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THE CALIBRATION OF THE DSNC FOR THE MEASUREMENT OF ^{244}Cm AND PLUTONIUM

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BACKGROUND

On 99-10-08, the DSNC was calibrated at the DFDF by the IAEA with assistance from KAERI and LANL. A ^{252}Cf source (K868) was used to calibrate the DSNC for the measurement of ^{244}Cm. The ratio of Pu/Cm is used to calculate the plutonium based on the curium measurement. The neutron emission rate from source K868 is traceable to the NIST reference source CR-5. The ratio of K868/CR-5 is 5.394. The reals (doubles) rate is used for the calibration and the relationship between the ^{252}Cf reals and the ^{244}Cm reals is given in Appendix A. The calibration is a straight line through the origin given by

$$R_{\mathrm{Cm}} = k\, m_{\mathrm{Cm}} \quad ,$$

$$k = \left(2.531 \times 10^{-6}\right)\left(\frac{R_{\mathrm{Cf}}}{m_{\mathrm{Cf}}}\right) \quad ,$$

where k includes a small (3.9%) multiplication correction in the doubles rate for the $U0_2$ rods and pellets.

The IAEA source K868 was used for the calibration and the ^{244}Cm calibration was checked by counting three standard spent fuel reference rods (10 cm long) that were prepared by KAERI. The standard PWR rods (B1, B6, and B8) each contained 3-4 mg of ^{244}Cm as determined from ^{137}Cs burnup measurements combined with ORIGEN2 Code calculations.

EQUIPMENT

The DSNC contains 18 ^{3}He tubes and each tube is directly attached to a PDT-110A amplifier. The amplifiers were adjusted in gain to give approximately the same LED blink rate for a ^{252}Cf source in the detector cavity.

The set-up parameters for the DSNC are given in Table I.

Table I. DSNC Set-up Parameters

Parameter	Value
HV	1820 V
Gate	64 µs
Predelay	4.5 µs
Dead time constants	
a	0.9
b	0.45
c	0
Multi	350
Efficiency	15.86%

RESULTS

To select the operating HV, three standard PWR fuel rods were placed in the DSNC and the bias was varied from 1600 to 1940 V. The results are given in Table II and Fig. 1. At about 1860 V, the gamma-ray pileup becomes significant and the reals rate starts to decrease.

Table II. High Voltage Plateau for Three PWR Rods

HV	PWR Rods (Totals)	PWR Reals
1600	3381	56
1700	7971	340
1800	13058	960
1820	14244	1059
1840	15853	1263
1860	18356	1385
1880	22236	1339
1900	29284	951
1920	39651	149
1940	58642	-142

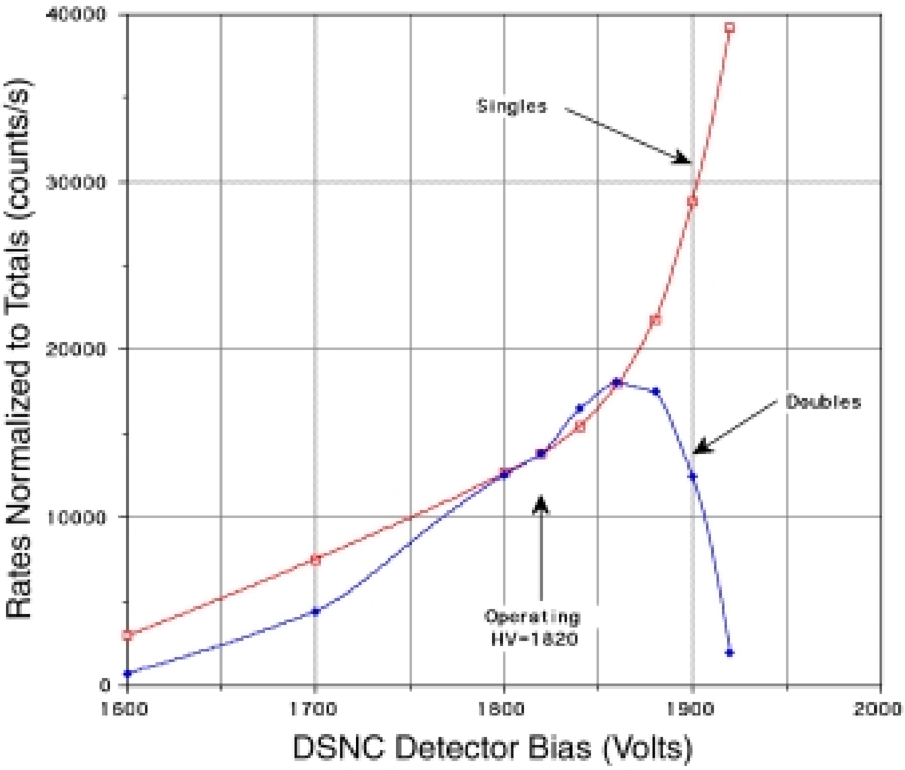


Fig. 1. High voltage plateau for singles and doubles using three standard spent fuel rods.

After completing the gain adjustment, the IMEF hot cell backgrounds were measured giving

T(bkg) = 427 counts/s, and

R(bkg) = 0.17 counts/s on 99-10-8.

The high totals (T) background was possibly caused by the storage of reactor fuel rods in a separate wing of the IMEF cells. The possibility of noise causing the high T rate was considered but the pulses were equally distributed across all of the 18 LED lights and the pulse shape was the same as for the ^{252}Cf neutron source. Also, the background rates in each output sector (A, B, and C) were constant to within 1% during an overnight measurement.

The source K868 was measured for 2000 s and the dead time corrected rates were

T(net) = 1496.5 – 426.5 = 1070 counts/s,

R(net) = 152.37 – 0.17 = 152.2 counts/s (on 99-10-08).

The neutron counting efficiency of the DSNC is a function of the HV setting and for 1820 V, the efficiency was

$$\varepsilon = \frac{1070\ \text{count/s}}{6747\ \text{n/s}} = 0.1586 \quad .$$

The mass of source K868 was 2.888 x 10^{-9} g on 99-10-08 so the calibration constant

$$k = \left(2.531 \text{ x } 10^{-6}\right)\left(\frac{152.2}{2.886 \text{ x } 10^{-9}}\right) = 1.335 \text{ x } 10^{+5}$$

$$\sigma_k = 3.6\% \ , \text{ and}$$

$$m_{(\mathrm{Cm})} = \frac{R_{\mathrm{Cm}}}{k} \quad .$$

To check the ^{244}Cm calibration, three PWR spent fuel rod segments (B1, B6, and B8) were counted in the DSNC on 99-10-08 and the results are given in Table III.

Table III. Curium and ^{252}Cf Calibration Measurements on 99-10-08

Sample	Δt	T(gross)	T (count/s) (net)	R (net) (count/s)	Declared Cm (mg)	Decay Corr. Cm (mg)*	Meas Cm (mg)	Diff (%)
Room bkg	1000	427	0	0.17	—	—	—	—
1 STD Rod (B1)	100	5284	4657	379	3.29	2.99	2.84	-5.03
2 STD (B1,B8)	100	9884	9357	763	6.42	5.83	5.72	-1.9
3 STD (B1,B6,B8)	100	14244	13818	1073	9.55	8.67	8.04	-7.8
K868	500	1497	1070	152.2	—	—	—	—

*Decay corrected (0.03827y^{-1}) to 99-10-08 based on ORIGEN calculations for Cm-244 on approximately 97-04-01.

Figure 2 shows the declared ^{244}Cm mass from ORIGEN2 calculations in the standard rods vs the measured ^{244}Cm using the DSNC and the K868 source calibration. The declared mass in the PWR rods have an average difference to the measured mass of ~4.9%. Part of this difference can be attributed to the uncertainty in the DSNC calibration for ^{244}Cm. However, the major source of the difference is probably caused by the ORIGEN 2 calculation of the ^{244}Cm mass that has an uncertainty greater than 10%.

In general, the ORIGEN2 calculated masses for ^{235}U and plutonium are much more accurate than the mass for ^{244}Cm because of the complex neutron reaction chain needed to produce ^{244}Cm. We have used the DSNC measured values for ^{244}Cm and the ORIGEN2 calculated values for plutonium and uranium to obtain:

$$\text{Pu/Cm} = \frac{0.63}{0.00284} = 222$$

$$^{235}\mathrm{U/Cm} = \frac{0.30}{0.00284} = 106$$

$$\mathrm{U/Cm} = \frac{58.66}{0.00284} = 20655$$

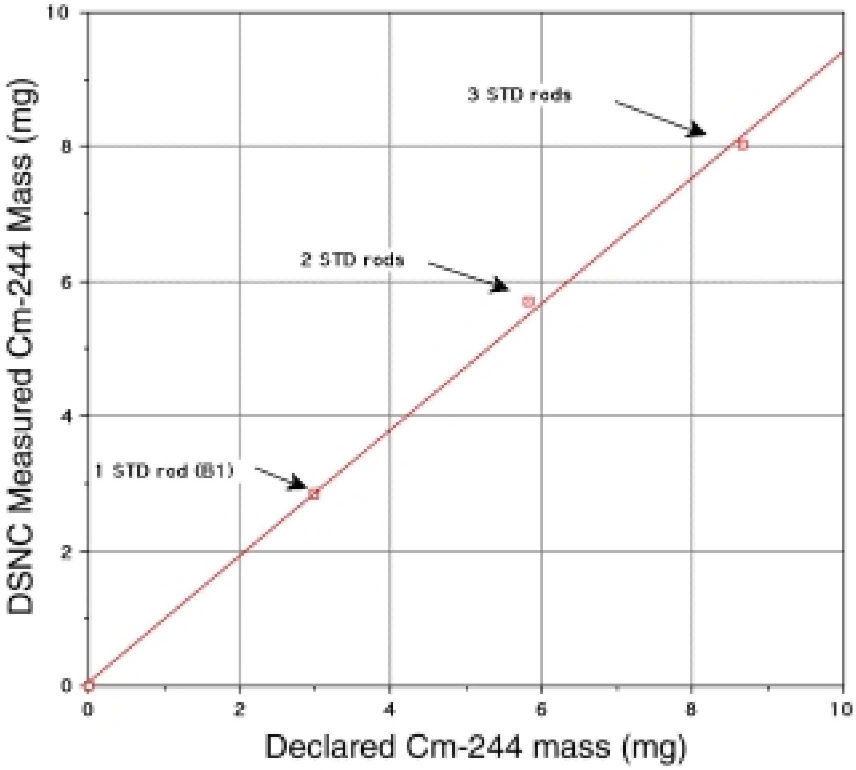


Fig. 2. Comparison of measurements and declarations for the ^{244}Cm mass in three PWR standard rods.

for rods B1 that had a burnup value of 41,000 MWd/tU. Future DUPIC spent fuel with lower burn-up values will have lower Cm/Pu ratios.

SUMMARY

The ^{244}Cm calibration of the DSNC is a simple straight line through the origin as shown in Fig. 3. The small amount of multiplication expected from a pin or pellet or a power grab sample is built into the calibration line. The calibration is linear even with the multiplication because the primary source of multiplication is ^{238}U rather than ^{244}Cm or plutonium.

The calibration slope is given by

$$k = 1.335 \text{ x } 10^5 \text{ counts/s} \bullet \text{g}\,^{244}\text{Cm} \quad .$$

The ^{244}Cm mass measured with this doubles calibration agrees well with the singles calibration obtained from the ^{252}Cf source K868.

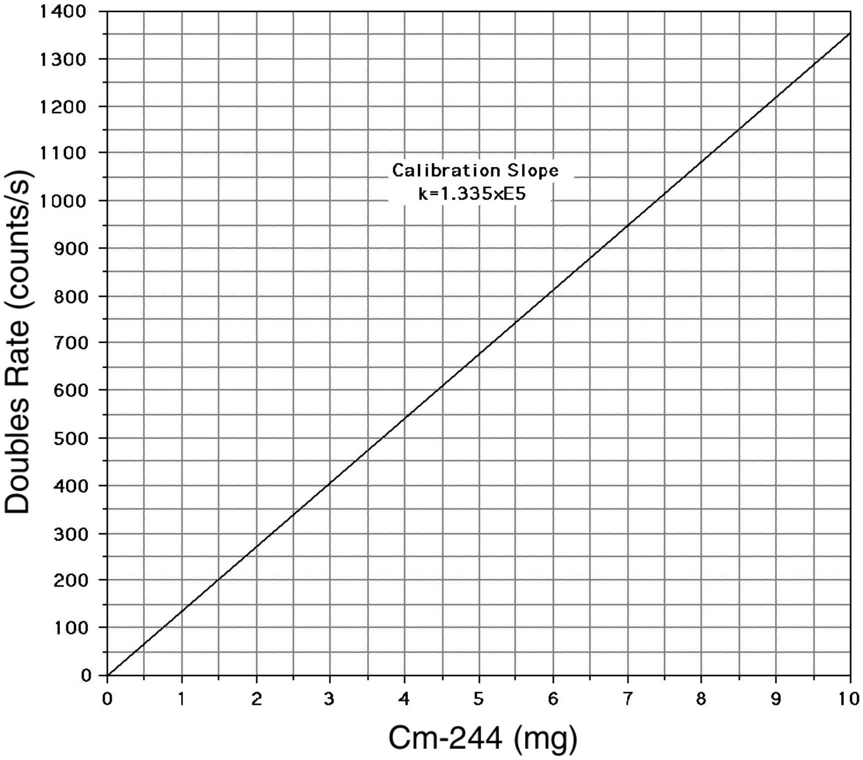


Fig. 3. Calibration line for DSNC ^{244}Cm measurements.

APPENDIX A

DUPIC SAFEGUARDS NEUTRON COUNTER CALIBRATION FOR ^{244}CM THROUGH CORRELATION WITH ^{252}CF

Introduction

The DUPIC Safeguards Neutron Counter (DSNC) at the DUPIC Fuel Demonstration Facility (DFDF) needs a calibration for ^{244}Cm mass but there are no calibration sources of ^{244}Cm available. It is proposed that the following steps be used.

(a) Measure the real coincidence count rate for a ^{252}Cf source of known mass and emission rate located in the center of the assay chamber.

(b) Correlate the real coincidence count rates from point sources with identical masses of ^{244}Cm and ^{252}Cf through Monte Carlo and point model calculations.

(c) Multiplication and (α,n) reactions are nonexistent in step (b) for point sources but have to be taken into account when bundles are measured.

The DSNC was designed to have a uniform spatial detection efficiency for point sources and cylindrical sections of a CANDU bundle. Maximum variations along the axis were less than 2.5% for a point source and about 3% for cylinders. So the calibration need not be adjusted for differences in spatial distributions between a small ^{252}Cf source and a pin or bundle.

The IAEA will supply the ^{252}Cf source. This report gives LANL calculations that compare point sources of ^{244}Cm and ^{252}Cf as noted in steps (a) and (b).

Real Coincidence Count Rate Expressions

An expression for a passive counter's real coincidence count rate *R*, including (α,n) reactions and multiplication, is taken from Ref. A-1 [page 483, Eq. (16-34)].

$$R = \tfrac{1}{2}\, m\, F\, \varepsilon^2\, f_d\, M^2\, \{<\nu_s(\nu_s-1)>/(1+\alpha\, \nu_s) + (M-1)\,(1+\alpha)\, <\nu_i(\nu_i-1)>/[(\nu_i-1)\,(1+\alpha\, \nu_s)]\}, \quad \text{(A-1)}$$

where

m = mass of the fissile material,
F = spontaneous fission rate per gram,
ε = fission neutron detection efficiency,
M = neutron multiplication by the fuel,
α = ratio of neutrons created from (α,n) reactions to spontaneous fissions,
ν_s = average number of neutrons released by a spontaneous fission,
$<\nu_s(\nu_s-1)>$ = reduced second moment for neutrons from spontaneous fission,
ν_i = average number of neutrons released by an induced fission,
$<\nu_i(\nu_i-1)>$ = reduced second moment for neutrons from induced fissions,
f_d = doubles gate fraction, the fraction of the real coincidences that is detected,
= $(2/\varepsilon)\,(R/T)_m <\nu>/<\nu(\nu-1)>$, measured with a ^{252}Cf source in a bare chamber; $(R/T)_m$ is the measured value with the ^{252}Cf source and $<\nu>$ and $<\nu(\nu-1)>$ are moments for spontaneous fission of ^{252}Cf source (3.757 and 11.962, respectively).

For the point sources used in steps (a) and (b), $\alpha = 0$ and $M = 1$. Eq. (A-1) simplifies to the following [Ref. A-1, page 470, Eq. (16-15)].

$$R = \tfrac{1}{2}\, m\, F\, \varepsilon^2\, f_d <\nu_s(\nu_s\text{-}1)> . \qquad \text{(A-2)}$$

^{244}Cm and ^{252}Cf Masses Correlated for Point Sources

The ratio of ^{244}Cm and ^{252}Cf count rates from point sources follows from Eq. (A-2). The doubles gate fraction is the same for both sources and therefore is cancelled in the ratio.

$$R_{Cm} / R_{Cf} = (m_{Cm} / m_{Cf})\ (F_{Cm} / F_{Cf})\ (\varepsilon^2_{Cm} / \varepsilon^2_{Cf})\ (<\nu_s(\nu_s\text{-}1)>_{Cm} / <\nu_s(\nu_s\text{-}1)>_{Cf}). \qquad \text{(A-3)}$$

Of relevance to the calibration is the count rate per unit mass. Equation (3) can be rewritten to display how this ratio for ^{244}Cm is related to the ratio for ^{252}Cf.

$$(R_{Cm} / m_{Cm}) = (R_{Cf} / m_{Cf})\ (F_{Cm} / F_{Cf})\ (\varepsilon^2_{Cm} / \varepsilon^2_{Cf})\ (<\nu_s(\nu_s\text{-}1)>_{Cm} / <\nu_s(\nu_s\text{-}1)>_{Cf}). \qquad \text{(A-4)}$$

The specific count rate (R_{Cf} / m_{Cf}) will be measured with the IAEA ^{252}Cf source at the DFDF. The subsections that follow give values for the other ratios in Eq. (A-4).

Specific Spontaneous Fission Rates

One gram of ^{244}Cm emits 1.08×10^7 neutrons per second from spontaneous fissions. The yield from a gram of ^{252}Cf is 2.34×10^{12} neutrons per second (Ref. A-1, page 339). Their ratio is

$$(F_{Cm} / F_{Cf}) = 4.62 \times 10^{-6} . \qquad \text{(A-5)}$$

Reduced Second Moments

The reduced second moments for ^{244}Cm (Refs. 2 and 3) and ^{252}Cf (Ref. A-1, p. 342) are 6.32 and 11.962, respectively. Their ratio is therefore

$$<\nu_s(\nu_s\text{-}1)>_{Cm} / <\nu_s(\nu_s\text{-}1)>_{Cf} = 0.5283 . \qquad \text{(A-6)}$$

Detection Efficiencies

The MCNP geometry used to design the bundle counter was again used to calculate detection efficiencies of neutrons from point sources of ^{244}Cm and ^{252}Cf located at the center of the assay chamber. Their ratio is essentially unity.

$$\varepsilon^2_{Cm} / \varepsilon^2_{Cf} = 0.998 \pm 0.014 \approx 1.000 . \qquad \text{(A-7)}$$

The two efficiencies are virtually identical because the energy spectra of spontaneous fission neutrons from ^{252}Cf and ^{242}Cm are very similar, as shown in Fig. A-1.

These numerical results are put into Eq. (4) to give this simplified expression for a curium sample with negligible multiplication.

$$(R_{Cm} / m_{Cm}) = (2.436 \times 10^{-6})\ (R_{Cf} / m_{Cf}). \qquad \text{(A-8)}$$

A minimum for the uncertainty of the numerical multiplier can be estimated from the calculated detection efficiencies; uncertainties for the other factors are unknown. On that basis, the uncertainty in Eq. (A-8) can be said to be at least 6.9 x 10^{-8} (or 3.0%).

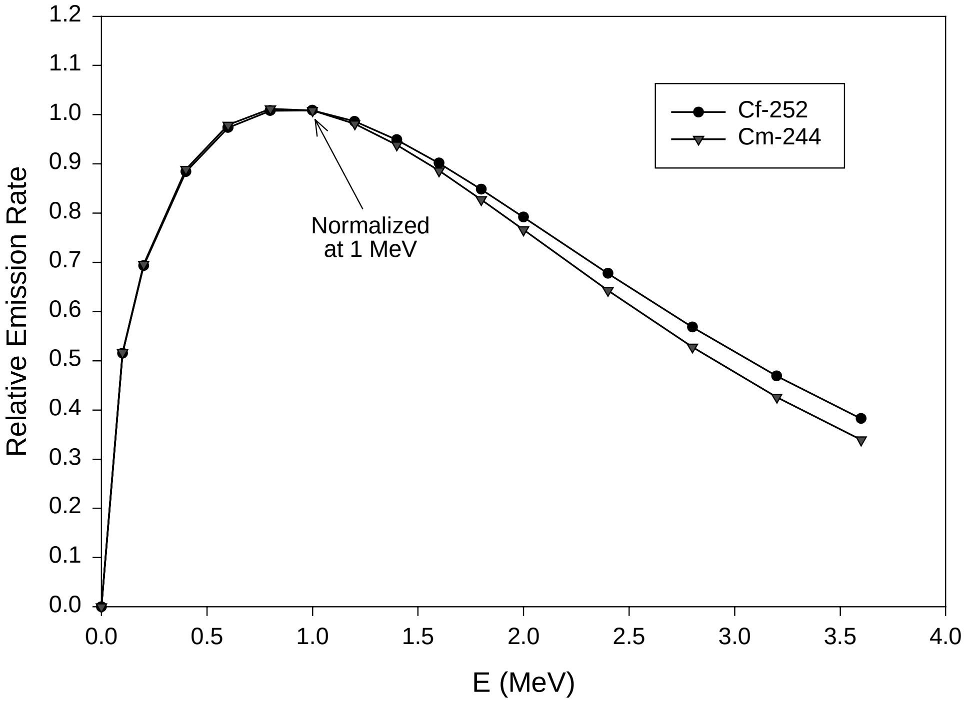


Fig. A-1. Neutron energy spectra for ^{252}Cf and ^{244}Cm are shown, normalized to the same emission rate at 1 MeV. The ^{252}Cf spectrum is slightly "harder" than that of ^{244}Cm. These are Watt spectra, using parameters from the MCNP manual.

Calibration for Non-Multiplying Items

The multiplication effects are negligible for the ^{252}Cf sources to be measured in the DSNC. The mass of ^{244}Cm in non-multiplying items is small, so the calibration curve is taken to be a straight line through the origin.

$$R_{Cm} = a\, m_{Cm}\ . \qquad \text{(A-9)}$$

The slope *a* is found from Eqs. (4), (8) and (9).

$$\begin{aligned} a &= R_{Cm} / m_{Cm} \\ &= (R_{Cf} / m_{Cf})\, (\varepsilon^2_{Cm} / \varepsilon^2_{Cf})\, (F_{Cm} / F_{Cf})\, (<\nu_s(\nu_s-1)>_{Cm} / <\nu_s(\nu_s-1)>_{Cf}) \\ &= (2.436 \times 10^{-6})\, (R_{Cf} / m_{Cf}). \end{aligned} \qquad \text{(A-10)}$$

The ratio of the real count rate from a ^{252}Cf source with its mass was measured with the IAEA's ^{252}Cf source K868. Some further details on the source are given in Appendix B. The result is $R_{Cf}/ m_{Cf} \approx 5.276 \times 10^{10}$ counts/(s•g-^{252}Cf). Therefore, the slope *a* from Eq.(A-10) is

1.285×10^5 counts/(s•g-^{244}Cm) for small samples with negligible multiplication. This slope increase to 1.335×10^5 for PWR pins and typical powder samples.

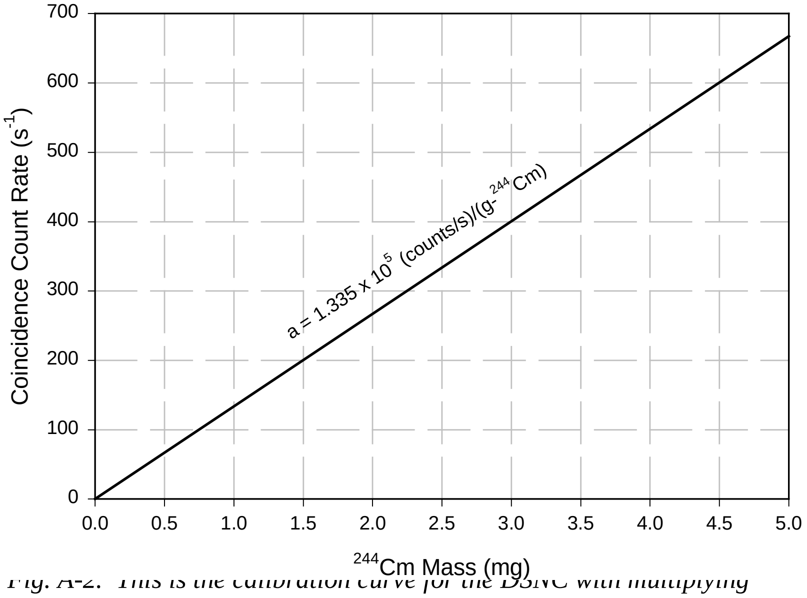


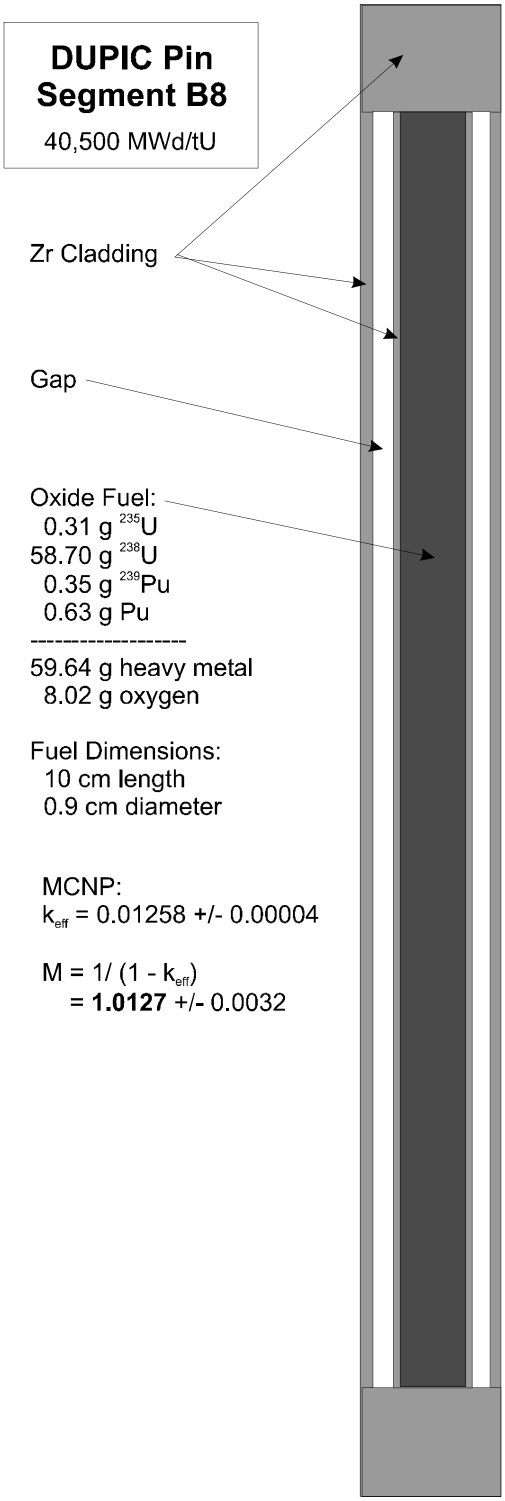
Fig. A-2. This is the calibration curve for the DSNC with multiplying items such as pins and pellets.

Calibration for CANDU Pins and Bundles

If a pin or pellet or a complete bundle of DUPIC pins is measured in the DSNC, the real coincidence count rate per gram of ^{244}Cm will be increased over the small sample calibration because of neutron multiplication. The primary neutrons from spontaneous fissions and (α,n) reactions can cause additional fission reactions in the uranium and plutonium to increase the measured coincidence rate.

This multiplication effect is fairly small because the fissile mass is small and the sample is in air surrounded by a cadmium liner. In practice, the multiplication correction to the small sample calibration curve can be calculated using the MCNP code and the point model equations. Figure A-2 gives the calibration curve for PWR rods and pellets.

An example of the multiplication effect is illustrated by measurements on PWR fuel pin segment B8 with a burnup of 40,500 MWd/tU. Figure A-3 shows the segment inside CANDU cladding and some of the details on the pin's composition. MCNP was used to calculate $k_{eff} = 0.01258 \pm 0.00004$, so $M = 1.0127 \pm 0.0032$. This rather small increase in M over unity nevertheless leads to a 3.9% increase in the coincidence count rate.



0.31 g ^{235}U
58.70 g ^{238}U
0.35 g ^{239}Pu
0.63 g Pu

59.64 g heavy metal
8.02 g oxygen

Fuel Dimensions:
10 cm length
0.9 cm diameter

MCNP:
k_{eff} = 0.01258 +/- 0.00004

M = $1/(1 - k_{eff})$
= **1.0127** +/- 0.0032

Fig. A-3. This is a sketch of the MCNP model of PWR pin segment B8. The segment is enclosed in its own cladding, which in turn is enclosed in cladding similar to that of a CANDU fuel pin. The material composition is indicated, followed by the results of the MCNP calculation and the consequences for the multiplication M.

References

A-1. D. Reilly, N. Ensslin, and H. Smith, Jr, editors, *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550, LA-UR-90-732, United States Nuclear Regulatory Commission, March 1991, pp. 190-192.

A-2. N. Miura and H. O. Menlove, "The Use of Curium Neutrons to Verify Plutonium in Spent Fuel and Reprocessing Wastes," Los Alamos National Laboratory report LA-12774-MS (May 1994).

A-3. D. A. Hicks, J. Ise, Jr., and R. V. Pyle, "Probabilities of Prompt-Neutron Emission from Spontaneous Fissions," *Physical Review* **101**(3), 1016-1020 (February 1, 1956).

APPENDIX B

^{252}CF SOURCE CHARACTERIZATION AND APPLICATION

The ^{252}Cf source K868 will be used to calibrate the DSNC for ^{244}Cm measurements. The ^{244}Cm calibration is a straight line, Eq. (A-9), with a slope *k* given by Eq. (10), repeated here for convenience.

$$R_{Cm} = k\, m_{Cm} \, . \tag{B-1}$$

$$k = (2.436 \text{ x } 10^{-6})\,(R_{Cf} / m_{Cf}). \tag{B-2}$$

for a sample with negligible multiplication.

The value of (R_{Cf} / m_{Cf}) is to be measured with source K868 of known ^{252}Cf mass. The value of m_{Cf} is determined by cross-calibrating source K868 with source CR-5 that is traceable to a NIST calibration.

The measured source rate from K868 was 5.394 times larger than from the standard ^{252}Cf source CR5. On January 1, 1987 CR5 had a yield of ($3.54\text{x}10^4$ ± 1.5%) neutrons/s. The decay constant for ^{252}Cf is 0.2623 y^{-1} so the ^{252}Cf mass in CR5 on October 1, 1999 will be $5.37\text{x}10^{-10}$ g and the ^{252}Cf mass in K868 will be ($5.37\text{x}10^{-10}$ x 5.394) = $2.90\text{x}10^{-9}$ g on October 1, 1999.

The value of m_{Cf} used to calculate *k* from Eq. (10) or (A-2) will be corrected for the decay from October 1, 1999 to the actual date of the calibration. The ^{252}Cf decay constant per day is $7.172\text{x}10^{-4}$ d^{-1}.

The efficiency of the DSNC is about 15.86%, depending on the operating high voltage, and the neutron yield from K868 on October 1, 1999 is ($2.90\text{x}10^{-9}$ g)x($2.34\text{x}10^{12}$ n/s•g) = 6786 n/s. Therefore the expected net singles rate is about 1076 counts/s on that date.

At the time of the initial installation and calibration, source K868 will be counted in the DSNC to establish the ^{244}Cm calibration. The source K868 will then be put under IAEA seal and placed inside the hot cell for future verification of the DSNC performance and calibration.

APPENDIX C

DSNC BACKGROUND MEASUREMENTS FOR MONITORING SPENT FUEL IN THE HOT CELL

The DSNC provides a time record of the neutron background in the DFDF hot cell. The DSNC is surrounded by about 15 cm of CH_2 and 10 cm of lead to isolate the detector from the backgrounds.

Three PWR standard rods (B1, B6, and B8) were placed at different locations in the hot cell to determine the totals counting rate vs the spent fuel position. There was no measurable reals background from the fuel in the hot cell.

Figure C-1 shows a diagram of the positions of the 3 PWR rod segments.

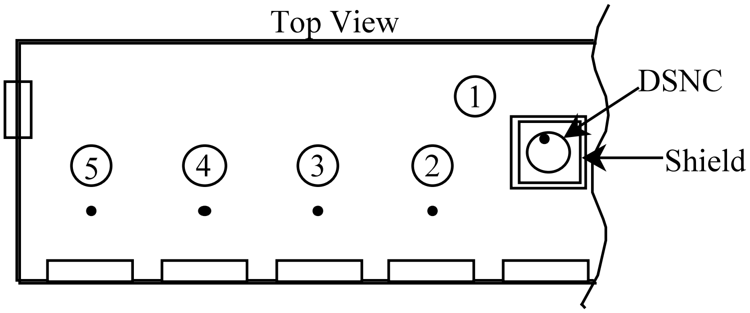


Fig. C-1. Diagram of the measurement positions for the 3 PWR spent fuel rods.

Table C-I gives the total rates and measurement time for each position.

Table C-I.

Time 99-10-08	Position	Totals (counts/s)
15:00	P1	510
15:05	P2	750
15:07	P3	448
15:08	P4	429
15:10	P5	430
15:15	Outside	426

The increase in the totals rate was negligible for positions P4 and P5. The close position P1 is lower than P2 because the DSNC shield is more effective for the close position.