LA-12737-M
Manual

UC-700
Issued: March 1994

NBC Operation Manual Including the Multi-position Add-A-Source Function

H. O. Menlove
L. A. Foster
J. Baca

Los Alamos
NATIONAL LABORATORY
Los Alamos, New Mexico 87545

MASTER

CONTENTS

CONTENTS (cont.)

NBC OPERATION MANUAL INCLUDING THE MULTI-POSITION ADD-A-SOURCE FUNCTION

by

H. O. Menlove, L. A. Foster, and J. Baca

ABSTRACT

This manual describes the design modifications and operating characteristics of a 200-ℓ-drum neutron coincidence counter. The counter has six shielded banks of ^{3}He tubes and JSR-11 shift register coincidence electronics. The modified design has a counting efficiency of 19.3%. The neutron counter measures the spontaneous-fission rate from the plutonium, and when this is combined with the plutonium isotopic ratios, we can determine the plutonium mass. The system includes the new multi-position add-a-source (AS) technique that uses a small ^{252}Cf source to determine the drum's matrix perturbation to the plutonium assay. The ^{252}Cf source is measured at three positions on the exterior of the drum to obtain the spatial distribution for the matrix correction. This manual gives the performance and calibration parameters. The matrix corrections by the AS technique are accurate to a few percent for typical applications.

GENERAL

This manual describes the design modifications and operating characteristics of the Neutron Barrel Counter (NBC). This drum counter will be used at TA-55 to determine the plutonium content of 200-ℓ drums.

Los Alamos prepared the specifications and conceptual design of the drum counter.[1] Jomar Systems/Canberra designed and built the counter. After fabrication, the counter was delivered to the Los Alamos National Laboratory for a program of performance measurements and acceptance testing, add-a-source (AS) installation,[2] initial calibration, software installation and checkout, and preparation of documentation. This manual describes the measured drum counter performance characteristics and the instrument settings required for operation and constitutes part of the documentation for the counter.

The system includes the following hardware:

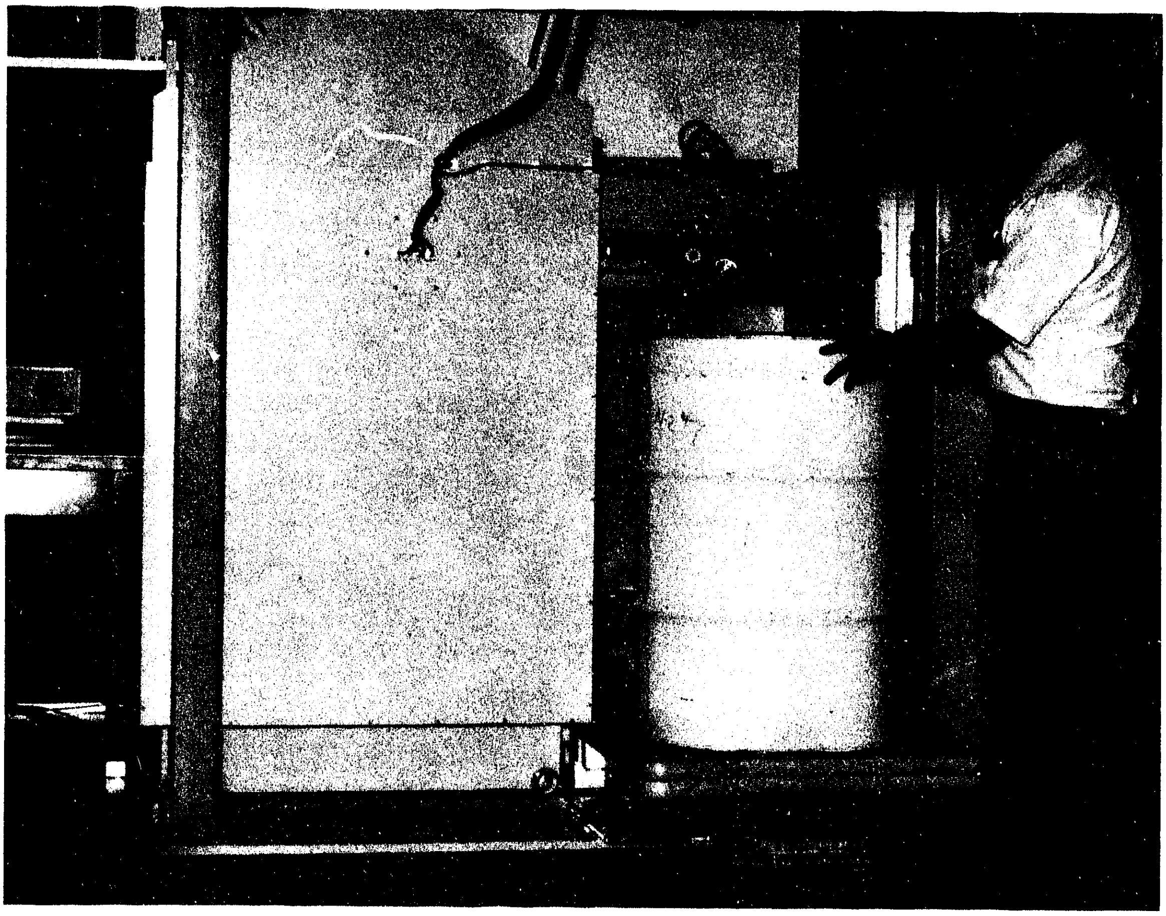
- detector head,
- Compumotor drive system for AS,
- ^{252}Cf source for the AS and normalization measurements,
- electronics rack,
- JSR-11 coincidence counting electronics,
- personal computer, and
- printer.

PHYSICAL DESCRIPTION

Figure 1 shows a photograph of the drum counter with the door open. The interior well is 711 by 711 by 965 mm high and can easily hold a standard 200-ℓ drum, which can be loaded by rolling it along the fixed platform of rotating wheels as shown in the figure. The door is moved back and forth on its wheels by a motor positioned on top of the counter.

The counter has six banks of ^{3}He tubes–one in each of the four sides and one on the top and bottom. The four vertical side banks each contain ten 914-mm-active-length ^{3}He tubes, and the top and bottom horizontal banks each contain ten 508-mm-active-length ^{3}He tubes. The four vertical side banks each require two AMPTEK counting channels consisting of AMPTEK Model A-111 preamp/discriminator

PHYSICAL DESCRIPTION
(cont.)



(Neg. No. CN91 3959)

Fig. 1. Photograph of neutron detector showing the open sample cavity and a 200-ℓ drum.

PHYSICAL DESCRIPTION
(cont.)

boards. The two horizontal banks use shorter ^{3}He tubes with less capacitance and each requires only one AMPTEK counting channel. The electronics of the NBC are similar to those of the HLNC-II.[3,4] The detector counts the totals and coincidence neutrons from the spontaneous fission of the even isotopes of plutonium.

On one side of the counter is a panel with 10 numbered lights, each corresponding to one of the 10 AMPTEK channels. Table I lists the 10 channel numbers and the corresponding location of the AMPTEKs inside the counter. Table II gives the characteristics of the ^{3}He tubes.

Each of the six banks of ^{3}He tubes is embedded in a 100-mm-thick slab of high-density polyethylene (CH_2). Each bank is also shielded on the outside by another 100-mm-thick slab of CH_2. Within the six detector banks, the ^{3}He tubes are centered 4.16 cm from the inside edge of the CH_2.

Table I. AMPTEK Channel Location

Channel No.	Position
1	Bottom bank
2	Right bank front end
3	Right bank back end
4	Back bank right end
5	Back bank left end
6	Top bank
7	Door bank right end
8	Door bank left end
9	Left bank back end
10	Left bank front end

PHYSICAL DESCRIPTION
(cont.)

Table II. Characteristics of ^{3}He Tubes

Model (sides)	RS-P4-0836-203
Model (top & bottom)	RS-P4-0820-203
Active length	914 mm vertical
	508 mm horizontal
Diameter	25 mm
Fill pressure	4 atm
Gas quench	Argon + CH_4
Cladding	Stainless steel
Operating high voltage	1660 V

The cadmium liners that Jomar Systems/Canberra normally places on the sample cavity walls and also between the detector slabs and the exterior CH_2 shield were removed for the NBC modifications. The liners were removed to increase the detector's efficiency and to decrease the *coincidence* neutron background. The cadmium increases the coincidence background from cosmic-ray spallation reactions and its removal improves the sensitivity of the system for low-background applications.

DOOR OPERATION

Figure 2 shows the front panel of the control box that the operator uses to open and close the door. To activate the door, the operator must hold down the "DOOR ENABLE" button as well as either the "OPEN DOOR" or "CLOSE DOOR" button. Once the door has reached its opening or closing limit, it will trigger the limit sensor, the appropriate "LIMIT" light on the control box will turn on, and the door will automatically stop. Just beyond the opening limit sensor is a second sensor that will stop the door in the event that the first fails to do so. In that situation, the "EMERGENCY LIMIT" light on the control box will turn on. Once the door has reached its limit, it can be moved only in the opposite direction.

COMPUTER HARDWARE

System interconnections between the computer, the JSR-11, the control box, and the drum counter are illustrated in Fig. 3.

PHYSICAL DESCRIPTION
(cont.)

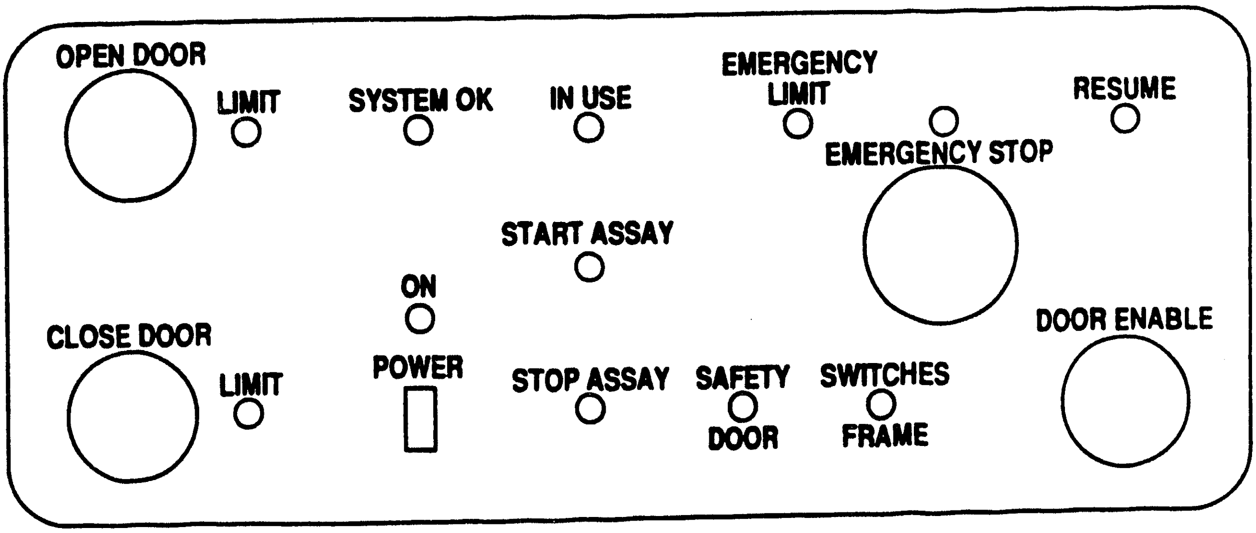


Fig. 2. Schematic diagram of the NBC door controller face panel.

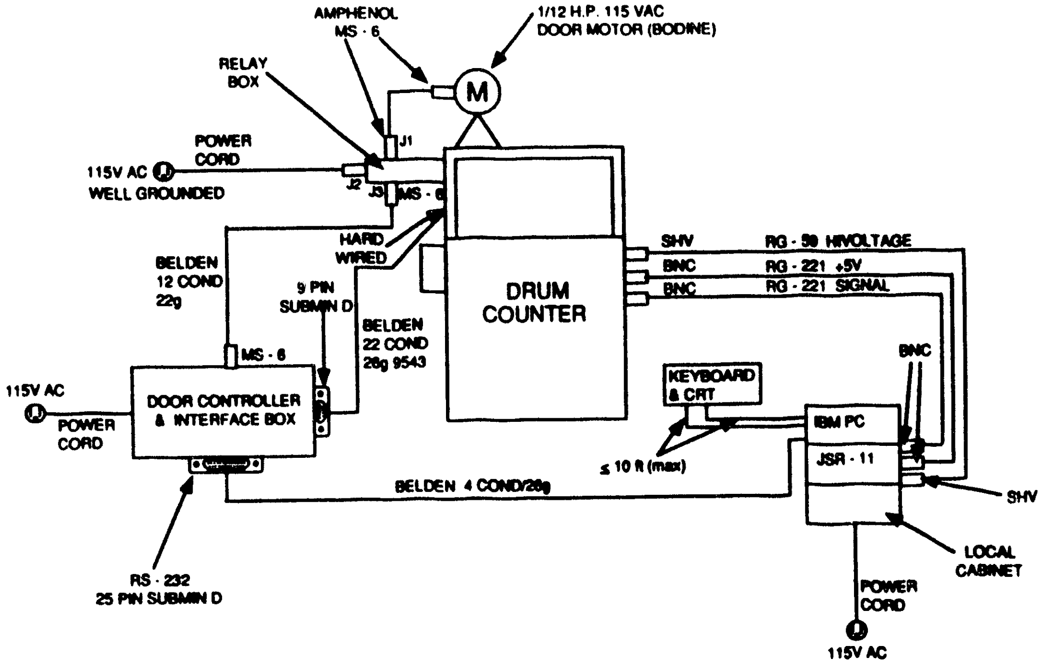


Fig. 3. Wiring diagram for the NBC door motor and controller.

CALIBRATION SOURCE

System Check

A ^{252}Cf reference source was assigned to the NBC system. This source was counted at the time of calibration and during subsequent use of the detector for normalization. The source, with an initial emission rate of 2.5 x 10^4 n/s, is adequate for at least 10 years in the NBC.

For convenience, the same source is used for both the normalization and the AS function. The normalization measurement is made with an empty chamber.

ANALOG ELECTRONICS

The NBC uses the fast-counting circuitry[4] based on the miniature AMPTEK hybrid chip. These chips are located near the end of the ^{3}He tubes (see Fig. 4) and contain the preamplifier, amplifier, and discriminator circuits. Ten of these amplifier units are located in the top part of the six detector banks. The output signals from these 10 amplifiers are added and sent to the input of the shift-register board. The connection is made through the "external SR input" on the back panel of the JSR-11 shift-register.*

HIGH-VOLTAGE PLATEAU

The high-voltage (hv) plateau for the NBC ^{3}He tubes is shown in Fig. 5 where the totals counting rate is plotted as a function of the hv bias on the detector tubes. The "knee" or start of the plateau is at 1630 V, and we set the operating voltage at 1660 V. A higher setting can result in a coincidence bias at high counting rates and increased gamma-ray sensitivity at high dose levels. The setting at 1660 V is not sensitive to gamma rays for dose levels up to ~1 R/h at the surface of the drums.

MOISTURE SEAL

To avoid moisture buildup in the hv junction box of the detector, the box contains desiccant and the openings are sealed with silicone rubber. The humidity and degree of moisture saturation in the desiccant can be read from outside the junction box. Also, the desiccant tubes can be exchanged for fresh tubes.

---

*Model JSR-11, JOMAR Systems, Los Alamos, NM 87544.

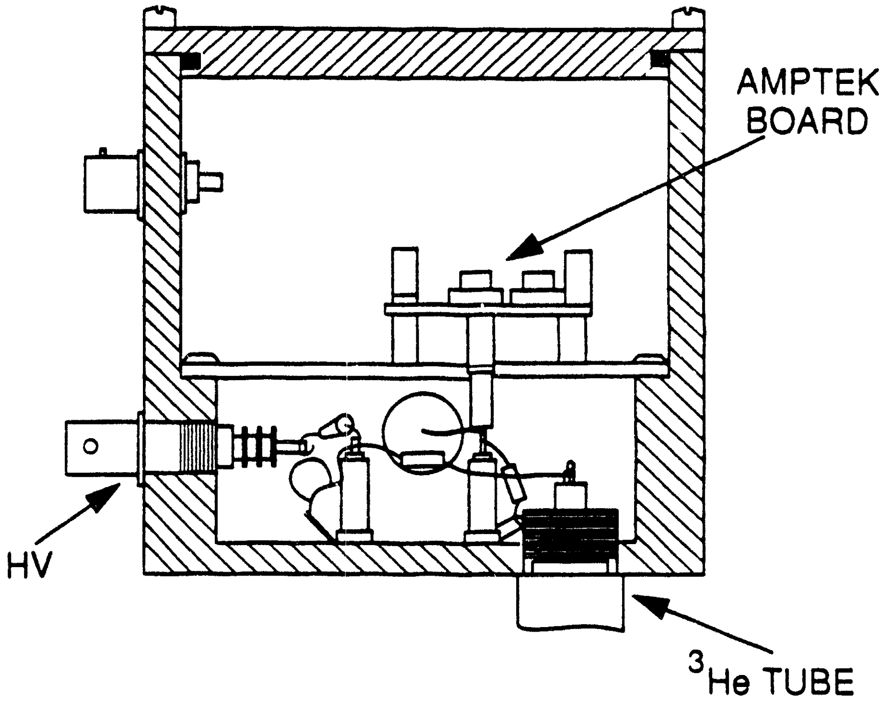


Fig. 4. Diagram of the hv junction box that contains the AMPTEK board.

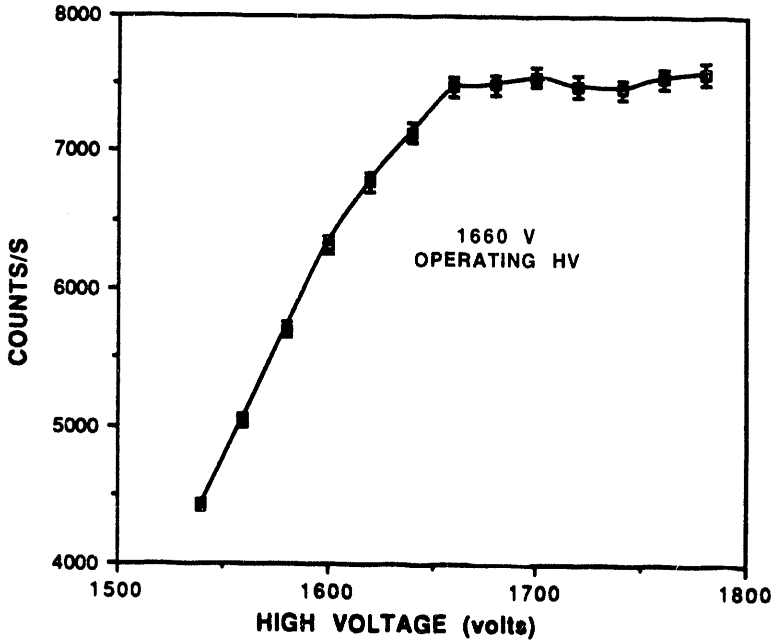


Fig. 5. Totals counting rate vs the hv setting of the ^{3}He tubes in the NBC.

EFFICIENCY

The efficiency of the NBC system was measured using a calibrated ^{252}Cf source in the detector. The results are listed in Table III.

Table III. NBC Performance Characteristics
(date: 93-03-25)

Parameter	NBC
Efficiency (no sample)	19.3%
Die-away time (center)	79 μs
Gate setting	128 μs
Ratio 128 μs/64 μs Gates	1.443
High voltage	1660 V
Dead time coefficient *a*	0.65 μs
Dead time coefficient *b*	0.21 μs
Californium reference rate (on 93-02-18)	
T(CR-4)	518 counts/s
R(CR-4)	87.1 counts/s
Add-a-source reference (93-02-18)	
R(G-370)	751 counts/s

We used a ^{252}Cf source to measure a die-away time of 79 μs.

DETECTOR DEADTIME

The NBC uses the same basic electronics and amplifiers as does the HLNC-II. The dead time coefficient δ is given by

$$\delta = (a + bT \times 10^{-6})\ \mu\text{s} ,$$

where T is the measured totals rate in counts/s and a and b are constants given in Table III. The corrected counting rates are

$$T(\text{corr.}) = T\, e^{\delta T/4}$$

and

$$R(\text{corr.}) = R\, e^{\delta T} .$$

DETECTOR DEADTIME

(cont.)

It is important to use the same dead-time coefficient for both calibration and assay so that any errors in the correction will cancel to a first approximation.

At very high counting rates ($T \geq 1$ MHz), a small positive bias appears in the dead-time corrected rates.

Our counting rates are expected to be less than 1 MHz, so we shall use the standard 4.5-μs predelay setting.

VERTICAL RESPONSE PROFILES

To determine sample positioning effects, we counted a ^{252}Cf point source at a variety of vertical and radial positions in an empty drum. The measurements were made using the (WDAS) detector[2] that has the same geometry as the NBC. The vertical profile measurements were made at a radius of 20 cm from the center of the 200-ℓ drum. The outside edge of the drum has a radius of 28 cm and the 20-cm radius is approximately the volume-averaged mean radius. That is, the drum volume inside 20 cm equals the volume outside 20 cm.

Figure 6 shows the normalized vertical totals and reals rates for the ^{252}Cf source. The dips at the top and bottom are caused by the gaps in the detector coverage at the ends of the detector banks.

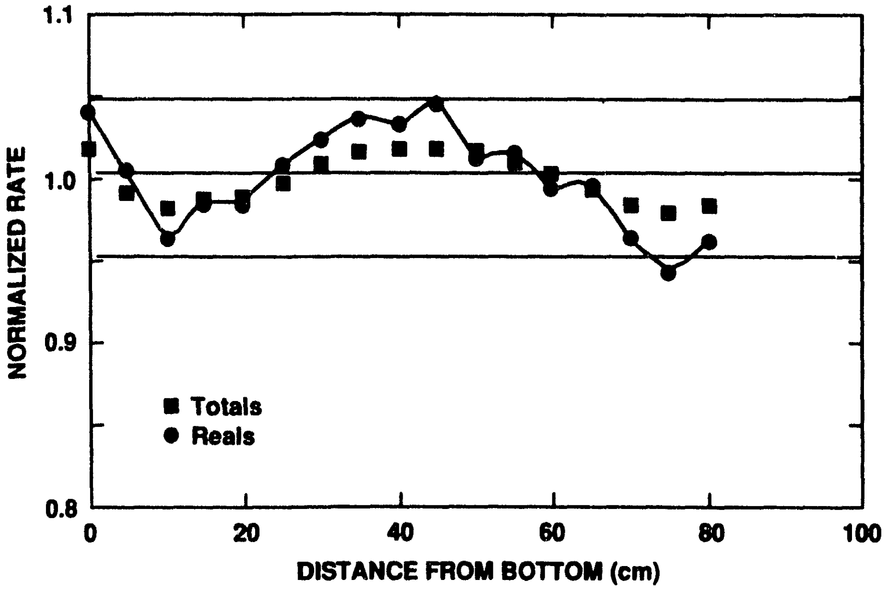


Fig. 6. Totals and reals vertical response profiles measured using ^{252}Cf at a radius of 20 cm in an empty 200-ℓ drum.

RADIAL PROFILES

For the radial profile, the ^{252}Cf source was positioned at four different radial positions and three different vertical positions. Figure 7 shows the radial profile for the average of the three vertical positions. The vertical positions were 15, 35, and 55 cm above the bottom of the drum.

The multiplication-corrected reals profiles are not given because, for the waste drums, the multiplication-corrected rates are *not used.* This is because the multiplication is negligible and normally the α parameter is unknown.

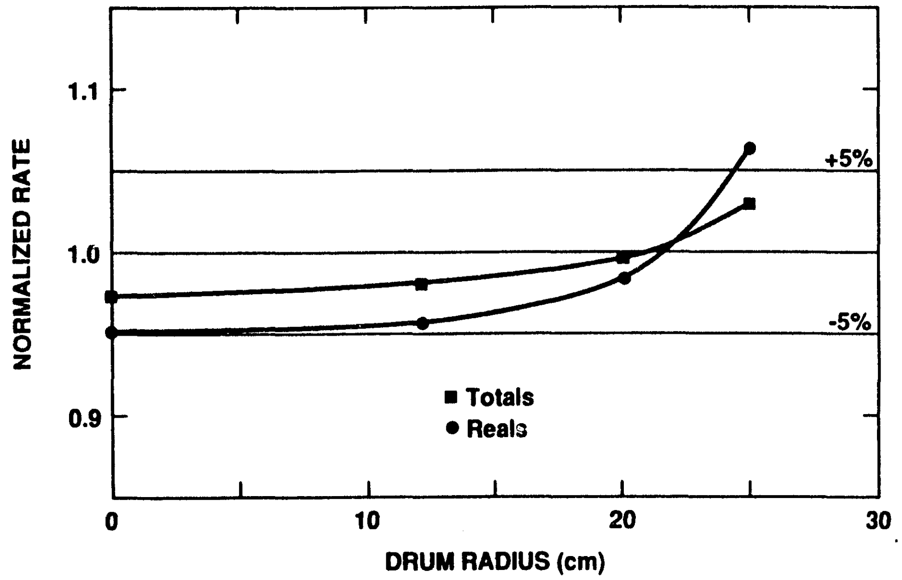


Fig. 7. Totals and reals radial response profiles averaged over the heights of 15, 35, and 55 cm in an empty 200-ℓ drum.

ADD-A-SOURCE METHOD

The basis of the AS method is to measure the matrix perturbation to the counting rate from a small ^{252}Cf source (2.5 x 10^4 n/s) on the outside of the sample and to use the information to correct for the matrix perturbation on the inside of the sample. For the present case, we have positioned the AS neutron source at three positions on the 200-ℓ drum. Figure 8 shows a schematic diagram that illustrates the technique in which the AS neutrons originate from ^{252}Cf and the sample neutrons originate from plutonium. The figure shows the location of the three positions on the bottom and side of the drum.

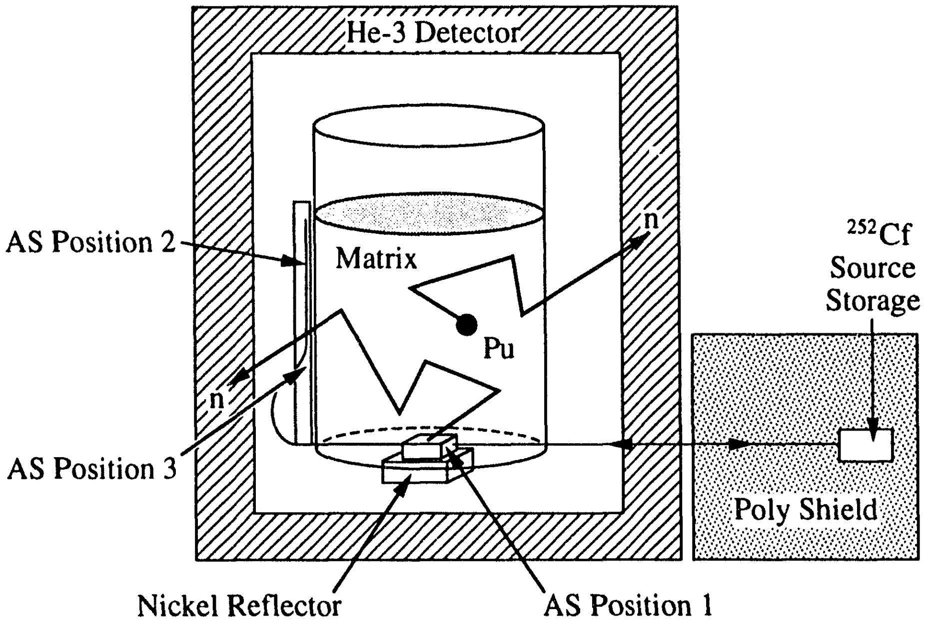


Fig. 8. Schematic illustration for the AS concept showing the Teleflex cable transfer system that moves the ^{252}Cf source from the shielded storage position to the matrix interrogation position.

The sample matrix has two primary effects on the neutrons: (1) energy reduction by scattering reactions, and (2) neutron absorption of the low-energy neutrons. The counter is designed with an optimum moderator (CH_2) thickness to be relatively insensitive to the energy reduction; however, as the hydrogen density in the drum increases, the *absorption* process significantly reduces the measured neutron signal.

ADD-A-SOURCE METHOD (cont.)

To correct for the matrix perturbation on the neutron signal, the AS method measures each drum both with and without the ^{252}Cf source on the outside of the drum. The measured quantities are

T_0, R_0 = Totals and reals rates from ^{252}Cf for an empty drum.

T, R = Totals and reals rates from a sample drum without ^{252}Cf.

T(Cf), R(Cf) = Totals and reals rates from a sample drum with the ^{252}Cf.

The net ^{252}Cf reals rate for the ^{252}Cf and a loaded sample drum is

$$R(\mathrm{Cf}) - R = R(\mathrm{net}).$$

We use the ratio of the empty drum (after source decay correction) to the net loaded drum to make the matrix correction as follows:

$$\left(\frac{R_0\,\mathrm{e}^{-\lambda t}}{R(\mathrm{net})} - 1\right) = x \ ,$$

and the correction factor (CF) is defined as

$$CF = 1 + f(x)$$

where $f(x)$ is a polynomial function of x based on empirical measurements. The measured R for a drum is corrected to give

$$R(\mathrm{corrected}) = R(\mathrm{measured})\ CF \ .$$

ADD-A-SOURCE CALIBRATION

The functional relationship between the AS perturbation x and the volume averaged sample perturbation $f(x)$ was determined empirically by measuring a large variety of matrix loadings with the AS. The matrix materials in the drums are listed in Table IV.

ADD-A-SOURCE CALIBRATION

(cont.)

Table IV. Matrix Materials

Matrix	Matrix Weight (kg)	ρ_H (g/cm^3)
Empty (19.3 kg steel drum)	0.0	0.0
Vermiculite + Borax	34.0	0.0008
Paper	20.9	0.007
Boron Glass	142.0	0
Polyethylene Shavings	11.8	0.0086
Polyethylene Tubes	42.6	0.031
Wood	54.6	unknown
Vermiculite + 29.5 kg of Poly Beads	40.0	0.0212
Vermiculite + 59.1 kg of Poly Beads	78.5	0.0423
Concrete (top) + Poly Shavings (bottom)	100	unknown
Concrete (bottom) + Paper (top)	118	unknown

A separate neutron source was counted at nine positions in the drum to give a *volume averaged* matrix effect. The nine positions are illustrated in Fig. 9. The average of the nine positions was then ratioed to the empty drum case to give the volume averaged perturbation

$$\left[\frac{R_0'(\text{empty vol})}{R'\ (\text{matrix vol})} - 1\right] = y \text{ (vol av perturbation)} \ ,$$

where

R_0' = reals rates averaged over the volume of an empty drum and

R' = reals rates averaged over the volume of the drum with matrix material.

ADD-A-SOURCE CALIBRATION

(cont.)

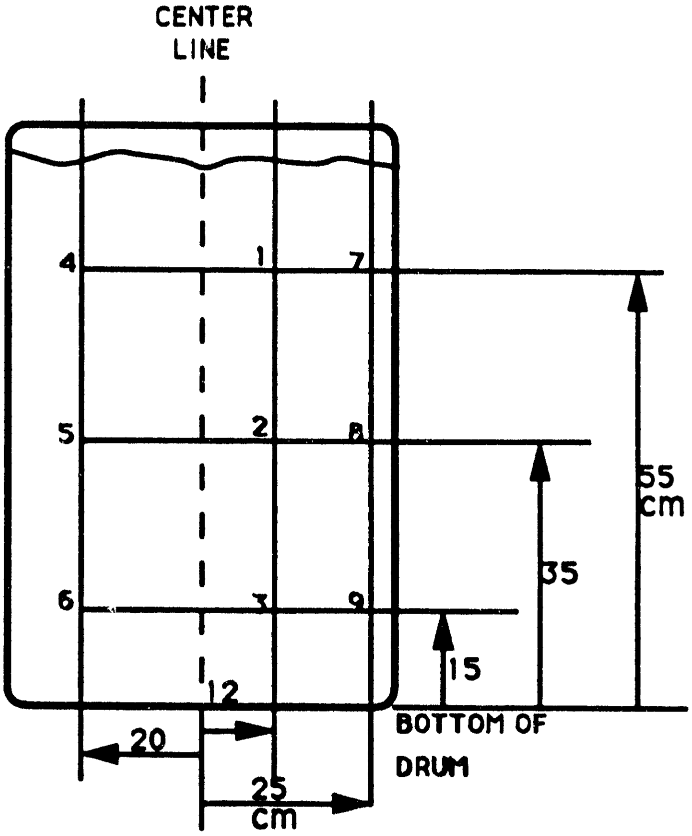


Fig. 9. Diagram of the neutron source positions to give a volume averaged response in a 200-ℓ drum.

Table V list the totals and reals rates and errors for the volume averaged case for a few of the matrix materials. A typical drum contained ~190 ℓ of matrix material.

Table VI gives the AS reals data for the three positions (see Fig. 9) on the drum. The ratio of each count to the average of the three positions can be used to flag discontinuities in the matrix loading. Position 1 (bottom of drum) picks up the hydrogen increase in the matrix with more sensitivity than positions 2 and 3 (side positions) because of the better geometric coupling to the bottom of the drum.

ADD-A-SOURCE CALIBRATION

(cont.)

The AS correction factor is derived from the volume averaged reals ratio for the empty drum over the matrix filled drum compared with the corresponding AS reals ratio. This data is given in Table VII.

A plot of the volume averaged perturbations *y* vs the AS perturbation *x* is shown in Fig. 10. The data point with the highest AS perturbation corresponds to a drum loaded with CH_2 beads and vermiculite ($\rho = \rho_H = 0.0423$ g/cm^3). The hydrogen content in this drum is equivalent to 38% of the hydrogen content of water.

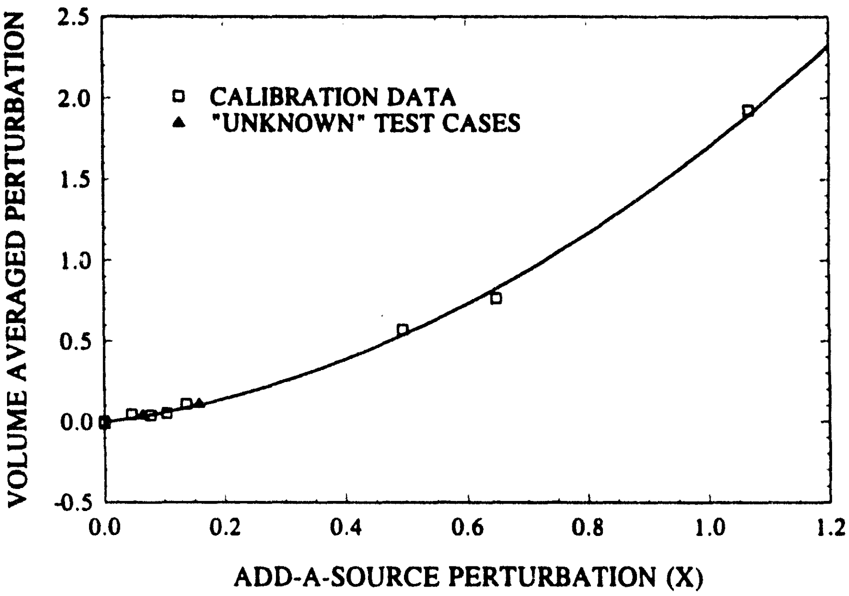


Fig. 10. The AS perturbation (x) in R vs the volume averaged perturbation (y) in R′ for a variety of matrix materials in 200-ℓ drums.

ADD-A-SOURCE CALIBRATION
(cont.)

Table V. Totals and Reals Data as a Function of Position and Matrix Material

Sample	Position	Meas Time (s)	Reals (1/s)	Sigma Reals	Totals (1/s)	Sigma Totals
Empty Drum	1	480	79.9	0.47	483.1	0.94
2/24/93	2	420	82.3	0.47	490.1	0.91
	3	450	76.5	0.54	473.4	1.64
	4	450	84.2	0.73	493.7	1.28
	5	540	85.1	0.59	500.6	1.11
	6	480	81.2	0.78	487.7	1.16
	7	480	82.3	0.73	491.5	1.27
	8	480	84.2	0.66	495.5	1.64
	9	420	75.6	0.56	470.8	0.73
	Av	-----	81.3	0.21	487.4	0.41
Boron Glass	1	420	73.0	0.41	434.2	1.15
3/2/93	2	420	81.5	0.54	456.5	1.10
	3	450	79.1	0.39	448.4	1.03
	4	420	74.0	0.95	437.3	1.18
	5	420	82.4	0.71	456.9	1.09
	6	420	74.7	0.87	439.2	2.05
	7	420	75.4	0.92	438.4	0.86
	8	450	83.2	1.04	456.9	1.45
	9	420	71.9	0.60	425.8	0.85
	Av	-----	77.2	0.25	443.7	0.41
Poly Shavings	1	420	77.3	0.71	482.3	1.10
2/25/93	2	450	76.2	0.59	487.5	1.73
	3	450	71.8	0.58	469.6	1.31
	4	450	81.4	0.69	495.3	1.50
	5	420	82.2	0.73	500.7	1.50
	6	450	78.2	0.86	484.9	0.96
	7	420	78.4	0.76	484.5	1.35
	3	480	80.4	0.59	491.1	0.96
	9	480	70.1	0.81	465.1	1.12
	Av	-----	77.3	0.24	484.6	0.43
Vermiculite + Poly Beads 59.1 kg	1	450	20.1	0.64	282.1	1.51
3/30/93	2	420	13.4	0.58	241.4	1.53
	3	420	16.4	0.63	255.8	1.59
	4	240	31.6	0.91	345.2	1.90
	5	180	24.6	0.64	307.1	1.45
	6	180	25.1	0.65	305.5	1.45
	7	180	41.3	0.84	372.0	1.64
	8	180	36.5	0.78	348.6	1.58
	9	180	34.4	0.75	337.0	1.55
	Av	-----	27.0	0.24	310.5	0.53
Concrete + Poly Shavings	1	120	67.5	1.36	450.4	2.30
3/30/93	2	120	66.0	1.35	453.3	2.30
	3	120	65.2	1.32	445.8	2.28
	4	120	78.5	1.48	481.4	2.40
	5	120	80.0	1.50	482.2	2.41
	6	120	73.5	1.42	462.9	2.35
	7	120	72.1	1.41	457.5	2.33
	8	120	73.5	1.43	461.6	2.35
	9	120	64.1	1.30	437.2	2.26
	Av	-----	71.2	0.47	459.1	0.78

ADD-A-SOURCE CALIBRATION

(cont.)

Table VI. Add-a-Source Data as a Function of Position

Sample	Position	Reals	Sigma R	Ratio to Av
Empty Drum	1	793.77	4.85	0.939
2/24/93	2	867.87	4.93	1.027
	3	873.69	3.90	1.034
	Av	845.11	2.65	1.000
Boron Glass	1	686.40	3.99	0.878
3/2/93	2	819.81	3.24	1.049
	3	837.89	3.67	1.072
	Av	781.36	2.11	1.000
Poly Shavings	1	675.12	3.10	0.883
2/25/93	2	810.53	3.66	1.060
	3	809.02	2.98	1.058
	Av	764.89	1.88	1.000
Vermiculite + Poly Beads 59.1 kg	1	341.34	4.26	0.855
3/30/93	2	454.03	5.12	1.137
	3	402.08	4.82	1.007
	Av	399.15	2.74	1.000
Concrete + Poly Shavings	1	613.04	12.13	0.860
3/30/93	2	793.29	13.97	1.113
	3	732.49	13.56	1.027
	Av	712.94	7.65	1.000

ADD-A-SOURCE CALIBRATION

(cont.)

Table VII. ^{252}Cf Counting Rates for Add-a-Source and the Average over the Matrix Volume

	Volume Average			Add-a-Source		
	T_{av}	R_{av}	T_0/T	R_0/R	R_{av}	R_0/R
Empty Drum	487.4	81.3	1.000	1.000	845.1	1.000
Vermiculite and Borax	468.4	82.2	1.040	0.988	844.6	1.001
Paper-1	485.7	77.9	1.003	1.043	802.9	1.053
Paper-2	485.8	77.3	1.005	1.048	813.1	1.039
Wood	479.4	73.2	1.018	1.107	744.0	1.136
Boron Glass	450.1	78.3	1.083	1.037	784.7	1.077
Poly Shavings	484.9	77.4	1.005	1.050	765.4	1.104
Poly Tubes	404.4	46.1	1.205	1.763	512.6	1.649
Vermiculite + Poly Beads, 29.5 kg	405.5	51.8	1.202	1.569	564.8	1.496
Vermiculite + Poly Beads, 59.1 kg	318.2	27.7	1.535	2.924	408.9	2.067
Concrete + Poly Shavings	470.5	72.9	1.038	1.112	730.4	1.157
Concrete + Paper	481.3	78.0	1.014	1.039	794.2	1.064

A second order polynomial was fitted through the y vs x data using the Deming Code to give the predicted volume averaged matrix perturbation $f(x)$ based on the AS measured perturbation,

$$y = f(x) = a_0 + a_1 x + a_2 x^2$$

where

$a_0 = -0.004893 \pm 0.0105$
$a_1 = 0.5020 \pm 0.1152$
$a_2 = 1.200 \pm 0.1293$

ADD-A-SOURCE CALIBRATION
(cont.)

with the variances and covariances as follows:

1	1	1.674E - 2
1	2	-1.410E - 2
1	3	8.102E - 4
2	2	1.327E - 2
2	3	-8.471E - 4
3	3	1.104E - 4

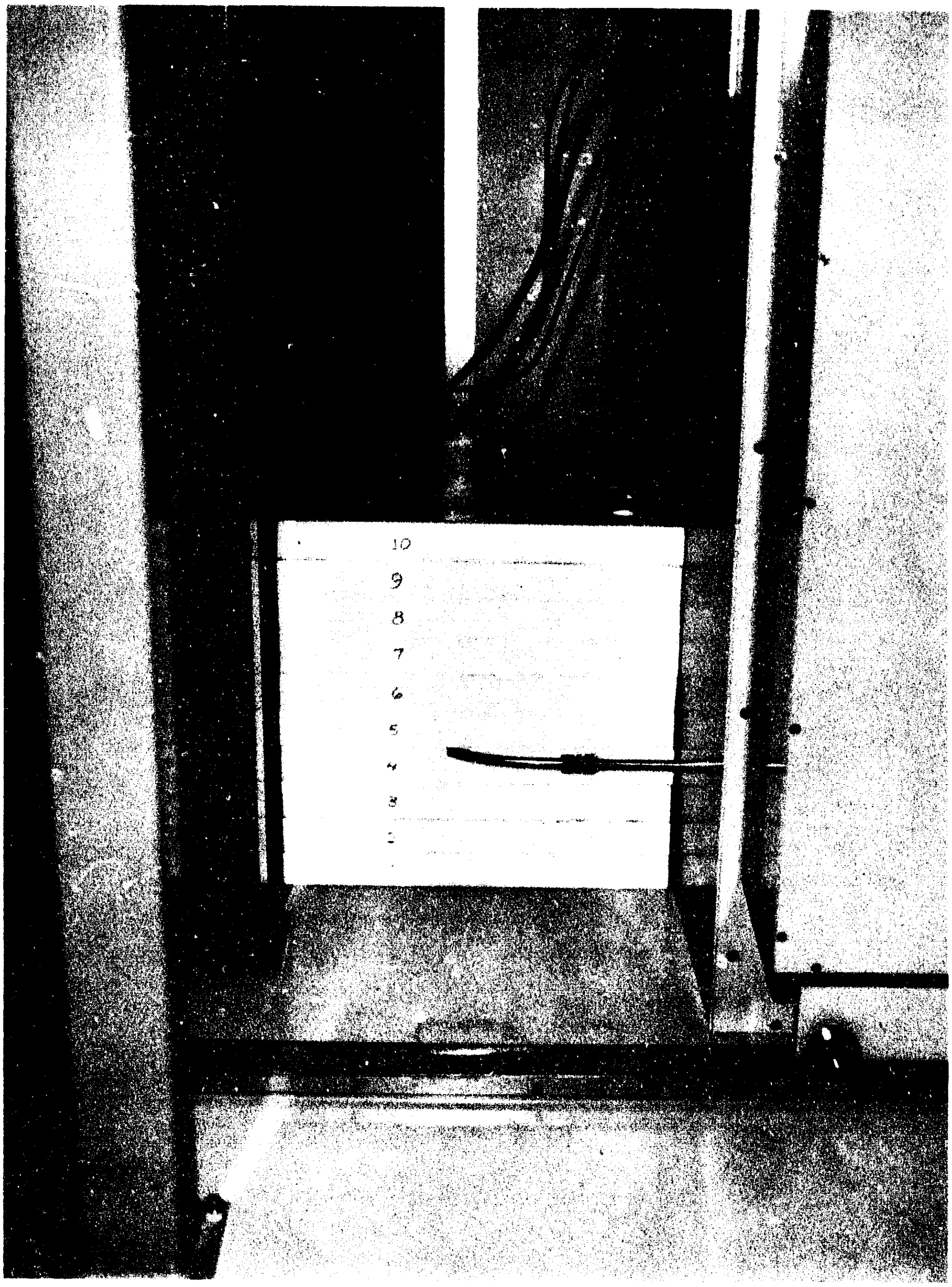
ADD-A-SOURCE TRANSFER SYSTEM

The ^{252}Cf source (2.5 x 10^4 n/s) is automatically transferred from the shielded location shown in Fig. 11 to the drum by means of a Teleflex cable and Compumotor drive system.[5] Figure 12 shows the AS transfer motor. At the stopping point under the drum, a 5-cm-thick nickel reflector reflects additional source neutrons into the drum. On the side of the drum, 5-8 cm-thick graphite reflectors are used for the same purpose.

It is necessary to measure an empty drum at the time of calibration, and the resulting R_0 value is stored in the computer for subsequent data evaluation.

Each unknown drum is placed into the counter, the door is closed, and the AS measurement is performed for ~3 min. The ^{252}Cf source is automatically removed from the detector and an ~10-min passive neutron measurement completes the assay. All of the time intervals are adjustable in the computer parameter files.

ADD-A-SOURCE TRANSFER SYSTEM
(cont.)

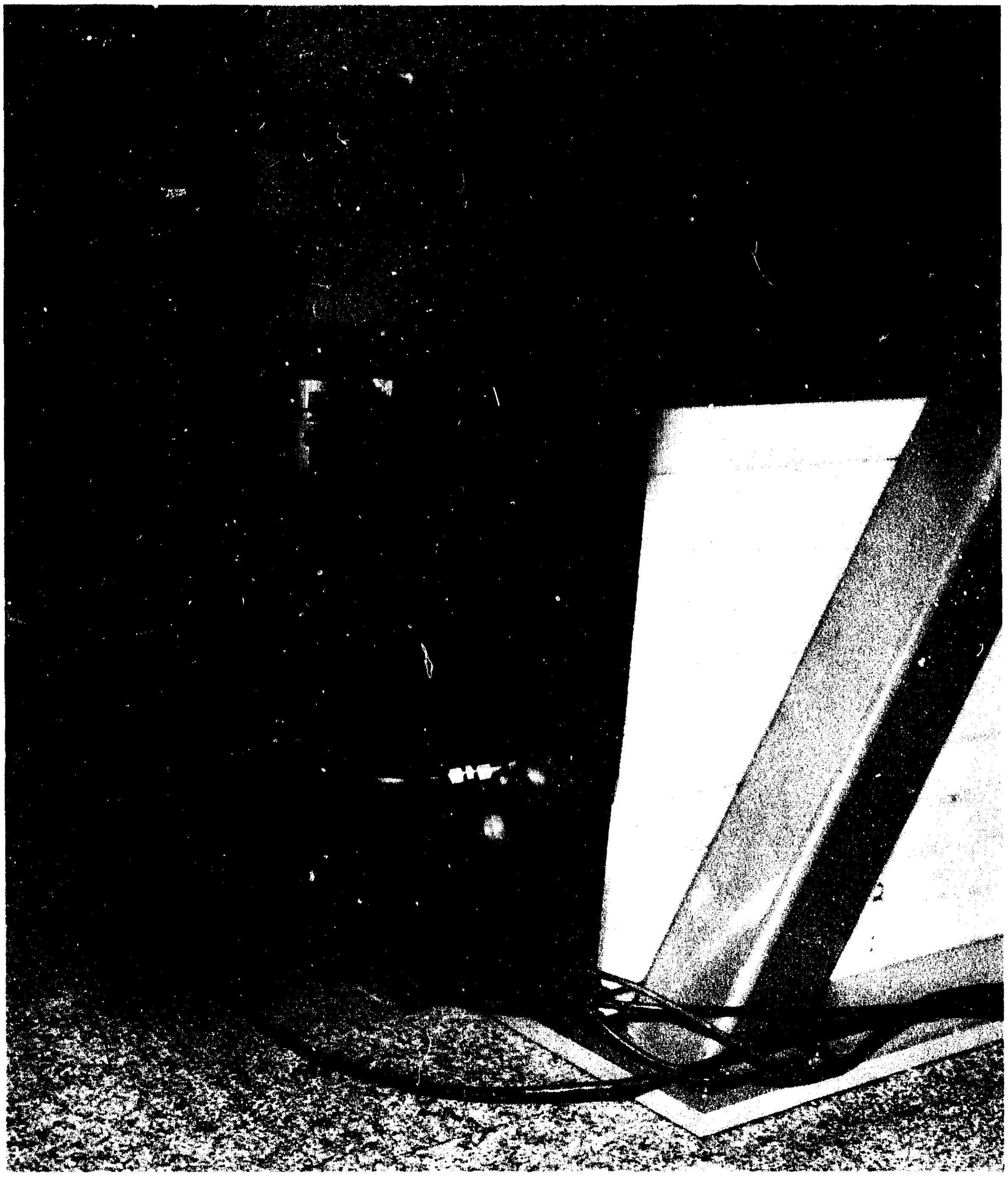


(Neg. No. CN91 3957)

Fig. 11. Photograph of the AS neutron shield positioned on the side of the detector.

ADD-A-SOURCE TRANSFER SYSTEM

(cont.)



(Neg. No. CN91 3955)

Fig. 12. Photograph of the Compumotor and teleflex transfer system on the back of NBC for the transfer of the AS.

MATRIX STUDY RESULTS

To evaluate the error in using the AS correction for the matrix materials listed in Table IV we used the fitted function $f(x)$ to give

$$CF = 1 + f(x) \ .$$

All of the measured R values were normalized to the empty drum case and corrected by CF.

The ratios and corrected results are listed in Table VIII and illustrated in Fig. 13. The CF-corrected reals deviate from the empty drum with a standard deviation of only ±1.7%.

Table VIII. Add-a-Source Matrix Correction for 200-ℓ Drums

Sample	Volume Average R_0/R	Relative	Correction Factor CF	Relative R(corr.)
Empty Drum	1.000	1.000	0.978	0.978
Vermiculite and Borax	0.988	1.012	0.979	0.990
Paper-1	1.043	0.959	1.026	0.984
Paper-2	1.048	0.954	1.013	0.967
Wood	1.107	0.904	1.103	0.997
Boron Glass	1.037	0.964	1.048	1.010
Poly Shavings	1.050	0.953	1.073	1.022
Poly Tubes	1.763	0.567	1.798	1.020
Vermiculite + Poly Beads, 29.5 kg	1.569	0.637	1.536	0.979
Vermiculite + Poly Beads, 59.1 kg	2.924	0.342	2.914	0.996
Concrete + Poly Shavings	1.112	0.900	1.124	1.011
Concrete + Paper	1.039	0.962	1.036	0.997
			1 sigma =	1.70%

These same drums (except for wood and concrete) were used to determine the $f(x)$ function so the results show the scatter of our CF calibration. However, after the CF calibration was established, drums filled with concrete plus paper, concrete plus polyethylene, and wood were measured as unknowns and the corrected response ($R \bullet CF$) was within 1% of the empty-drum case. A typical drum of organic waste is expected to have a hydrogen loading that is equivalent to ρ_{CH_2} ~0.014 g/cm^3, and thus the correction factor will be smaller than the present calibration range.

Future work will include additional matrix materials to help establish the accuracy of the AS method for a variety of matrix materials.

MATRIX STUDY RESULTS
(cont.)

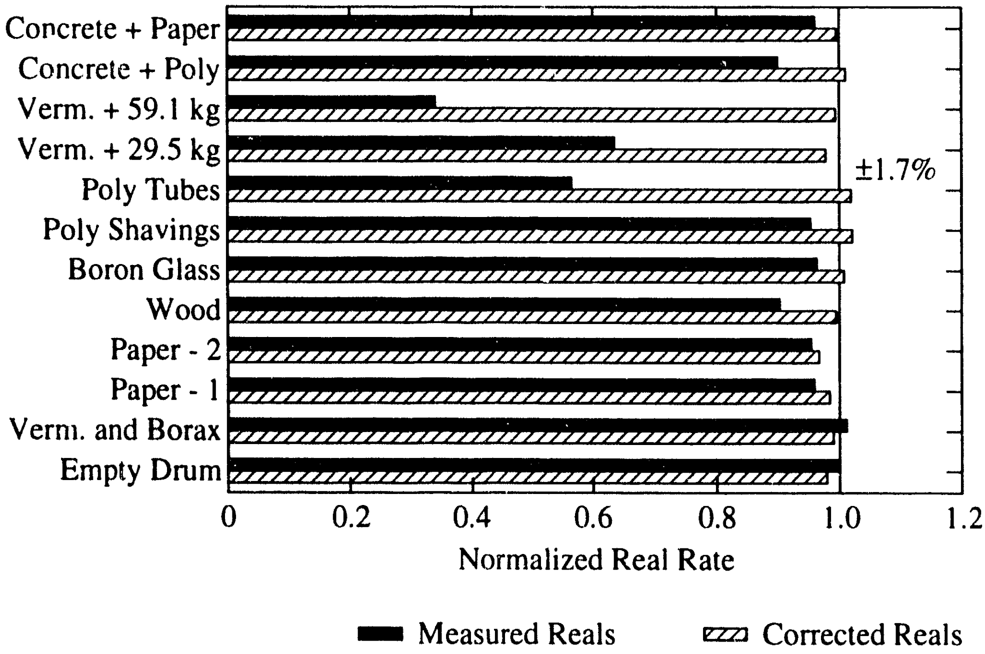


Fig. 13. The measured reals and the AS CF-corrected reals for various matrix materials in 200-ℓ drums.

PRECALIBRATION

Calibration measurements using small plutonium samples and MOX pellets were made at Los Alamos TA-35 before the installation of NBC at TA-55. The calibration range of interest for the waste drums is from zero to a few hundred grams of plutonium. Additional calibration measurements were performed at TA-55 after installation.

The specifications for the MOX pellets that we used for the Los Alamos calibration are listed in Table IX and the plutonium powder standards are listed in Table X. The calibrations were performed in an empty drum.

PRECALIBRATION
(cont.)

Table IX. MOX Pellet ^{240}Pu-eff Corrected to 3/19/93

		Weight % relative to total plutonium mass					
Sample ID	Total Pu (g)	238	239	240	241	242	g ^{240}Pu-eff
A1-066	0.1525	0.0846	88.4881	10.3149	0.8211	0.2953	0.0168
A1-081	0.5073	0.0846	88.4881	10.3149	0.8211	0.2953	0.0559
A1-078	0.8055	0.0846	88.4881	10.3149	0.8211	0.2953	0.0888
A1-089	0.2264	0.2329	77.9335	18.9059	1.6844	1.2434	0.0489
A1-119	0.2649	0.0513	87.2169	11.8545	0.6647	0.2127	0.0327

Table X. SGS Can Standards Corrected to 3/19/93

		Weight % relative to total plutonium mass					
Sample ID	Total Pu (g)	238	239	240	241	242	g ^{240}Pu-eff
STDSGA10	9.9970	0.0061	96.3390	3.5610	0.0756	0.0180	0.3605
STDSGCAL20	20.0070	0.0061	96.3390	3.5610	0.0756	0.0180	0.7216
STDSGA30	29.9770	0.0061	96.3390	3.5610	0.0756	0.0180	1.0811
STDSGC30	29.9790	0.0061	96.3390	3.5610	0.0756	0.0180	1.0812

Because of the geometric variation of the counting efficiency, we used several positions for the standards and we used the volume averaged rates.

TA-35 RESULTS

Table XI gives the volume averaged reals rates for the standards and Fig. 14 gives a plot of R vs the ^{240}Pu-eff mass where

$$^{240}\text{Pu-eff} = 2.52\,^{238}\text{Pu} + {}^{240}\text{Pu} + 1.68\,^{242}\text{Pu}\ .$$

TA-35 RESULTS
(cont.)

Table XI. Initial Plutonium Calibration Measured at TA-35 Bldg. 27

Sample ID	Time (s)	g ^{240}Pu-eff	Reals/s	sigma R	Reals/s - g ^{240}Pu-eff
66 + 78 + 119	9000	0.138	2.492	0.022	18.06
66 + 78 + 81 + 89 + 119	9000	0.243	4.298	0.031	17.69
STDSGA10	4800	0.3605	6.015	0.109	16.69
STDSGCAL20	3600	0.7217	12.437	0.174	17.23
STDSGA30	3600	1.0813	19.139	0.292	17.70
STDSGA30 + STDSGC30	2400	2.1626	39.345	0.715	18.19

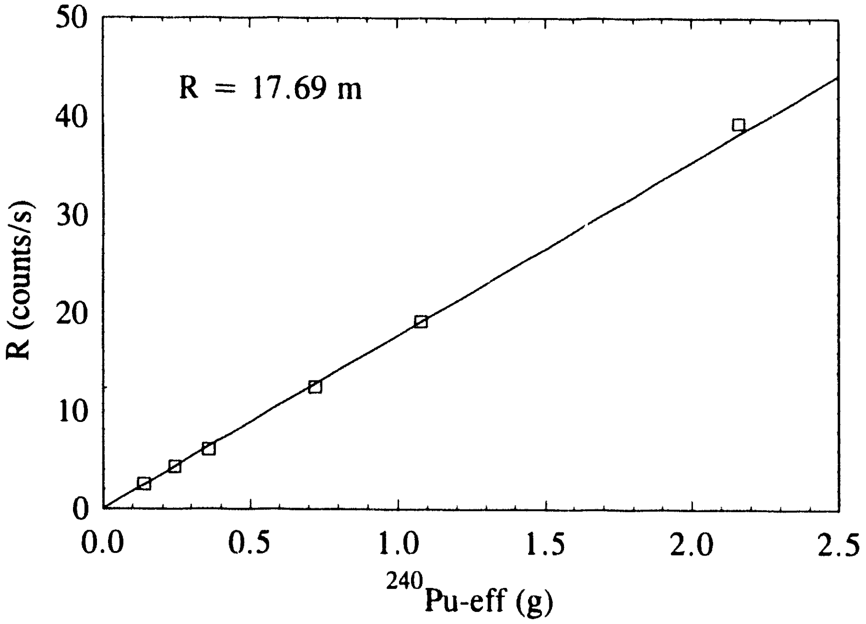


Fig. 14. Preliminary calibration curve for (CF) R vs the ^{240}Pu-eff mass for MOX pellets at Los Alamos.

TA-35 RESULTS
(cont.)

There is negligible multiplication in the standards so we fitted a linear calibration line that went through the origin. The Deming fitted calibration line is

$$R(\text{corr.}) = a\, m + b,$$

where m is the ^{240}Pu-eff mass and

$a = 17.69$ counts/s • g ^{240}Pu-eff., and
$b = 6.42 \times 10^{-5}$.

When matrix material is in the drum, the measured reals are corrected by

$$R(\text{corr}) = R(\text{meas})\ CF\ ,$$

before they are fitted to the calibration line. The *CF* correction factor is applied to *both* the calibration standards and the assay samples. Thus, any error in *CF* is limited to matrix differences between the standards and the unknowns.

TA-55 CALIBRATIONS

After the NBC was moved from TA-35 to TA-55 at LANL, the system was recalibrated using TA-55 standards. The efficiency change after the move was negligible.

Table XII gives the specifications for the TA-55 (PF-4 Count Room) standards. The mass values are about an order of magnitude larger than the standards used at TA-35.

Table XII. Standards and Data for Calibration at TA-55 PF-4 Count Room

		Weight % relative to total plutonium mass					
Sample ID	Total Pu (g)	238	239	240	241	242	^{240}Pu-eff (g)
STDR3	20.93	0.00122	94.073	5.765	0.129	0.0021	1.22
STD3	60.05	0.0298	91.993	7.617	0.283	0.077	4.7
STD4	59.89	0.0175	93.501	6.327	0.129	0.025	3.84
STD6	119.76	0.0175	93.501	6.327	0.129	0.025	7.68
STD8	239.58	0.0175	93.501	6.327	0.129	0.025	15.37

TA-55 CALIBRATIONS
(cont.)

The standards are cans of PuO_2 powder with no attempt to dilute the PuO_2 through the 200-ℓ volume.

Table XIII gives the measurement results for the TA-55 standards and Fig. 15 shows a fit of the data using a quadratic function.

Table XIII. TA-55 Measurement Results

Standard ID	Reals/s	sig Reals	Totals/s	sig Totals	Time (s)
STDR3	21.032	0.327	714.1	0.51	6000
STD4	69.573	0.733	2389.7	0.78	6000
STD6	149.225	1.711	4204.5	1.46	3000
Background	0.414	0.084	756.2	0.23	9000

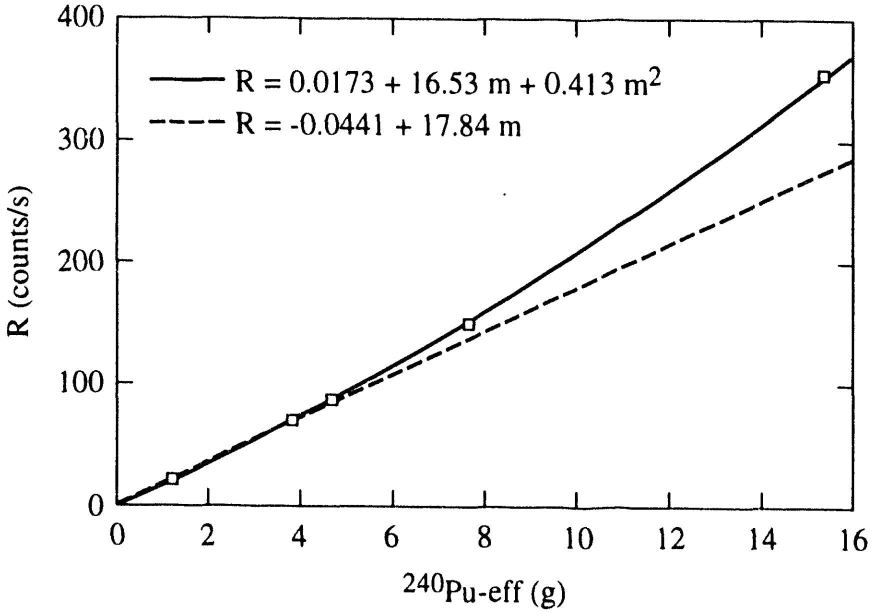


Fig. 15. Calibration curve for NBC after the installation at TA-55. The lower linear curve is the appropriate calibration for plutonium distributed in a 200-ℓ drum.

TA-55 CALIBRATIONS
(cont.)

The PuO_2 powders are concentrated in the small cans so there is significant multiplication for the higher mass values. However for normal waste drums, the plutonium is spread over a much larger volume than the standard cans so the actual multiplication is negligible. Thus, we should use the straight line fit through the smallest two standards that gives a slope of 17.84 counts/s • g ^{240}Pu-eff. This compares well with the TA-35 small-mass standards that gave a calibration slope of 17.64.

MULTIPLICATION

For the normal NBC application to 200-ℓ drums using the add-a-source software (ADDASRC), there is no significant neutron multiplication ($M = 1$) and the assay results are based on the *CF*-corrected reals rates (no multiplication correction). The AS correction changes the *R* rates but not the *T* rates, so the *CF* invalidates any subsequent multiplication corrections.

However, if a high-mass multiplying sample is measured in NBC, the normal neutron coincidence counter software (NCC) can be used and the multiplication-corrected result is available. For this type of application, we have measured the multiplication constant for NBC using the small pellets listed in Table IX.

The result was

$$\rho_0 = \frac{R}{T}(1 = \alpha) = 0.096 \ ,$$

where α is the ratio of (α,n)/spontaneous fission neutrons. The coincidence gate was set at 128 µs and if the gate is reduced to 64 µs, ρ_0 will be reduced by a factor of ~1.4.

DETECTABILITY LIMIT

The detectability limit *d* (in grams of ^{240}Pu) at 3 standard deviations above background was calculated for the counter using the calibration curve and the equation

$$d = (3/a) \bullet \left(\frac{B + ad}{t}\right)^{1/2} \ ,$$

where

DETECTABILITY LIMIT
(cont.)

a = response of counter in counts/(s • g ^{240}Pu) = 17.64,
B = room background reals rate = 0.096 counts/s (shielded location), and
t = counting time = 1000 s.

For the coincidence mode, d = 1.89 mg of ^{240}Pu in the TA-35 shielded location; however, the detectability limit after installation will depend on the background rate at TA-55. Figure 16 shows a graph of the detectability limit vs the neutron coincidence background for NBC.

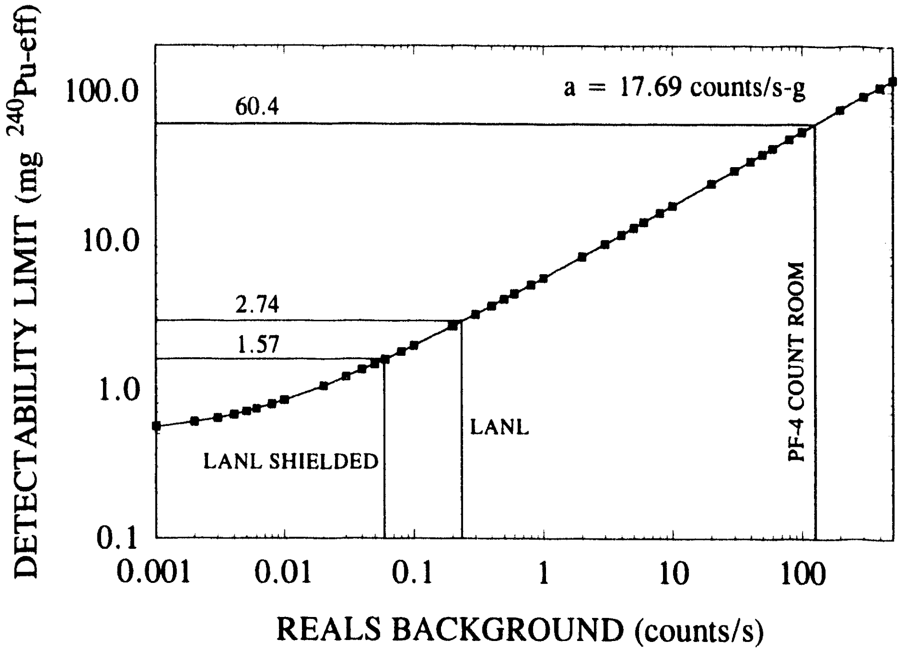


Fig. 16. Calculated NBC detectability limit vs neutron coincidence background for a 1000-s measurement time.

The coincidence background is primarily from cosmic-ray spallation except for the case of a high totals background rate. When the totals rate T is high, then the accidental coincidence rate A can become large. The accidental rate can be calculated from

$$A = (T)^2 G,$$

where G = the gate length in seconds.

DETECTABILITY LIMIT
(cont.)

For example, a high totals background rate of 500 counts/s gives

$A = (500)^2\ (128 \times 10^{-6}) = 32$ counts/s,

that is much higher than the cosmic-ray rate of ~0.2 counts/s. For this example, a 64-µs gate should be used rather than a 128-µs gate. Additional external CH_2 shielding is the most effective method to reduce a high room background (totals rate). For example 10 cm of CH_2 will reduce the background by a factor of ~10, and this gives a factor of ~100 reduction in the accidental coincidence rate.[6]

The detectability limit is a function of the neutron coincidence background, and we have reduced our background by a factor of ~1.8 by eliminating the cosmic-ray spallation events with high multiplicity. The cosmic-ray events can be counted as prompt charged-particle reactions in the detector tubes or as spallation-source neutrons that extend in time over the slowing-down time of the detector body.

The predelay (4.5 µs) eliminates the first category because the events are short lived and the predelay vetoes them from the coincidence gate. The spallation neutrons fall within the coincidence gate but often with high multiplicity. We use the data collection software to isolate the high-multiplicity events and to eliminate them from the data averages. We are currently using statistical techniques to accomplish this. In the future, we could use multiplicity electronics to eliminate these types of cosmic-ray events.

HYDROGEN DETERMINATION

Because the neutron scattering, moderation, and absorption in the drum are dominated by the hydrogen content, we can use the AS measurement to determine the approximate hydrogen content in a drum. The data given in Table XIV have been graphed in Fig. 17 with the AS perturbation (x) as a function of the hydrogen density (ρ_H). For comparison, full density ($\rho = 1$) H_2O has $\rho_H = 0.11$ g/cm^3.

HYDROGEN DETERMINATION
(cont.)

Table XIV. Add-a-Source Perturbation as a Function of Hydrogen Density

Sample	Hydrogen Density	AS Perturbation (x)
Empty	0	0
Polyethylene Shavings	0.0086	0.104
Vermiculite + 29.5 kg of Poly Beads	0.0222	0.496
Polyethylene Tubes	0.031	0.649
Vermiculite + 59.1 kg of Poly Beads	0.0444	1.067

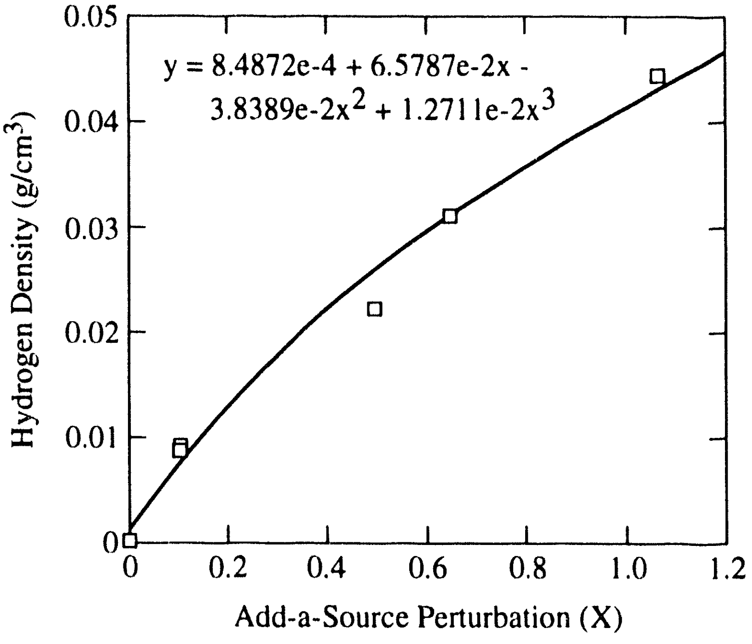


Fig. 17. AS perturbation (x) as a function of hydrogen density in 200-ℓ drums containing CH_2.

HYDROGEN DETERMINATION
(cont.)

For applications of AS to hydrogen determination, calibrations should be performed using a known hydrogen loading in the matrix materials of interest.

Thermal-neutron absorbers, such as boron, in the matrix have almost no effect on the measurement because the AS perturbation x is based on the ratio of R values. The 64 to 128-μs coincidence gate for R eliminates all neutrons that are at thermal energy inside the drum because the flight time to the ^{3}He tubes is longer than the coincidence gate.

The method of neutron moderation has been used in many fields (for example, borehole logging) to determine the amount of hydrogen. The present application of AS to determine the hydrogen is new in that it uses neutron coincidence counting. This gives an important improvement to the hydrogen measurement because the coincidence timing criteria prevent the results from being dependent on the impurities that absorb thermal neutrons.

NORMALIZATION PROCEDURE

The standard method of using a ^{252}Cf source to correct for efficiency changes in the detector system should *not* be used when using the AS procedure. The AS correction factor (*CF*) picks up any efficiency changes in the detector system and automatically corrects the final result. However, the correction is based on the assumption that the efficiency change was from the matrix rather than the detector, thus the *CF* calibration would need to be rechecked.

SUMMARY

The NBC provides a sensitive (19.3% efficiency) and accurate method to measure plutonium in 200-ℓ waste drums. Other samples such as boxes that fit into the sample cavity can be measured as well as the drums. The sensitivity of the system will depend on the room background and the cosmic-ray spallation background at the measurement location. The accuracy will depend on the particular matrix type and the plutonium mass level.

For the typical drum-matrix loadings, the detector has been optimized to be insensitive to matrix changes. The detector wall thickness was chosen to give approximately the same counting rate for an empty drum and a drum containing 20 kg of combustible wastes such as paper and rags. As the matrix density increases, the AS correction is effective for correcting the measurement back to the calibration condition.

SUMMARY
(cont.)

The AS correction significantly improves the final assay accuracy as illustrated in Fig. 13. The AS feature also gives the capability to flag outlier samples that contain an unexpected matrix. If neutron shielding material is present in the drum, the AS procedure picks up the problem with good sensitivity.

REFERENCES

1. David W. Miller and Norbert Ensslin, "Drum Counter Design and Operation Manual," Los Alamos National Laboratory report LA-11892-M (July 1990).

2. H. O. Menlove, et al. "WDAS Operation Manual including the Add-a-Source Function," Los Alamos National Laboratory report LA-12292-M (April 1992).

3. H. O. Menlove and J. E. Swansen, "A High-Performance Neutron Time-Correlation Counter," *Nucl. Technol.* **71**, 497-505 (November 1985).

4. J. E. Swansen, P. R. Collinsworth, and M. S. Krick, "Shift-Register Coincidence Electronics System for Thermal Neutron Counters," Los Alamos Scientific Laboratory report LA-8319-MS (April 1980). Also in *Nucl. Instrum. Methods* **176** (3), 555-565 (1980).

5. Phillip M. Rinard, "Shuffler Instruments for the Nondestructive Assay of Fissile Materials," Los Alamos National Laboratory report LA-12105 (May 1991).

6. H. E. Hootman, "Estimation of ^{252}Cf Shielding Requirements," E. I. DuPont de Nemours & Co., Savannah River Laboratory, Aiken, South Carolina, report TID-4500, UC-34 (April 1970).

*U S GOVERNMENT PRINTING OFFICE 1994-0- 573-024/87071

END

DATE
FILMED

4/11/94