the ^{252}Cf source next to the fuel respectively. The net signal above background is primarily due to the multiplication of Cf source neutrons caused by the fissile content. It is almost uniformly sensitive to diversions at different locations aross the assembly. A 100-μg ^{252}Cf source was proven strong enough to provide sufficiently high signal above background. The concept of $^{239}Pu_{eff\_CIPN}$ was introduced to represent the three major fissile isotopes in a single term. Burnup (BU) and cooling time (CT) corrections were introduced to $^{239}Pu_{eff\_CIPN}$ to account for the neutron absorption caused by different neutron absorbers. The results shows that there exists a coherent universal relation between CIPN count rate and "corrected fissile content". With the schemes presented in this paper, together with given BU and CT (or quantified using other techniques), the fissile content of the target spent FA (or $^{239}Pu_{eff\_CIPN}$) can be determined within a few percent.

Keywords: fissile content, spent fuel, plutonium, nuclear safeguards, CIPN, NGSI, MCNP, NDA

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Quantifying Fissile Content in Spent Nuclear Fuel Assemblies Using ^{252}Cf Interrogation with Prompt Neutron (CIPN) Detection

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Outline

- ❖ Overview of NGSI Spent Fuel Project
- ❖ CIPN principle and design
- ❖ CIPN signature, statistical uncertainty
- ❖ Detection of diversion
- ❖ Determination of fissile mass
- ❖ Conceptual experiment setup
- ❖ Summary

Why measure Pu in spent fuel?

- Independently verify the mass of Pu
- Re-verification following a loss of continuity of knowledge
- Determinate the input accountability mass reprocessing facility
- Shipper/receiver difference
- Determine mass of non-self-protecting assemblies
- Tangent - enable "finger printing" to assure what leaves one facility arrives at another – no need to quantify mass

Overview of NGSI Spent Fuel project

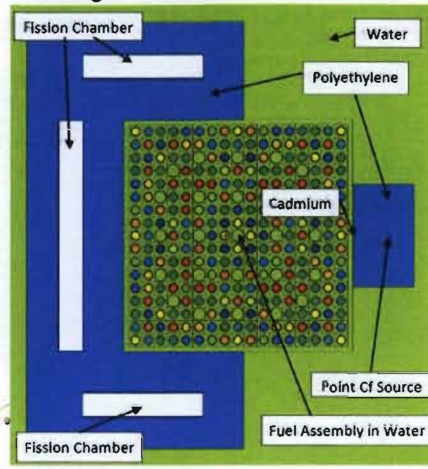
- Next Generation Safeguard Initiative (NGSI) Spent Fuel project is multi-million dollar, multi-institute (including international collaborators), 5+ year project sponsored by US DOE.
- Determine the Pu mass in Spent Fuel Assemblies ... and detect the absence of Pu mass (detect diversions)
- 14 Nondestructive Assay (NDA) techniques were studied. Complete list follows.
- Down-selections were performed in summer 2011, and four Integrated NDA systems (including CIPN) were chosen to move forward.

14 NDA Techniques were investigated:

^{252}Cf Interrogation with Prompt Neutron Detection	Neutron Resonance Transmission Analysis (time of flights)
Delayed Gamma	Nuclear Resonance Fluorescence
Delayed Neutrons	Passive Prompt Gamma
Differential Die-Away	Passive Neutron Albedo Reactivity
Differential Die-Away Self-Interrogation	Self-integration Neutron Resonance Densitometry
Lead Slowing Down Spectrometer	Total Neutron (Gross Neutron)
Neutron Multiplicity	Passive X-Ray Fluorescence

CIPN design

- Quantify the fissile content in the sample by quantifying multiplication using a ^{252}Cf source

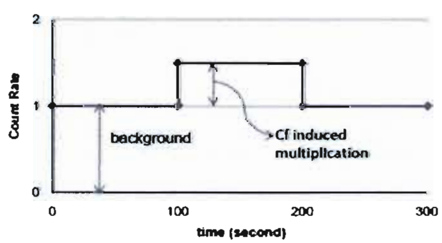


- Fission chambers (FC)
  - Thin layer of 93% enriched ^{235}U
  - 1" diameter
- Cadmium liner
  - Between ^{252}Cf and spent fuel assembly (SFA) for uniformity
  - Outside wall of U-shaped poly block (minimize background signal)

CIPN principle

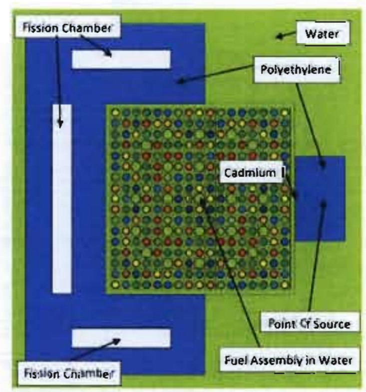
I. BACKGROUND - passive neutron count rate from the SFA

II. ACTIVE - neutron count rate measured in presence of ^{252}Cf source

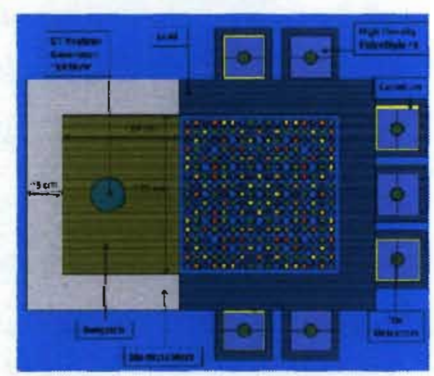


- NET SIGNAL = ACTIVE – BACKGROUND → multiplication in the SFA induced by Cf neutrons→ fissile content

CIPN with DT generator

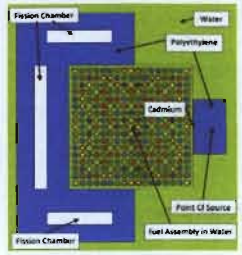


Delayed Neutron Instrument



CIPN design goals/optimization

OPTIMIZATION:

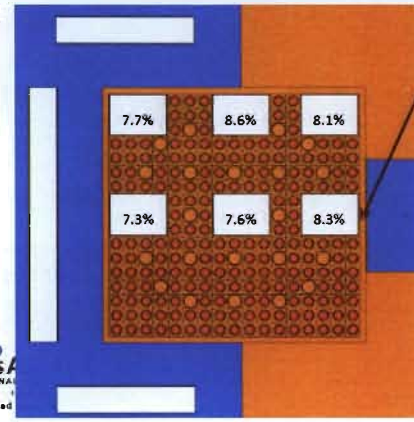


- Source strength, detector design and efficiency
  - *Key design factors under control*
  - Hundreds of MCNPX cases run to optimize detector design
- FC arrangement/length/position optimized for uniform sensitivity across SFA
- Thickness of polyethylene optimized to maximize count rate
- ^{252}Cf interrogating source
  - Optimized for broad range of SFAs
  - Optimized position for sufficient neutron moderation and large solid angle

CIPN uniformity

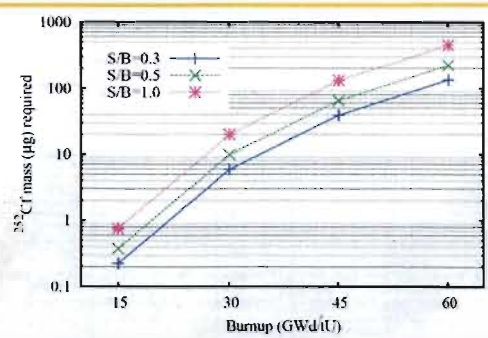
- 1.0 mm thin cadmium sheet inserted between the SFA and the source polyethylene block
  - Increase uniformity

Cd sheet



- Relative count rate change (%) when 12 fuel rods (~4.5% mass) replaced by DU (0.2% ^{235}U) in each zone

^{252}Cf source strength



- 60 μg ^{252}Cf source corresponds to S/B of 50% for a 45 GWd/tU BU case
- ^{252}Cf half-life = 2.6 yr (100 μg corresponds to 2.34×10^8 n/s)
  - 200 μg needed at the start of operation for ~5 years of operation
  - Largest ^{252}Cf commercially available = 10,000 μg

CIPN signature

CIPN signature

- Multiplication in SFA in presence of ^{252}Cf source

sources of neutrons:

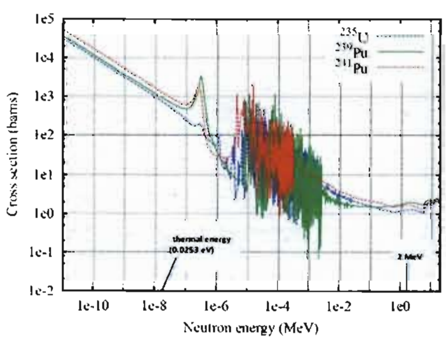
- Background
  - ➢ Spontaneous fission of transuranic isotopes (^{244}Cm, ^{242}Cm, ^{240}Pu…)
  - ➢ (α, n) reaction with light elements (^{18}O)
- Active ^{252}Cf interrogation
  - ➢ Induced fissions by ^{252}Cf neutrons
  - ➢ Direct detection of source neutrons….small in water and predictable
  - ➢ *Net count rate - after subtracting of the background, the measured signal dominated by neutrons produced from the chain of multiplication caused by the fissile content*

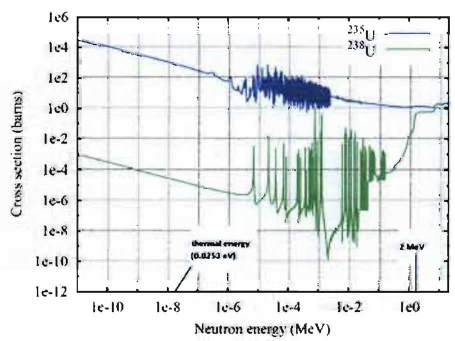
CIPN signature (cont'd)

Balance between induced fissions and capture

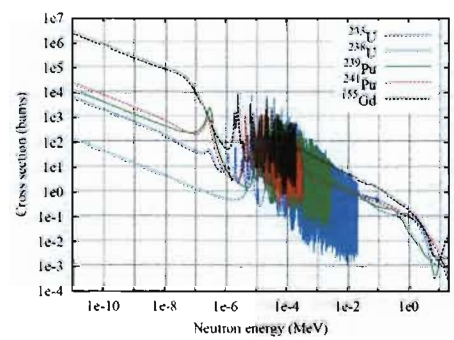
- Induced fission
  - ➢ Main contributing isotopes: ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu
  - ➢ NGSI spent fuel effort … to quantify the elemental Pu mass
  - ➢ Necessary to minimize the contribution of ^{238}U or to quantify and separate it from the Pu contribution
- Capture
  - ➢ Neutron flux in SFA reduced by isotopes that absorb neutrons
  - ➢ Main contributors – fissile isotopes ^{235}U, ^{239}Pu and ^{241}Pu, actinides such as ^{240}Pu and ^{238}U; fission fragments Gd, Nd, and Sm;

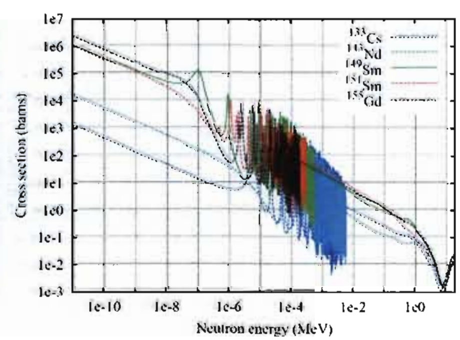
CIPN signature – fission content



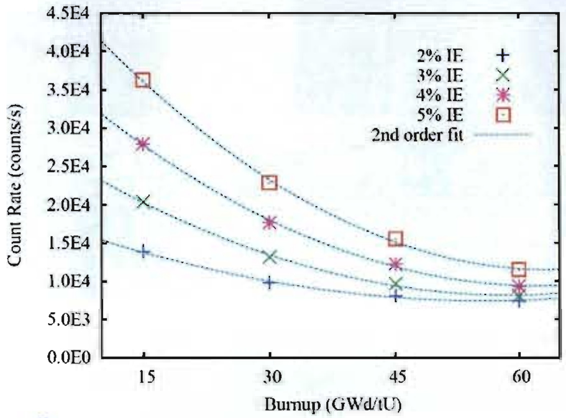


CIPN signature – absorbers



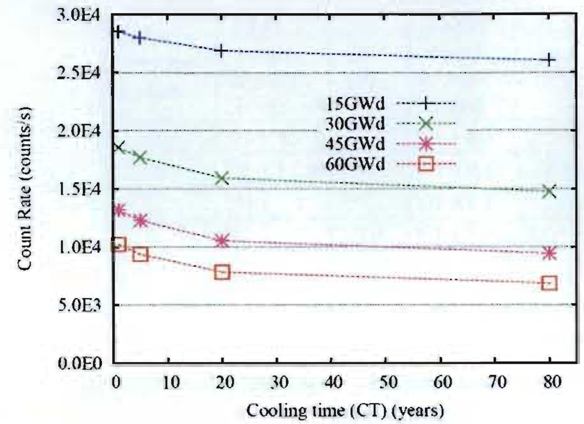


CIPN signature vs SFA parameters (BU)



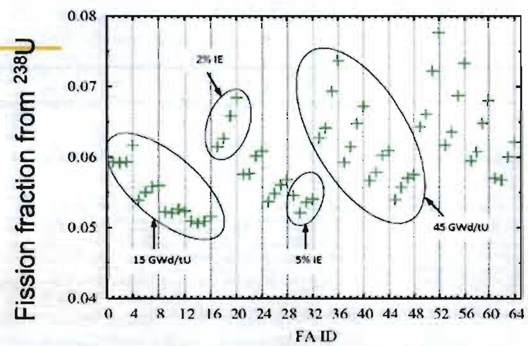
- Fissile content decreases with higher BU
  - ➤ lower fissile content - ^{235}U reduces
  - ➤ greater neutron absorber mass - increase of Pu isotope
- Count rate increases with IE

CIPN signature vs SFA parameters (CT)



- Count rate decreases with longer CT, but to a lesser degree than the variation with BU and IE
  - ➤ ^{241}Pu decays (14 yr)
  - ➤ ^{241}Am (14 yr) and ^{155}Gd (5 yr) increase

CIPN signature – contribution of ^{238}U



- Neutron contribution from ^{238}U ~ 5% to 8% among all investigated SFAs
  - ➤ *only ~10% of ^{238}U fission directly caused by Cf source, so ~90% of them associated with multiplication of fissile isotopes*
  - ➤ *^{238}U contribution treated as a background component*

CIPN signature - uncertainties

statistical

- ➤ Measurement time – optimize to achieve desired precision

$$S = A - B \qquad \sigma_S^2 = \sigma_A^2 + \sigma_B^2 + \sigma_{S\_MCNPX}^2.$$

- ➤ Electronics stability ~ 0.05%
- ➤ *MCNPX ~ 0.3% in this work (can be reduced)*

systematic

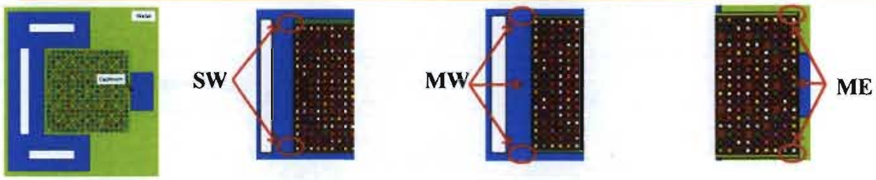
- ➤ Likely dominant contributor to final uncertainty
- ➤ Fuel assembly position
- ➤ uncertainty in the source rate
- ➤ uncertainty in ^{238}U and direct source neutron contribution

CIPN signature – statistical uncertainties

BU (GWd /tU)	Bgd Count Rate, B (cts/s)	σ_B (%)	Active Count Rate, A (cts/s)	σ_A (%)	Net Count Rate, S (cts/s)	σ_S w/o MCNP (%)
15	2.9 x 10^2	0.59	3.8 x 10^4	0.05	3.8 x 10^4	0.05
30	4.8 x 10^3	0.14	2.9 x 10^4	0.06	2.4 x 10^4	0.08
45	2.2 x 10^4	0.07	3.9 x 10^4	0.05	1.7 x 10^4	0.15
60	5.8 x 10^4	0.04	7.1 x 10^4	0.04	1.3 x 10^4	0.28

- The uncertainty in the signal (s_s) stays below 0.3 % for all assemblies for count time of 100 s
  - ➢ Measurement precision limited by systematic uncertainty

CIPN signature – systematic uncertainties

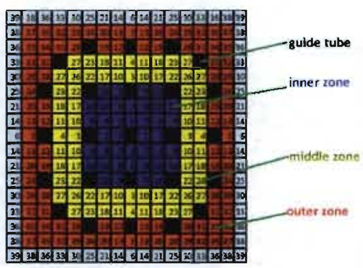


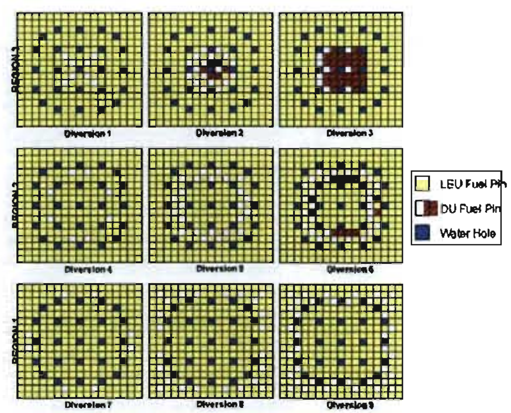
case	15GWd/tU		45GWd/tU	
	CR (cts/s)	change, %	CR (cts/s)	change, %
CENTER	3.77E4	--	1.70E4	--
SW	3.69E4	-1.9 ±0.3	1.67E4	-1.5 ±0.4
MW	3.70E4	-1.6 ±0.3	1.66E4	-2.2 ±0.4
ME	3.86E4	2.6 ±0.3	1.71E4	0.5 ±0.4

- Uncertainties of up to 2.6% between independent measurements of the same SFA
- **Solutions:** 1) Engineering constraints to fix SFA position
  2) Ratio to other integrated techniques

Detection of diversion

CIPN – detection of diversion





- 8, 24 and 40 rods from each zone replaced by depleted uranium (0.2% ^{235}U)

CIPN – detection of diversion

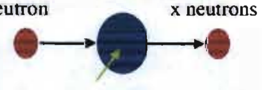
case ID	location of diverted rods	# rods diverted	% mass diverted	15GWd/tU, (%)	30GWd/tU, (%)	45GWd/tU, (%)
1	inner zone	8	3.0	-4.6 ±0.30	-3.2 ±0.27	-2.4 ±0.32
2		24	9.1	-15.4 ±0.28	-10.1 ±0.29	-8.0 ±0.32
3		40	15.2	-27.5 ±0.28	-19.4 ±0.28	-14.1 ±0.31
4	middle zone	8	3.0	-3.8 ±0.30	-2.8 ±0.31	-2.0 ±0.32
5		24	9.1	-11.6 ±0.29	-7.8 ±0.30	-6.1 ±0.32
6		40	15.2	-20.0 ±0.28	-14.1 ±0.26	-11.0 ±0.32
7	outer zone	8	3.0	-4.1 ±0.30	-2.7 ±0.31	-2.4 ±0.33
8		24	9.1	-9.8 ±0.29	-7.2 ±0.30	-5.9 ±0.32
9		40	15.2	-17.3 ±0.28	-12.8 ±0.26	-10.4 ±0.32

- *The diversion of at least 8 fuel rods can be detected if the count rate of the full case is known*
  - Case of no reference measurement needs future research

Determination of fissile content

CIPN – determination of fissile content

$$^{239}Pu_{eff,CIPN} \equiv C_1\,^{235}U_m + ^{239}Pu_m + C_2\,^{241}Pu_m.$$



$$x = C_0 * (\overline{\nu} * \overline{\sigma_f} - \overline{\sigma_f} - \overline{\sigma_a})$$

C_1 and C_2 – net neutron contribution of ^{235}U and ^{241}Pu relative to ^{239}Pu

neutrons from fission of ^{235}U (^{241}Pu)

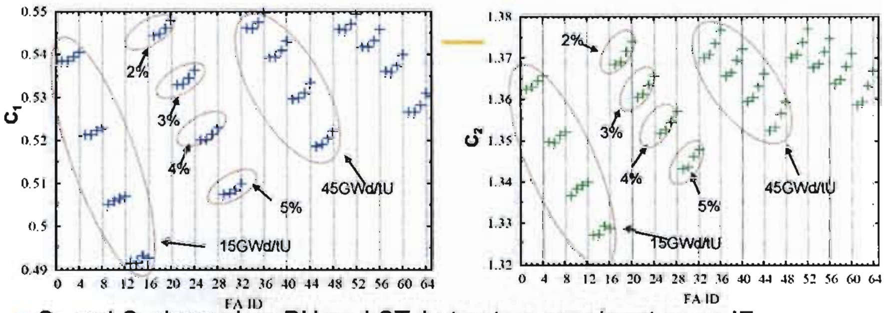
neutrons absorbed by ^{235}U (^{241}Pu)

$$C_i = \frac{\int_V \int_E \sigma_f(E)_i(\nu_i(E) - 1)\Phi(E,V)dEdV - \int_V \int_E \sigma_a(E)_i\Phi(E,V)dEdV}{\int_V \int_E \sigma_f(E)_{^{239}Pu}(\nu(E)_{^{239}Pu} - 1)\Phi(E,V)dEdV - \int_V \int_E \sigma_a(E)_{^{239}Pu}\Phi(E,V)dEdV}$$

neutrons from fission of ^{239}Pu

neutrons absorbed by ^{239}Pu

CIPN – quantifying C_1 and C_2 coefficients



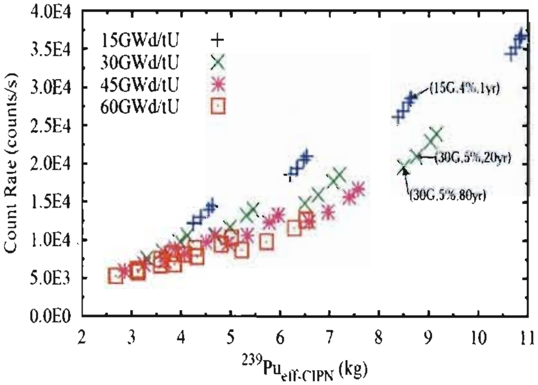
- C_1 and C_2 depend on BU and CT, but not as prominent as on IE
- $<C_1> = 0.529$; $<C_2> = 1.359$
  - C_1 (C_2) fluctuates over a range of -7% to 4% (-2% to 1%) around average
- Assuming realistic range of IE, BU, CT values
  - *C_1 and C_2 vary in the ~ 1% to 2% range around average*

CIPN – determination of fissile content

- C_1 and C_2 averaged over all 64 assemblies

$$^{239}Pu_{eff\_CIPN} \equiv C_1{}^{235}U_m + {}^{239}Pu_m + C_2{}^{241}Pu_m.$$

<C_1> = 0.529
<C_2> = 1.359



Lessons learned:

- Why the scatter: neutron absorbers.
- Two types of neutron absorbers: actinides and FPs (e.g., ^{240}Pu, ^{149}Sm etc.).
- Most absorbers scale with BU, such as ^{240}Pu and ^{149}Sm .
- Some change dramatically with CT, e.g. ^{155}Gd & ^{241}Am.

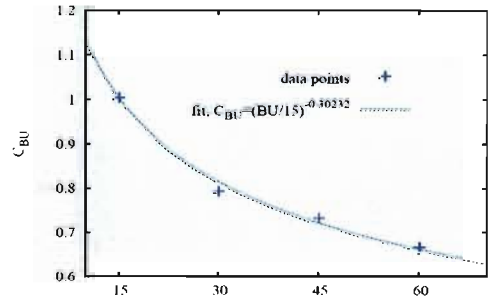
Correction for neutron capture: *half-empirical correction*

Burnup correction

Cooling time correction

$$X = C_{BU}(^{239}Pu_{eff\_CIPN} + C_3{}^{155}Gd_m + C_5{}^{241}Am_m)$$

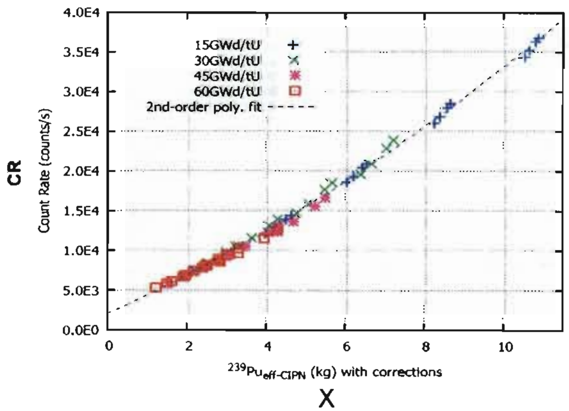
C_3 = -49
C_5 = -0.66



$$C_{BU} = (BU/15)^{-0.30232}$$

- Coefficients C_3, C_5 generated similarly as C1 and C2;
- mass of ^{155}Gd and ^{241}Am required for CT correction
- Simple formulation; easy to implement

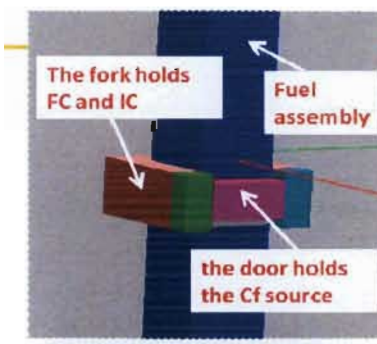
CIPN – determination of fissile content

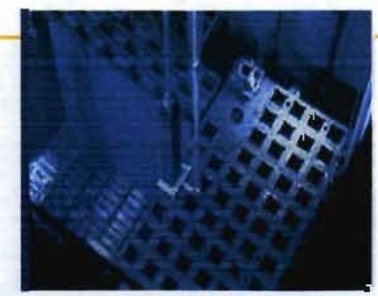


- BU and CT correction to account for parasitic absorption
- A smooth relation between count rate and adjusted Pu_{eff} are found

$$CR = 80.648X^2 + 2307.4X + 2029.68$$

Conceptual experiment setup





CIPN – summary

- **~80% of CIPN signal** produced by **^{235}U, ^{239}Pu and ^{241}Pu**
  - Remaining ~20% primarily from fission in ^{238}U and direct detection of the ^{252}Cf neutron source
  - Both signals expected to be quantified and subtracted from the total signal as background
  - Capable to determine total fissile content with relatively ***uniform spatial sensitivity*** (~15% variation)
  - ***Diversion of 3% of the mass detectable*** assuming reference measurement with full SFA available

CIPN – summary

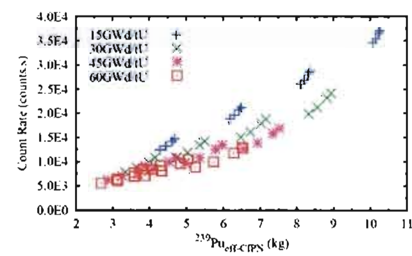
- For the full range of SFAs (15 to 60 GWd/tU), the **statistical precision of the CIPN signal is below 1% in 10 s**
  - Assay times of 100 s expected
  - ***Precision dominated by systematic error***
  - Systematic uncertainty due to motion (± 0.5 cm) of the SFA inside the detector resulted in a signal variation of ~2%
- Correlation between the CIPN signal and the fissile mass in terms of $^{239}Pu_{eff\_CIPN}$ established
  - Dependence of signal on neutron capture
  - Two approaches developed to correct for neutron capture
  - ***Smooth relation between count rate and $^{239}Pu_{eff\_CIPN}$ established***

Thank you

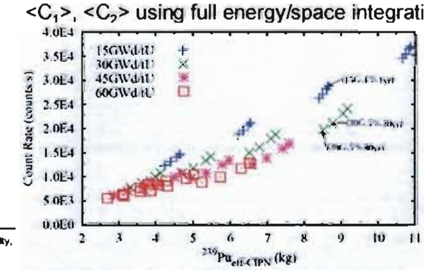
Questions?

C_1, C_2 comparison

Variable C_1, C_2

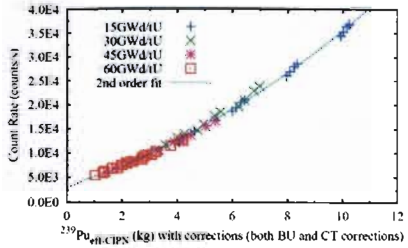


<C_1>, <C_2> using full energy/space integration



C_1, C_2 comparison – CT and BU correction

Variable C_1, C_2 – RSD~1.6%



<C_1>, <C_2> using full energy/space integration – RSD~1.9%