

6. PASSIVE NEUTRON MULTIPLICITY COUNTING

N. Ensslin, M. S. Krick, D. G. Langner, M. M. Pickrell, T. D. Reilly, and J. E. Stewart

6.1 INTRODUCTION

6.1.1 Purpose of the Chapter

A nondestructive assay (NDA) technique for plutonium, called passive neutron multiplicity counting, has been developed as an extension of neutron coincidence counting (Ref. 1). The new technique has led to the design and fabrication of neutron multiplicity counters, one of which is pictured in Figure 6.1. The development of new neutron counters has been accompanied by advances in data-processing electronics, analysis algorithms, and data-analysis software. Development activities have been funded primarily by the Department of Energy (DOE) Office of Security Policy, Technology Development Branch. The new technology has led to significantly better measurement accuracy for plutonium metal, oxide, scrap, and residues.



Fig. 6.1. Photo of the Plutonium Scrap Multiplicity Counter, used for accurate assays of plutonium metal, oxide, mixed oxide, or scrap.

This chapter describes the principles of multiplicity counter design, electronics, and mathematics. Existing counters are surveyed, and their operating requirements and procedures are defined. Current applications to different plutonium material types are described and estimates of the expected assay precision and bias are given.

6.1.2 Definition of Neutron Multiplicity Counting

Multiplicity is a word with a multiplicity of meanings! Our use of the word begins with the fact that an important NDA signature for plutonium is spontaneous fission, leading to the nearly simultaneous emission of multiple, indistinguishable neutrons. The number of neutrons emitted in spontaneous fission can vary from zero to eight. The distribution of the number of neutrons is

called the multiplicity distribution. The multiplicity distributions for spontaneous fission in ^{240}Pu and the 2-MeV-neutron-induced fission for ^{239}Pu are illustrated in Figure 6.2.

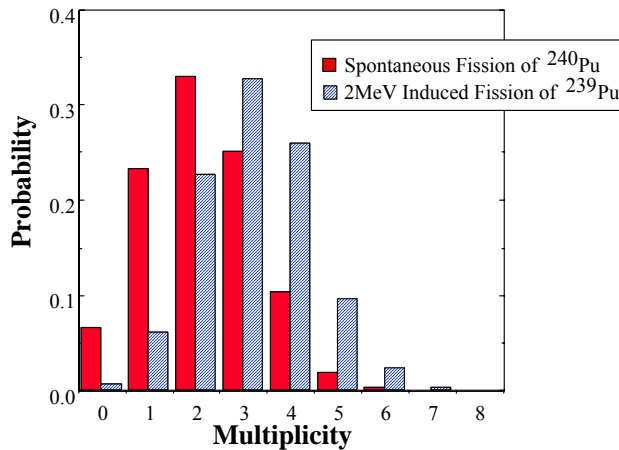


Fig. 6.2. The spontaneous fission multiplicity distribution for ^{240}Pu and the 2-MeV-neutron-induced fission multiplicity distribution for ^{239}Pu .

Multiplicity counting sums up separately the number of 0, 1, 2, 3, 4, 5, 6, 7, etc. neutrons within the coincidence resolving time or “gate width” of the electronics package. This measures the multiplicity distribution of neutrons that are emitted, detected, and counted within the gate width. For this reason, the word multiplicity is specifically associated with the extension of conventional coincidence counting to the collection of higher-order multiples of neutrons. However, we also associate the word multiplicity with a special neutron counter design and with the mathematics of the data analysis process.

In practice, multiplicity data analysis is usually not based directly on the observed multiplicity distribution, but on its factorial moments. The first moment is the “singles,” or “totals,” the second factorial moment is the “doubles” or “reals,” and the third factorial moment is the “triples.” Neutron multiplicity analysis works with all three of these moments, whereas conventional coincidence counting only uses the singles and doubles. Thus, when we use the word “multiplicity,” we really mean that we add a third measured parameter, triple coincidences.

6.1.3 Basic Principle of Neutron Multiplicity Counting

Coincidence counting is an NDA technique that extracts quantitative information from the neutrons emitted by plutonium. Ideally, this information should determine the actual grams of ^{240}Pu -effective in the sample, where this is defined as the mass of ^{240}Pu that would give the same double coincidence response as that obtained from all the even isotopes in the sample:

$$^{240}\text{Pu}_{\text{eff}} = 2.52^{238}\text{Pu} + ^{240}\text{Pu} + 1.68^{242}\text{Pu} . \quad (6-1)$$

Gamma-ray spectroscopy or mass spectroscopy is then used to obtain the isotopic composition of the plutonium, which makes it possible to obtain the total Pu mass:

$$\text{Total Pu} = ^{240}\text{Pu}_{\text{eff}} / (2.52f_{238} + f_{240} + 1.68f_{242}) , \quad (6-2)$$

where f_{238} , f_{240} , and f_{242} are the fractions of the plutonium isotopes present in the sample.

In practice, the neutron flux emitted by the sample can be affected by a number of usually unknown, or incompletely known, sample or detector properties. The list of potentially unknown parameters includes the following:

1. Spontaneous fission rate—the goal of the assay,
2. Induced fission, or sample self-multiplication, and its variation across the sample,
3. The (α,n) reaction rate in the sample,
4. The energy spectrum of the (α,n) neutrons,
5. Spatial variation of the neutron multiplication,
6. Spatial variation in neutron detection efficiency,
7. Energy spectrum effects on detection efficiency,
8. Neutron capture in the sample, and
9. The neutron die-away time in the detector.

Clearly there are potentially more unknowns than conventional coincidence counting can determine. We need N measured parameters to solve for N unknown parameters. Conventional coincidence counting provides only two measured parameters, singles and doubles.

The basic principle of neutron multiplicity counting is the use of a third measured parameter, the triples, so that one can solve for three unknown sample properties, typically the fission rate (mass $^{240}\text{Pu}_{\text{eff}}$), sample self-multiplication, and the (α,n) reaction rate. The fourth and fifth unknowns are discussed in sections 6.6.5 and 6.5.8. The sixth and seventh unknown parameters, related to detection efficiency, are usually eliminated as unknowns by careful counter design and calibration. The other potential unknowns are usually less important, and are assumed to be small or constant.

Because we are solving three equations for three unknowns, the solution is exact, complete, and self-contained! This has some interesting consequences. For samples that meet the assumptions in the derivations, the assay is bias free and accurate within counting statistics. However, if a sample does not meet the assumptions, the assay may be biased. In principle, there is no need for calibration with physical plutonium standards, but it is nevertheless important to use standards for validation and bias reduction.

6.1.4 Historical Reasons for Multiplicity Counting

Historically, the benefit of passive neutron counting has been the great penetrability of neutrons. Neutrons are often the only way to rapidly assay large, dense samples. Neutrons can usually measure the entire volume of the item and they are not easily shielded, except by hydrogenous materials with or without neutron poisons such as boron. The first neutron instruments used only the total neutron count rate. However, very few plutonium materials could be accurately assayed, as implied by the long list of potential unknowns presented above.

The next development was neutron coincidence counting, which uses the spontaneous fission signature and is not affected directly by (α,n) neutrons. Coincidence counting has had wide application for international safeguards inspections. It has had a more limited application domestically because large errors can occur when measuring impure materials. The fundamental limitation of coincidence counting is that it measures only two parameters. For a typical sample, there are at least the first three unknowns listed above. Therefore, it is usually not possible to obtain accurate assays of impure samples with conventional coincidence counting. One must either assume that the (α,n) rate is known, and solve for mass and self-multiplication (Ref. 2), or assume that self-multiplication is known, and solve for mass and (α,n) rate. If the assumed information is incorrect, large errors can occur. In fact, for many impure or heterogeneous samples, neither the multiplication nor the (α,n) yield can be known beforehand.

Based on the need for better accuracy, the goal of neutron multiplicity analysis is to correctly assay in-plant materials without prior knowledge of the sample matrix. The availability of a third measured parameter makes this possible for many materials, including moist or impure plutonium oxide, oxidized metal, and some categories of scrap and waste. For the design of neutron multiplicity counters, the goal is to obtain detection efficiencies that are high and nearly independent of the sample matrix. A useful multiplicity counter should also provide relatively fast assays. At present, a practical goal for assay precision is 1% relative standard deviation (RSD) in 1000 s. The limiting factor here is the poorer precision of the triples.

6.1.5 Areas of Application for Multiplicity Counting

Passive multiplicity counting has applications in a number of different areas: improved materials accountability measurements, verification measurements, confirmatory measurements, and excess weapons materials inspections. Although the historical motivation for developing the technique was improved accountability measurements of impure plutonium, new applications have arisen in the areas of verification and confirmation because the technique does not require prior knowledge of the sample, or prior calibration with standards. For similar reasons, multiplicity counting is coming into use for IAEA inspections of excess weapons materials, where the goal is to verify materials with limited production records.

Multiplicity counting can be used for all plutonium samples, but the additional information is beneficial primarily on impure samples. For some material categories multiplicity may not be helpful because of the limited precision of the triple coincidences. These materials include small Pu samples, some Pu-bearing waste, or process residues that are so impure that the high (α, n) reaction rate ruins the precision of the triples. For pure Pu metal or oxide, the additional multiplicity information is not needed, and conventional coincidence counting provides better precision and sufficient accuracy. However, if there are any doubts about the Pu purity, the multiplicity and conventional results can be compared, and the more accurate result can be used. Additional information on multiplicity applications and expected performance is provided in Section 6.7.

6.1.6 Advantages and Disadvantages of Multiplicity Counting

The advantages of multiplicity counting are summarized in the following list:

1. The measurement accuracy for impure Pu is much greater than for conventional coincidence counting.
2. Information on sample self-multiplication and (α, n) reaction rate is obtained.
3. Calibration for many material types does not require representative standards.
4. Typical measurement time, 1000–2000 s, is short compared to other techniques.
5. If a high-efficiency multiplicity counter is used for conventional coincidence counting, one can use very short counting times, and obtain somewhat better accuracy.

The disadvantages of multiplicity counting are as follows:

1. Multiplicity counters are more costly than conventional coincidence counters.
2. Multiplicity counters requires somewhat more floor space and height than conventional counters of the same cavity size.
3. The measurement time for good precision on triples, typically 1000–2000 s, is longer than the 100–300 s counting time used for most conventional coincidence assays.
4. For plutonium samples that do not meet the analysis assumptions, some assay biases still remain. These biases must be removed using correction factors, special calibration procedures, physical standards, or calorimetry on outliers.

6.2 MULTIPLICITY COUNTER DESIGN PRINCIPLES

6.2.1 Multiplicity Counter Design Goals

The development of multiplicity counting has led to the development of a new generation of thermal neutron multiplicity counters. Like coincidence counters, multiplicity counters are thermal neutron well counters that use ^3He tubes embedded in polyethylene. However, their design represents an advance in the state-of-the-art. The overall goal of the design process is to minimize the effects of detector-dependent variables, such as those summarized in Section 6.1.3. In terms of these variables, the goals for multiplicity counter design include the following:

1. Maximize the detection efficiency to increase the triple coincidence count rate, which is proportional to the third power of the efficiency, typically 40% - 60%.
2. Minimize deadtime losses by substantially increasing the number of preamp/discriminator circuits used to read out the ^3He tubes, as described in Section 6.4.3. Multiplicity counters utilize 20 or more circuits, as compared to 6 in conventional counters. The triples rate is much more sensitive to electronic deadtime than the doubles and singles rates.
3. Minimize the detector die-away time to decrease accidental coincidences and thereby improve the “signal-to-noise” ratio for triples.
4. Minimize the effects of sample placement, or variable plutonium distribution, by making the radial and axial efficiency profile of the sample cavity as flat as possible.
5. Minimize the influence of the emitted neutron energy spectrum on the efficiency.

6.2.2 Monte Carlo Design Calculations

In the design of multiplicity counters, Monte Carlo (MCNP) and Figure of Merit codes are used to supplement what has been learned from past designs of conventional coincidence counters. The codes can be used to study design choices such as tube placement; number, size, and gas pressure of tubes; tube bank layout; placement of different neutron moderator or reflector materials; the use of cadmium liners; etc. After the design and fabrication of the counter is completed, the counter’s actual efficiency, die-away time, efficiency profiles, and counting precision are compared with the calculations. The results so far have been in excellent agreement, validating this approach. A recent version of the Monte Carlo code, MCNPX, can directly simulate the singles, doubles, and triples count rates from a known neutron source (Ref. 3). Monte Carlo design calculations are typically performed to a 1-sigma precision of 0.5% to 1.0%. Figure 6.3 is a schematic used in the Monte Carlo design of the Plutonium Scrap Multiplicity Counter pictured in Figure 6.1.

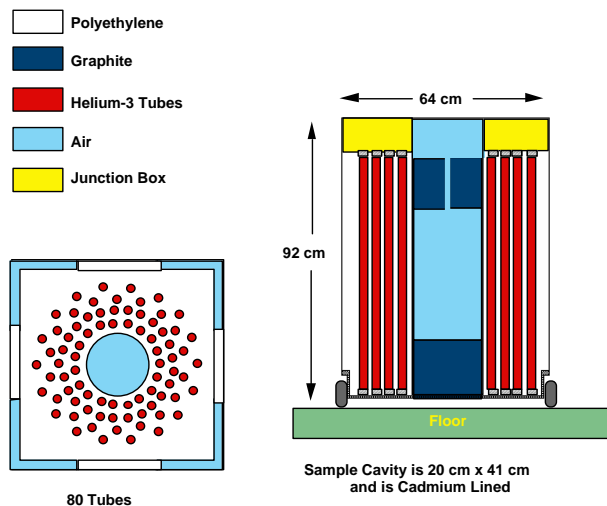


Fig. 6.3. Monte Carlo Design schematic for the Plutonium Scrap Multiplicity Counter (Ref. 2).

The MCNP code can be used to calculate the expected detection efficiency as a function of neutron energy to test the “flatness” of the multiplicity counter design. Multiplicity counters achieve their flat response largely through the use of multiple rings of ^3He tubes placed at different depths in the polyethylene moderator. Figure 6.4 plots the relative count rate for the four rings in the Pyrochemical Multiplicity Counter as a function of neutron energy. The Pyrochemical Multiplicity Counter has 4 rings of ^3He tubes and is specifically designed to measure samples with variable (α, n) yields and neutron energies. Each ring responds differently, but the sum of all four is nearly constant.

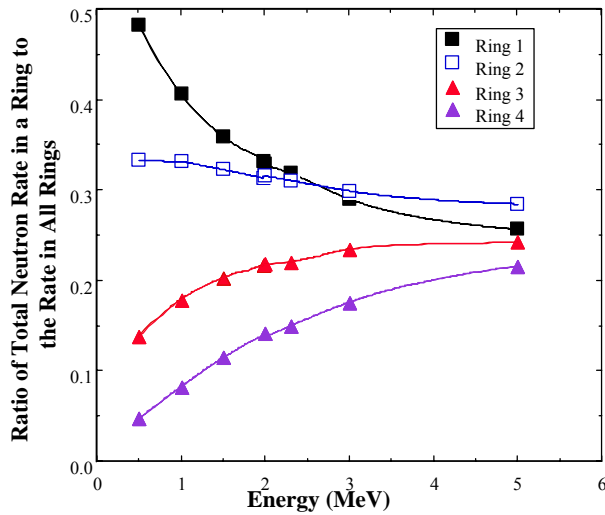


Fig. 6.4. Relative count rates for the four rings in the Pyrochemical Multiplicity Counter as a function of neutron energy. Ring 1 is the inner ring.

6.2.3 Figure of Merit Calculations

Because MCNP provides an estimate for the efficiency and die-away time, a Figure of Merit code can be used to determine the optimum design needed to achieve the desired measurement precision. One Figure of Merit code developed for multiplicity counting analysis (Ref. 4) determines assay variance from the neutron multiplicity distribution. This distribution is predicted from the detector design parameters obtained from MCNP. The expected values of the sample mass, self-multiplication, and (α, n) reaction rate, and the count time, electronic gate width, and pre-delay are entered into the code which then predicts the expected single, double, and triple count rates, and determines the assay variance.

Once the sample mass and size range have been defined, these calculations can be used to define the target efficiency and die-away time needed to obtain a given assay precision in a given time. Figure 6.5 illustrates the expected assay precision vs. ^{240}Pu mass for plutonium oxide samples in a 50% efficient counter, for a 1000-s count time. From this, one can determine whether 50% efficiency is sufficient. For impure samples, the assay precision deteriorates rapidly with increasing (α, n) rates. The Figure of Merit calculation is a fast way to estimate the efficiency, die-away time, and count time needed to provide a given assay precision.

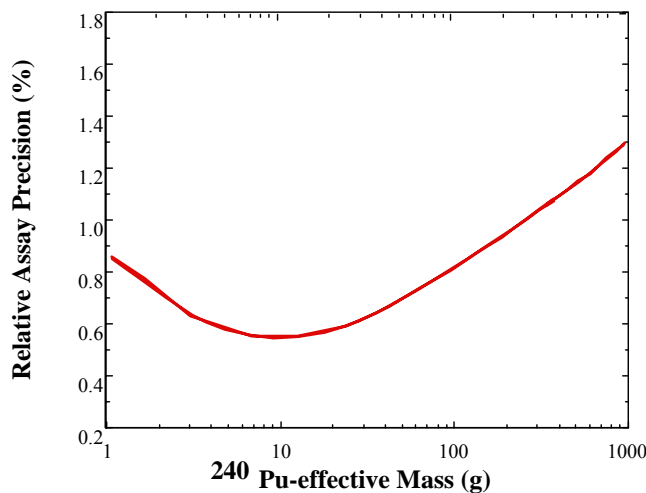


Fig. 6.5. Figure of Merit calculation of expected assay precision (RSD) vs. ^{240}Pu mass for PuO_2 in a 50% efficient multiplicity counter for 1000-s counts.

6.3 SOME EXISTING MULTIPLICITY COUNTERS

6.3.1 Basic Differences between Multiplicity and Conventional Coincidence Counters

Multiplicity counters are similar in construction to coincidence counters. Both are thermal neutron detectors that utilize polyethylene-moderated ^3He proportional counters. Both employ Amptek preamp/discriminators and shift-register-based electronics, although for multiplicity counting the electronics must collect the multiplicity distribution (see Section 6.4.3).

Multiplicity counters are designed to maximize efficiency and minimize die-away time, as described in the previous section. They have much lower deadtimes, and their detection efficiencies are less dependent on energy. Conventional coincidence counters can be used for multiplicity analysis, but their lower efficiencies and longer die-away times lead to very long counting times. Table 6.1 lists some of the multiplicity counters that are currently in use in DOE facilities and provides a summary of their most important features (Ref. 1).

6.3.2 In-Plant (Pyrochemical) Multiplicity Counter

Based on experience gained with earlier developmental counters, the In-Plant or Pyrochemical Multiplicity Counter was designed specifically for in-plant use to optimize the parameters important for multiplicity assay. The counter has two halves, so that it could be installed around a glovebox well. Figure 6.6 is the design schematic used in the Monte Carlo calculations to determine the optimum tube spacing, 1.59 cm, and the best choice of end plug materials. The result is a very high performance counter with a single-exponential die-away curve. The individual ring responses are illustrated in Figure 6.4.

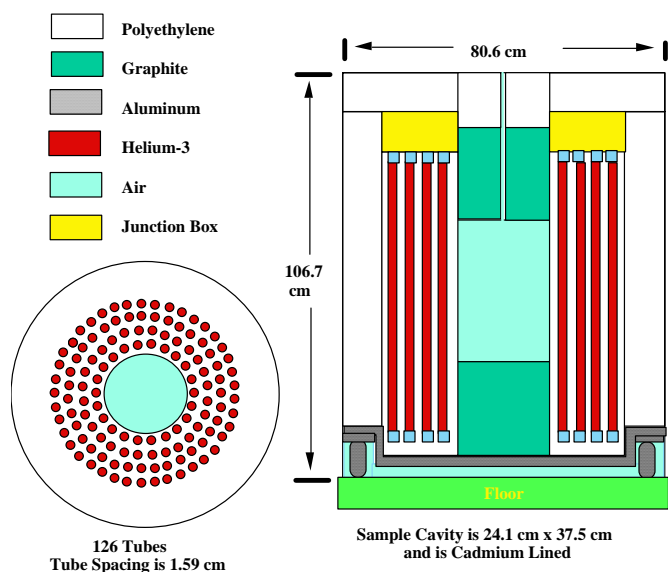


Fig. 6.6. Design schematic for the In-Plant (Pyrochemical) Multiplicity Counter (Refs. 6 and 7).

The In-Plant Counter was used in the Los Alamos Plutonium Facility to assay Pu metal, oxide, and high (α, n) electro-refining salts. The counter was also used at the Livermore Nuclear Materials Facility to assay low and high burnup Pu metal and oxide.

Table 6.1. Survey of Some Multiplicity Counters in DOE Facilities (Ref. 1)										
Multiplicity Counter Name	Location	Application	No. of tube rings	No. of ³ He tubes	No. of Amptek preamps	Derandomizer	Multiplicity deadtime (ns)	Neutron Det. Eff.(%)	Die-Away Time (μs)	Cavity Size
Five-Ring Multiplicity Counter	Los Alamos	Technology Development	5	130	34	Yes	36	53	49	16.5 cm D x 25.4 cm H
Three-Ring Multiplicity Counter	Los Alamos	Technology Development	3	60	12	Yes	83	45	63	20 cm D x 30 cm H
Pyrochemical Multiplicity Counter	Los Alamos	In-plant metals, oxides	4	126	36	No	90	57	47	24 cm D x 38 cm H
Plutonium Scrap Multiplicity Counter	Hanford, Japan	Pu Inventory Verification	3 ½	80	19	No	121	55	47	20 cm D x 41 cm H
ARIES Neutron Counter	Los Alamos	Pu metals and residues	3 ½	80	20	Yes	60	55	47	20 cm D x 41 cm H
FB-Line Multiplicity Counter	Savannah River	Metal, oxide inventory	4	113	24	Yes	50	58	50	20 cm D x 41 cm H
30-Gallon Multiplicity Counters	Rocky Flats, Livermore	Inventory Verification	3	126	54	Yes	25	42	55	30-Gallon Drum
High-Efficiency Neutron Counter	Los Alamos	Waste assay	2	113	16	No	171	32	50	55-Gallon Drum
Epithermal Neutron Multiplicity Ctr.	Los Alamos	Pu inventory Verification	4	121 10atm	27	Yes	37	65	22	20 cm D x 43 cm H
KAMS Neutron Multiplicity Counter	Savannah River	Receipts Verification	3	198 10atm	54	Yes	19	52	37.3	55-gallon Drum
Super HENC Multiplicity Counter	Rocky Flats,	Standard Waste Box Assay		260 10atm	32	Yes		40.3		Standard Waste Box

6.3.3 Plutonium Scrap Multiplicity Counter

The Plutonium Scrap Multiplicity Counter (PSMC) is a relatively compact, high efficiency counter for the measurement of impure Pu and mixed oxide scrap. The PSMC is pictured in Figure 6.1 and a design schematic is given in Figure 6.3. Relative to the In-Plant counter, the PSMC uses many fewer ^3He tubes for nearly the same efficiency. This was achieved by reducing the number of ^3He tubes in each ring in proportion to the decrease in the neutron flux density in the moderator. Thus the outer ring of ^3He tubes is only about half filled, and in Table 6.1 the PSMC is described as having $3\frac{1}{2}$ rings of tubes. The axial efficiency profile is constant to within $\pm 2\%$ over the height of the cavity, making it easier to place most scrap containers within the flat portion of the efficiency profile.

The first PSMCs were used for inventory verification campaigns in Japan and at Hanford, and additional counters are now commercially available. An in-line, active/passive, multiplicity counter very similar in design to the PSMC was developed for permanent installation in the Los Alamos Plutonium Facility (Ref. 5). This counter, called the ARIES Neutron Counter, has a split body so that it can be installed around a glove-box well.

6.3.4 FB-Line Multiplicity Counter

The FB-Line Neutron Multiplicity Counter (FBLNMC) is designed to measure impure plutonium at the Westinghouse Savannah River Site FB-Line Facility. The FBLNMC can be applied to impure samples that range in Pu mass from a few tens of grams to several kilograms; coincidence counting or multiplicity counting can be used. Monte Carlo calculations helped design the high-efficiency (57%) detector, which has 113 ^3He tubes. The axial efficiency profile varies by less than $\pm 2\%$ over the height of the cavity, and the radial efficiency variation over 16 cm is only 1.5% at the midplane of the sample cavity. The energy response profile is identical to that of the Pyrochemical Multiplicity Counter. A derandomizer circuit (see Section 6.4.4) reduces the deadtime by more than a factor of 2, to 50 ns. The individual ring outputs can be read by auxiliary scalars to diagnose sample anomalies. The ratio of rates in the inner and outer rings can also provide a sensitive indication of the mean energy of the neutrons from a sample and is strongly influenced by the sample matrix or (α, n) neutrons.

6.3.5 30-Gallon Multiplicity Counters

The 30-Gallon (109-liter) Multiplicity Counters are an important extrapolation of design concepts to a larger sample volume. The number of ^3He tube rings is reduced to three to save cost, and aluminum corner reflectors help maintain a good spatial response. To facilitate loading heavy drums or ATR400 storage containers, a hexagonal design is used, with the two front sides forming the doors. The mechanical arrangement of the counter and doors is shown in Figure 6.7. The counter has an efficiency of 42% and a die-away time of 55 μs , which is sufficient for assay of bulk plutonium in the kilogram range. Fifty-four Amptek preamp/discriminators and a derandomizer circuit are used to obtain an extremely low deadtime of 25 ns. One 30-Gallon Counter was used at Rocky Flats for IAEA inspections of excess weapons materials (Refs. 6 and 7). Another counter is installed at the Livermore Nuclear Materials Facility, where it is used for inventory verification.

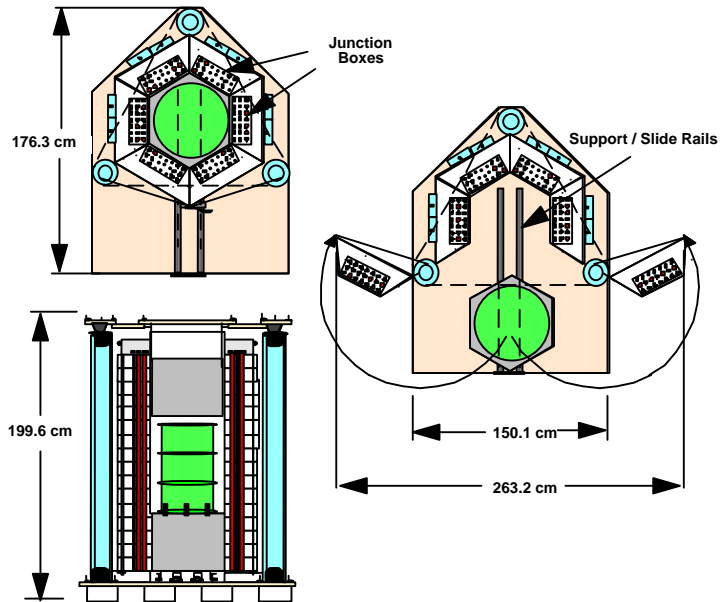


Fig. 6.7. Mechanical schematic for the 30-Gallon Multiplicity Counter.

6.3.6 High-Efficiency Neutron Counter (HENC)

The High-Efficiency Neutron Counter (HENC) is a waste-drum counter developed jointly by Canberra Industries and Los Alamos National Laboratory. This counter was designed to be a high efficiency, low detectability, passive neutron coincidence counter with multiplicity and segmented Add-a-Source matrix correction capability. The design was optimized on the basis of a detectability limit figure of merit, resulting in a limit of 0.5 mg $^{240}\text{Pu}_{\text{eff}}$ by singles counting and 1.7 mg by doubles counting at sea level. An automated drum handling system opens the assay chamber door, loads drums from the conveyor system, and rotates the drums while they are being assayed. Figure 6.8 is a top view of the HENC.



Fig. 6.8 High-Efficiency Neutron Counter (HENC).

6.3.7 Epithermal Neutron Multiplicity Counters

The thermal neutron multiplicity counters described above utilize 4-atm ^3He tubes. A new design concept, the Epithermal Neutron Multiplicity Counter (ENMC), uses 121 10-atm tubes in closely packed rings with less polyethylene moderator (Ref. 8). This enables the ENMC to detect both thermal and epithermal neutrons, resulting in an efficiency of 65% and a die-away time of only 22 μs . For bulk samples of plutonium, ENMC assay times are 5 to 40 times shorter than prior thermal neutron multiplicity counters. The largest relative gains are for the most impure items with high (α, n) rates, where such gains reduce counting times from hours to 20 or 30 minutes. Additional information on ENMC design and performance are given in Chapter 8.

An Inventory Sample Counter (INVS) was designed to fit inside the ENMC sample chamber for assay of small inventory samples. The INVS insert has 21 ^3He tubes in 3 tight rings. The combined ENMC/INVS has an efficiency of 80% and a die-away time of 19 μs . A new drum-sized multiplicity counter has been designed and fabricated for receipts verification at the Savannah River K-Area Material Storage Facility which utilizes 198 10-atm ^3He tubes to achieve an efficiency of 52%.

6.3.8 SuperHENC Multiplicity Counter

The largest neutron multiplicity counter built to date also uses epithermal neutron design concepts. The SuperHENC (Super High Efficiency Neutron Counter) has a cavity large enough to accommodate 1900- ℓ Standard Waste Boxes and also provides Add-a-Source matrix corrections using a ^{252}Cf source that is shuffled in and out of the assay cavity. SuperHENC uses 260 10-atm ^3He tubes to achieve an efficiency of 40.3 %. This counter is mounted in a 14.6 m x 2.6 m x 4.1 m high trailer, Figure 6.9. It was fabricated and used to measure transuranic waste containers before shipping to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico (Ref. 9).

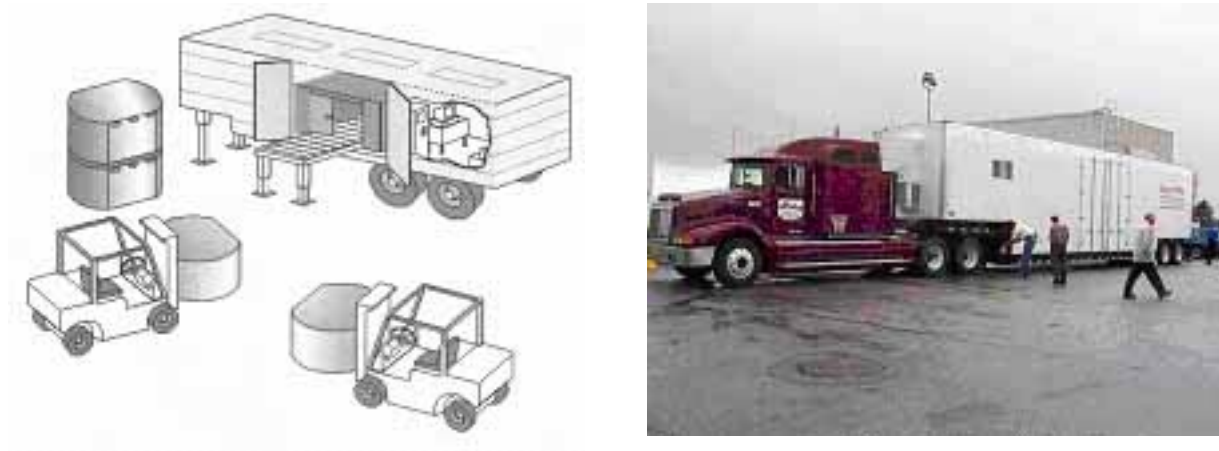


Fig. 6.9. SuperHENC Multiplicity Counter .

6.4 MULTIPLICITY ELECTRONICS

6.4.1 Thermal Neutron Detection Electronics

The electronics for thermal neutron multiplicity counters are similar to those used for conventional coincidence counters. They are based on the detection of neutrons with ^3He

proportional counters embedded in a polyethylene moderator (Refs. 1 and 10). Modern thermal coincidence and multiplicity detectors both utilize Amptek circuits to amplify the ^3He output pulses from the ^3He tubes and convert the pulses above a discriminator threshold to digital pulses. Multiplicity counters usually have 80-130 ^3He tubes, and many more Amptek circuits to provide very short electronic deadtimes.

Amptek A-111 hybrid circuits have a charge-sensitive preamplifier, an amplifier with a bipolar output, a discriminator set to provide 50-ns output pulses, and a pulse-shaping circuit. They provide sufficient gain and signal-to-noise ratio if the ^3He tubes are operated at 1680 V. The Amptek time constant is set to about 150 ns which is the minimum recovery time before they can provide another output pulse. Each Amptek is mounted on a small circuit board that also provides an output pulse to drive a light-emitting diode that flashes when a neutron is detected. Figure 6.10 illustrates one channel processing the input signals from three ^3He tubes.

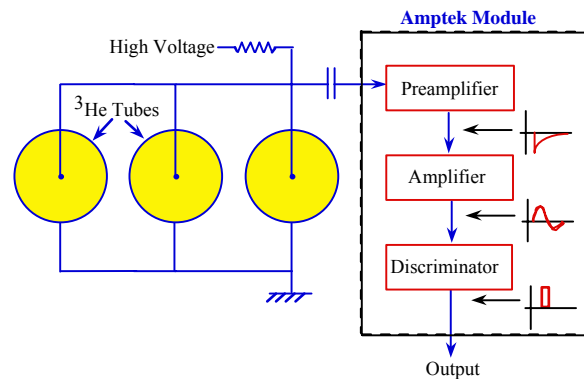


Fig. 6.10. Electronic layout with one Amptek channel processing the input signals from three ^3He tubes (Ref. 1).

The multiplicity counter contains one or more “junction boxes,” where ^3He tubes are mounted to provide good grounding. The junction boxes are split into two horizontal layers, with the bottom one providing high-voltage (HV) distribution lines and isolation capacitors. The top layer holds the Amptek boards, the wires that provide the boards with +5V DC, and the 50-ns discriminator output lines. Also, all of the LED outputs are brought to a single display panel on the side of the junction box. This provides the operator with a quick visual diagnostic to ensure that all Amptek channels are operating. The junction box has an O-ring seal and is equipped with several desiccant holders. This helps prevent breakdown in the HV distribution network, which can cause electronic noise bursts that are detected as spurious coincidences.

Individual Amptek modules are connected via an OR circuit to provide a single input to the shift register circuit used to collect correlated events (Section 6.4.5). The use of many Ampteks greatly reduces the deadtime, because it’s unlikely that neutrons detected at about the same time produce pulses that are processed by the same module. However, the OR-gate does have a small deadtime due to accidental overlaps of the 50-ns input pulses (Ref. 1).

6.4.2 Derandomizer Circuit

The OR circuit described above can be replaced with a “derandomizer” circuit that eliminates this source of deadtime. This is a useful addition to coincidence counters operating at high count rates or to multiplicity counters, because the triples counts are very sensitive to deadtime. The derandomizing buffer holds pulses from the Amptek modules and releases them synchronously with the derandomizer clock (typically 10-16 MHz) such that no pulses overlap. The word “derandomizer” refers to the fact that the input events are now synchronized.

One derandomizing circuit is implemented in a field-programmable gate array (FPGA) installed in the junction box. The derandomizer has 32 input channels that can hold up to three

events from each of 32 Amptek circuits. There are seven outputs: four of eight channels each, two of 16 channels each, and one of all 32 summed channels. The outputs are clocked at a 10-MHz synchronous rate and produce a 50-ns pulse for each input event. With a derandomizer, a conventional shift register can operate at count rates approaching 2 MHz with virtually no synchronizer counting losses. A multiplicity counter can have a triples deadtime correction of about 50% at a count rate of 500 to 900 kcps, but the correction depends on the type of neutron source as well as the count rate.

There is a second derandomizer. If a shift register coincidence circuit is operating with a 4 MHz clock, then each stage of the shift register corresponds to 250 ns. If two input pulses arrive within 250 ns, then one of them can be lost because one stage can only hold one pulse. To prevent this loss, a derandomizer is placed at the input to store pulses until there is space available in the shift register. The use of two derandomizers arose historically. The one at the shift register input is the original one. The Amptek derandomizer was added later to further reduce the deadtime.

6.4.3 The Neutron Pulse Stream and Coincidence Gates

The thermal neutron detector input electronics described above provide a stream of pulses, each representing one detected neutron, to the input of the shift register circuit. The pulse stream contains a combination of spontaneous fission, induced fission, (α,n) neutrons, and external background events. One way to visualize this pulse stream is the histogram shown in Figure 6.11. This is the distribution in time of additional detected neutrons that follow after each detected starting neutron. The data for the plot of count vs. time are obtained from a large number of starting neutrons. The neutrons detected after the starting neutron are counted in time bins according to their detection times relative to the starting neutron time ($t=0$). The starting neutrons can be correlated or uncorrelated to other neutrons. If only random, uncorrelated events are detected, the distribution is on the average constant in time. If correlated events from fission are also present, then the correlated neutrons produce an exponential distribution in time, where the time constant is the detector die-away time. The time distribution is given by

$$N(t) = A + R e^{-\left(\frac{t}{\tau}\right)}, \quad (6-1)$$

where $N(t)$ is the neutron population at time t , A is the accidental or random count rate, R is the real or correlated count rate, and τ is the mean neutron lifetime in the counter (die-away time). In Figure 6.11 the dark red bars represent fission neutrons correlated to the initial pulse (R in Eq. 6-1). The striped bars are from fissions that are not correlated to the starting neutron, because it was a random neutron or from a different fission. The white bars are (α,n) or background neutrons. Note that the accidental coincidences contain both the white and striped bars. Figure 6.11 also shows two coincidence counting intervals (or gates), the $R+A$ (Reals plus Accidentals) and A (Accidentals only). These are described in Section 6.4.5 below.

6.4.4 Predelay Circuit

Because of deadtime and pulse pileup in the ^3He tubes, Ampteks, OR gates, or other electronic components, the actual time distribution of Fig. 6.11 does not continue as a rising exponential all the way to $t=0$. So that this loss of events at short times does not bias the measurements, a short shift register called the “predelay” is located between $t=0$ and the $R+A$ gate. This circuit delays the start of the coincidence counting interval until a short time interval (the predelay) has passed. The length of the predelay is based on the speed of the amplifier, the storage capacity of any derandomizer, and the expected count rate. If the amplifier baseline is not fully restored in a time less than the predelay, the effect extends into the $R+A$ gate and a bias results. The Amptek A-111 amplifier requires a predelay of only 3 μs for a bias of less than

0.01% at a count rate of 500 kHz. At very high count rates, as the derandomizer stretches pulse strings out in time, it may create strings longer than the predelay and a bias.

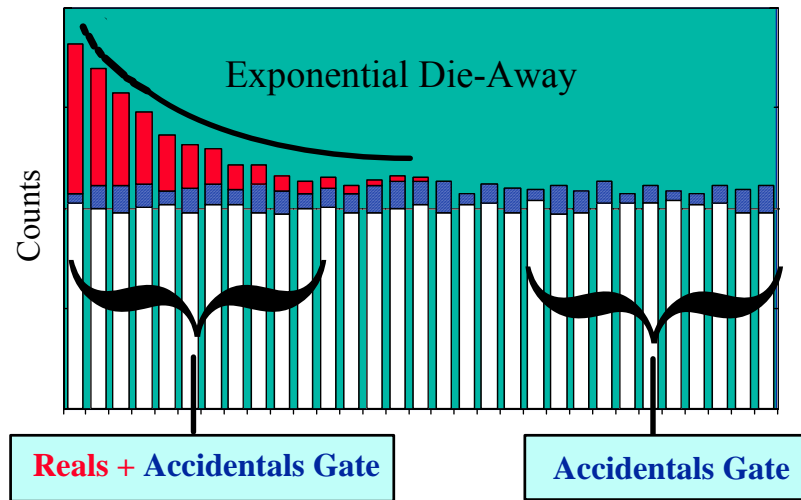


Fig. 6.11. Histogram of detected neutrons in a pulse stream. A measured distribution with exponential die-away is above the histogram, and the (Reals + Accidentals) and (Accidentals) coincidence gates are below the histogram.

6.4.5 Multiplicity Shift Register Basics

To extract the fission rate and multiplicity information from the neutron pulse stream in Figure 6.11, we need to extract the correlated neutrons from the background of uncorrelated ones. The multiplicity shift register circuit achieves this in an elegant fashion by separating the incoming pulse stream into correlated and uncorrelated events. Originally, this was done by storing all incoming pulses for a given interval, the gate-width G , in an integrated circuit called a shift register. The circuit has a series of clock-driven flip-flops linked in stages. For example, a 128-stage shift register driven by a 4-MHz clock ($0.25 \mu\text{s}/\text{stage}$) defines a gate of $32 \mu\text{s}$. Incoming pulses shift through the register one stage at a time and the whole process takes $32 \mu\text{s}$. The shift register collects all possible neutron pairs in an inherently deadtime-free manner.

Operation of a shift register can be visualized by referring to Figure 6.11. This figure shows a prompt gate of width G that opens after the pre-delay and that collects real and accidental coincidences. After a delay much longer than the detector die-away time (up to $4096 \mu\text{s}$), another gate is opened that collects only accidental events. Additional shift registers provide this long delay and second gate. Within counting statistics, the number of accidental events collected in the A scaler is the same as in the R+A scaler. Thus the difference between the counts collected in the R+A gate and those collected in the A gate is the desired real coincidence signal R . Additional details on the operation of the shift register are given in Refs. 1 and 11. In modern coincidence and multiplicity electronics, memory circuits are used to simulate the shift registers.

There is more information in a neutron pulse stream than single and double neutron events. In multiplicity counting, we look at the distribution of 0's, 1's, 2's, 3's, etc. to deduce the multiplicity distribution. Special electronics is required to measure the neutron multiplicity distributions in the R+A and A coincidence gates. These electronics record the number of times each multiplicity occurs in the gates. For example, if seven neutron pulses are in the R+A gate when another neutron arrives, then "1" is added to the R+A counter that tallies sevens. Figure 6.12 is a simplified circuit diagram of the multiplicity shift register electronics.

Separate multiplicity distributions are measured for the R+A and A gates. Table 6.2 shows typical R+A and A distributions obtained from a 60-g plutonium oxide sample measured in a multiplicity counter with roughly 56% efficiency. Each distribution contains the number of times each multiplicity occurred in the corresponding gate. As an example from this table, seven neutron pulses were found 183 times in the R+A coincidence gate, and 42 times in the A coincidence gate.

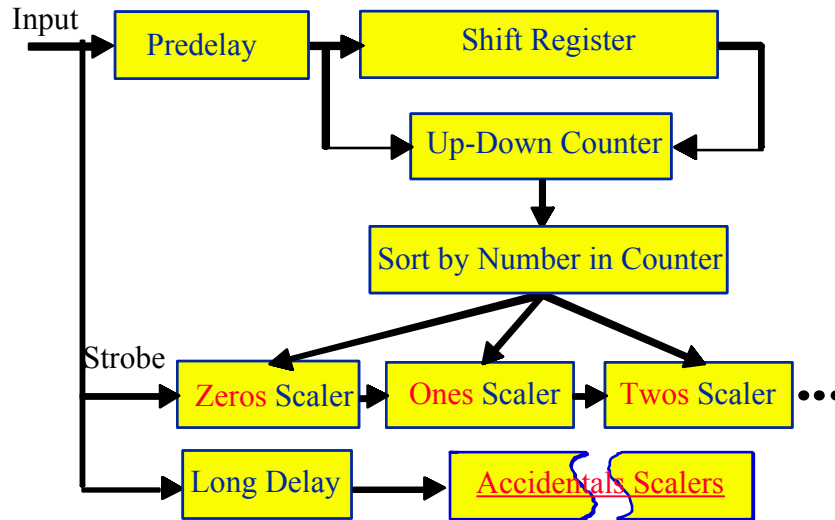


Fig. 6.12. Multiplicity shift-register block diagram.

The sum of all multiplicities in the A distribution (37,153,097) is the total number of triggers, because the singles scaler is situated at the output of the A scaler. The sum of all the multiplicities in the R+A distribution (37,153,123) is not always exactly equal to the total number of triggers because the R+A gate interval is shifted by 4096 μs from that of the A gate. (For a purely random pulse stream, the two distributions are the same within statistical errors.) For a correlated pulse stream, the R+A distribution has more high-multiplicity events, and the A distribution has more low-multiplicity events.

Multiplicity	Counts in (R+A Gate) f(i) distribution	Counts in (A Gate) b(i) distribution
0	26804360	29731130
1	8187530	6222207
2	1772831	1016603
3	325270	157224
4	53449	22387
5	8231	3093
6	1237	402
7	183	42
8	30	8
9	2	1
10	0	0

The two distributions in Table 6.2 can be analyzed to obtain the number of single, double and triple neutron pulses. Note that the number of 1's, 2's, and 3's in Table 6.2 is not what we call the singles, doubles, and triples counts! Instead, the singles is the sum of all the triggers. The doubles is the sum of all the triggers times the mean of the R+A distribution minus the mean of the A distribution, which is the same as a conventional shift register output. The triples is a more complex unfolding of the R+A and A distributions (Equations 6-5 and 6-6 in Section 6.5.5).

In other words, we are using the complete multiplicity distributions to compute their first, second, and third factorial moments which we can relate to singles, doubles, and triples. The reason that we have to measure very high multiplicities, 8's, 9's, 10's, etc., is that the average number of events inside the gate of the shift register is the singles count rate times the gate width. For example, if the singles rate is 100,000 cps and the gate width is 64 μ s, the average number of events in either the R+A or the A gate sampled at random times is 6.4. So even for a purely random neutron source, we will record two R+A and A multiplicity distributions that range from 0's to 15's or 20's, with their peak around 6 or 7.

6.4.6 Deployed Multiplicity Shift Registers

The first stand-alone multiplicity shift register is the MSR4, built as a doublewide nuclear instrumentation module (NIM). The singles counting register is 32 bits deep, and the R+A and A registers are 44 bits deep. There are 256 multiplicity registers for the R+A gate and 256 for the A gate, so multiplicities from 0 to 255 can be accumulated. There is no front panel display, and the unit must be controlled through a serial interface with a computer. Several commercial versions of this are now available. Some versions are designed to be more portable, and others utilize time correlation analyzers to collect all of the time intervals between incoming pulses as they arrive.

A recent development is the Advanced Multiplicity Shift Register (AMSR), which decouples sampling of the R+A and A gates. The A gate is sampled at the clock speed, much faster than the sampling of the R+A gate. This improves the precision of the A rate, thereby reducing count times for all coincidence and multiplicity measurements. The count times are reduced by roughly a factor of 2 for a given precision.

6.5 MULTIPLICITY MATHEMATICS

6.5.1 Assumptions in the Equations

This section provides equations for ^{240}Pu mass and other sample parameters in terms of the neutron time distributions measured by multiplicity counters. The equations are based on fundamental assumptions about the fission process and the process of neutron emission and detection which set limits on their accuracy and range of applicability. To the extent that the theoretical model matches the plutonium samples, the measured singles, doubles, and triples rates provide an exact solution for the $^{240}\text{Pu}_{\text{eff}}$ mass, its multiplication M , and its (α, n) rate. To the extent that the model is not a perfect match, we can expect to encounter some biases or limitations in the multiplicity technique. In this regard, the following assumptions are important to remember:

- (1) It is assumed that all induced fission neutrons are emitted simultaneously with the original spontaneous fission or (α, n) reaction (the superfission concept) (Ref. 12).
- (2) It is assumed that the detector efficiency and the probability of fission are uniform over the sample volume. This assumption is called the "point model" because it is equivalent to assuming that all neutrons are emitted at one point. The weighted point model described in Section 6.5.9 provides some improvements to this assumption.
- (3) It is assumed that (α, n) neutrons and spontaneous fission neutrons have the same energy spectrum, so that the detection efficiency, the fission probability, and the induced-fission multiplicity are the same for both neutron sources. This assumption is not valid for many (α, n) sources.
- (4) It is assumed that the neutron die-away time in the sample/detector combination is well approximated by a single exponential time constant.

6.5.2 The Spontaneous Fission Process

The starting point for the multiplicity equations is the spontaneous fission process in plutonium which provides the assay signature for multiplicity and conventional coincidence counting. The primary source of spontaneous fission neutrons in plutonium is usually the isotope ^{240}Pu , although the other even isotopes also contribute to the yield. The effective ^{240}Pu mass is that mass of ^{240}Pu that would give the same double coincidence response as that obtained from all the even isotopes in the actual sample:

$$^{240}\text{Pu}_{\text{eff}} = 2.52^{238}\text{Pu} + ^{240}\text{Pu} + 1.68^{242}\text{Pu} \quad (6-2)$$

The coefficients 2.52 and 1.68 are the ratios of the spontaneous fission decay rates and second factorial moments of the multiplicity distributions. The available nuclear data on these coefficients has an RSD of about 2 to 3% (Ref. 13). The emitted multiplicity distribution from ^{240}Pu is shown in Figure 6.2, and the spontaneous fission neutron yields of plutonium and related nuclides are tabulated in Ref. 1.

For ^{240}Pu , the average number of neutrons emitted per spontaneous fission, or the mean multiplicity ν_1 , is about 2.154. From the ^{240}Pu spontaneous fission yield of about 1020n/s-g, and the mean multiplicity of 2.154, we can deduce that ^{240}Pu has a spontaneous-fission rate F of about 473.5 fissions/s-g. The spontaneous-fission neutron energy spectrum follows a Maxwellian distribution and has an average energy of about 1.96 MeV. The second factorial moment ν_2 is about 3.789, and the third factorial moment ν_3 is about 5.211. (Published values will vary depending on the nuclear data selected for the computation.)

6.5.3 Description of (α ,n) Reactions

Many heavy nuclei, including the odd and even isotopes of plutonium, decay by alpha particle emission as well as by spontaneous fission. Indeed, alpha decay is much more common than spontaneous fission, even for a heavy nuclide such as ^{252}Cf . The alpha-decay half-lives and yields for these nuclei are tabulated in Ref. 1.

The alpha particles emitted by plutonium have an average energy of about 5.2 MeV, and those emitted by uranium are about 4.7 MeV. Alpha particles of this energy have a very short range and do not escape from even the thinnest sample cans. However, they can have a tremendous effect on the neutron count rate. This is because they lead to (α ,n) reactions in low-Z matrix materials, including oxygen, water, fluorine, etc. We define the ratio of (α ,n) neutrons to spontaneous fission neutrons by the parameter α . For pure plutonium metal samples, $\alpha=0$. For oxides and other elements, (α ,n) reaction yields are tabulated in Ref. 1 and in other publications. From these yields, we can compute α for samples of pure plutonium oxide (with ^{241}Am in-growth) from the following equation:

$$\alpha = \frac{13400f_{238} + 38.1f_{239} + 141f_{240} + 1.3f_{241} + 2.0f_{242} + 2690f_{\text{Am}241}}{1020(2.54f_{238} + f_{240} + 1.69f_{242})} \quad (6-3)$$

The thick target (α ,n) yields for other common elements, and the average energy of the neutrons that are emitted, are also tabulated in Ref. 1. From these tables we can see which low-Z elements are most likely to significantly increase the neutron emission rate of the sample.

Of course, the goal of neutron multiplicity counting is to provide an accurate assay independent of the sample (α ,n) yield and to provide a value for α without other available information. However, samples with $\alpha>0$ may have a neutron energy spectrum different from the spontaneous fission neutron energy spectrum. Fortunately, for the most common element present, oxygen, the average energy of the (α ,n) neutrons is 2.03 MeV, very close to the average of the spontaneous fission neutrons, 1.96 MeV.

6.5.4 Definition of Sample Multiplication

Neutrons from either spontaneous fission or (α,n) reactions can induce fissions in a sample. When a neutron induces a fission, a multiplication event has occurred. Neutron-induced fission is the most common multiplication event, but other reactions such as ($n,2n$) are possible. Some neutrons may be captured without causing the release of any neutrons, as in (n,γ) or (n,p) reactions. Or, neutrons may leak out of the sample without undergoing any interactions.

We define v_i to be the average number of neutrons created by induced fission, and p as the probability that a neutron will induce a fission. A neutron can induce a fission with probability p and disappear with probability $(1-p)$. The total multiplication M_T is the total number of neutrons after multiplication, ignoring capture, divided by the number of neutrons that were started. If 100 neutrons are started in the sample and an additional 59 are created by multiplication events, M_T is 1.59.

We also define a quantity called the net leakage multiplication M_L . This reflects the fact that not all the neutrons from induced fissions will escape the sample, instead some will be captured. M_L is always less than or nearly equal to M_T , depending on the value of p . The following approximate relationship is for the net leakage multiplication:

$$M = \frac{1-p}{1-pv_i} \quad (6-4)$$

We have dropped the subscript L, and from now on we will use M for the leakage multiplication. This is the appropriate quantity for multiplicity counting because it is the measure of the neutrons that escape the sample and are available for detection.

For actual samples, v_i , p , and M will depend on the neutron energy spectrum, the sample composition, and the sample density. There is no direct means of determining M for an unknown sample. It may be calculated by Monte Carlo codes, estimated from the observed doubles to singles ratio when α is known, or calculated from the observed triples to doubles ratio using the multiplicity equations. Because the ratio of triples to doubles is a strong measure of multiplication, this ratio is the primary determinant of M in the multiplicity equations.

Not only does neutron reflection or leakage affect sample multiplication, but sample multiplication itself affects the response of the counter. As M increases, the die-away time of the counter increases because the neutrons that induce fissions create a new source of neutrons. When the die-away time changes, the fraction of coincidence events accepted by the electronics also changes. However, if the detector well is lined with cadmium, thermal neutrons cannot return to the sample, and the increase in die-away time is small and usually negligible.

6.5.5 Measured Singles, Doubles, and Triples Count Rates

The singles, doubles, and triples count rates are obtained from the multiplicity shift register described above. The multiplicity shift register measures the foreground multiplicity distribution $f(i)$ in the R+A gate and the background distribution $b(i)$ in the A gate, as given in Table 6.2. The software computes the factorial moments $f_1, f_2, b_1,$ and b_2 of these distributions, as described in Ref. 1. The singles rate S is the total number of trigger events that arrive at the shift register per unit time. In terms of the factorial moments, the doubles rate D and the triples rate T are given by:

$$D = S(f_1 - b_1) \quad (6-5)$$

$$T = S(f_2 - b_2 - 2b_1(f_1 - b_1))/2 \quad (6-6)$$

Figure 6.13 is a histogram of the multiplicity distributions in the R+A and A gates from a 3.8-kg plutonium metal sample (Ref. 1). The high singles rate from this sample yields multiplicities as high as 20. At these high rates, the R+A and A distributions do not look very different to the eye, so the use of factorial moments and equations 6-5 and 6-6 is essential to unfold the correct doubles and triples count rates. These rates must also be carefully corrected for background count rates and electronic deadtimes.

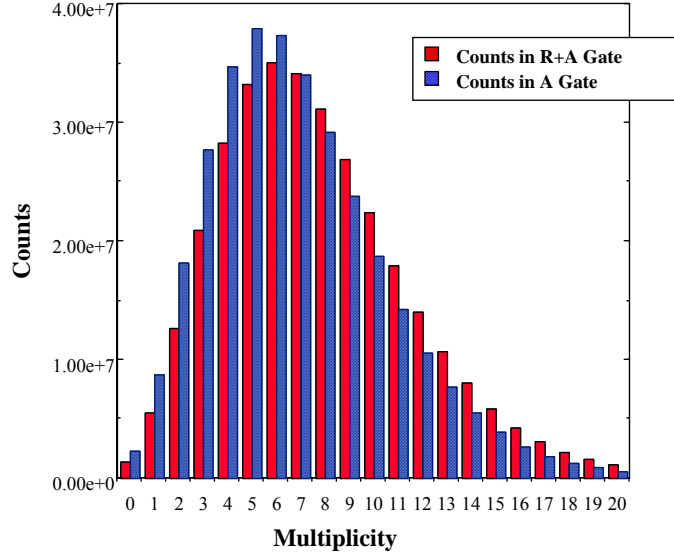


Fig. 6.13. Multiplicity distribution for a 3.8-kg plutonium metal sample measured in a 56% efficient multiplicity counter.

6.5.6 Analytical Definition of Singles, Doubles, and Triples Count Rates

We also need analytical expressions for singles, doubles, and triples count rates in terms of the multiplicity distribution emitted by the sample, detected by the multiplicity counter, and counted by the multiplicity shift register. These equations were derived by Böhnell (Ref. 12) and Cifarelli and Hage (Ref. 14) based on the point model and other assumptions described earlier, and are described in more detail in Reference 1.

$$S = F\varepsilon M v_{s1}(1 + \alpha) \quad (6-7)$$

$$D = \frac{F\varepsilon^2 f_d M^2}{2} \left[v_{s2} + \left(\frac{M-1}{v_{i1} - 1} \right) v_{s1} (1 + \alpha) v_{i2} \right] \quad (6-8)$$

$$T = \frac{F\varepsilon^3 f_t M^3}{6} \left\{ v_{s3} + \left(\frac{M-1}{v_{i1} - 1} \right) [3v_{s2}v_{i2} + v_{s1}(1 + \alpha)v_{i3}] + 3 \left(\frac{M-1}{v_{i1} - 1} \right)^2 v_{s1}(1 + \alpha)v_{i2}^2 \right\} \quad (6-9)$$

where F = spontaneous fission rate,
 ε = neutron detection efficiency,
M = neutron leakage multiplication,
 α = (α, n) to spontaneous fission neutron ratio,
 f_d = doubles gate fraction,
 f_t = triples gate fraction,
 v_{s1}, v_{s2}, v_{s3} = factorial moments of the spontaneous fission neutron distribution,

$v_{i1}, v_{i2}, v_{i3} =$ factorial moments of the induced fission neutron distribution.

The doubles gate fraction f_d is the fraction of the detected doubles that are actually counted inside the gate width G of the multiplicity shift register. For a neutron detector with a die-away time characterized by a single exponential with time constant τ , f_d is given by

$$f_d = e^{-P/\tau} (1 - e^{-G/\tau}). \quad (6-10)$$

where τ = detector die-away time,
 G = shift register gate width, and
 P = shift register pre-delay.

The triples gate fraction is then given by $f_t = f_d^2$ but is usually determined experimentally because the die-away curve for a real detector is not exactly a single exponential.

6.5.7 Final Solution for Sample Mass, Multiplication, α

Eqs. 6-7 through 6-9 relate singles, doubles, and triples rates to the unknown sample parameters, and Eqs. 6-5 and 6-6 calculate the doubles and triples rates from the multiplicity shift register; these are the relationships needed for multiplicity analysis. For measurements of large mass items in small containers, the detection efficiency ε is usually assumed to be a known parameter obtained from careful measurement of a californium reference source. Then Eqs. 6-7 through 6-9 can be solved for the $^{240}\text{Pu}_{\text{eff}}$ mass m_{240} , α , and M . The solution for M is obtained first by solving the following equation:

$$a + bM + cM^2 + M^3 = 0, \quad (6-11)$$

where the coefficients are functions of S , D , and T :

$$a = \frac{-6T v_{s2} (v_{i1} - 1)}{\varepsilon^2 f_t S (v_{s2} v_{i3} - v_{s3} v_{i2})}, \quad (6-12)$$

$$b = \frac{2D [v_{s3} (v_{i1} - 1) - 3v_{s2} v_{i2}]}{\varepsilon f_d S (v_{s2} v_{i3} - v_{s3} v_{i2})}, \quad (6-13)$$

$$c = \frac{6D v_{s2} v_{i2}}{\varepsilon f_d S (v_{s2} v_{i3} - v_{s3} v_{i2})} - 1, \quad (6-14)$$

Once M is determined, the sample fission rate, F , is given by

$$F = \frac{\left[\frac{2D}{\varepsilon f_d} - \frac{M(M-1)v_{i2}S}{v_{i1} - 1} \right]}{\varepsilon M^2 v_{s2}}. \quad (6-15)$$

The second term in the numerator of Eq. 6-15 represents the effect of sample self-interrogation due to induced fission, which must be subtracted from the emitted doubles to obtain the spontaneous fission rate. Once F is obtained, the sample's ^{240}Pu effective mass m_{240} is given by

$$m_{240} = \frac{F}{(473.5 \text{ fissions/s} - g)}. \quad (6-16)$$

Also, the sample's α value is given by

$$\alpha = \frac{S}{F\varepsilon V_{sl}M} - 1. \quad (6-17)$$

If the item's isotopic composition is known, the total Pu mass can be calculated from

$$Pu = m_{240} / (2.52 f_{238} + f_{240} + 1.68 f_{242}). \quad (6-18)$$

where f_{238} , f_{240} , and f_{242} are usually obtained by mass or gamma-ray spectroscopy.

For measurements of low Pu mass items in large containers, such as waste drums, the detection efficiency ε may vary from item to item. In this situation, it may be a good approximation to assume that sample self-multiplication, M , equals 1. Then M can be considered a known parameter, and one can solve Eqs. 6-7 to 6-9 for m_{240} , α , and ε . The equations for this case are given in Ref. 1.

6.5.8 Weighted Point Model Equations

The point model assumptions of constant neutron energy and uniform multiplication can cause significant biases in measurements of impure Pu metal and large, dense Pu oxides. For example, multiplication inside such items is not uniform, but is actually much larger in the center and falls off towards the surface. To correct for these biases, weighted point model equations (Ref. 6) have been developed to add variable-multiplication weighting factors to the expressions in Eqs. 6-8 and 6-9. The weighting factors are obtained from Monte Carlo simulations using the MCNPX code (Ref. 2) which supports the simulation of spontaneous fission sources and can tally the source and detected neutron multiplicity distributions.

The weighting factors are introduced for both the spontaneous fission and (α, n) contributions to the doubles and triples rates to account for multiplication variations. The MCNPX code is used to model items with widely varying geometries, densities, and impurities to obtain simulated singles, doubles, and triples count rates. The weighting factors are then selected to correct for the biases observed in the simulated assays. For unknown samples, the weighted point model equations are used to compute M , α , and m_{240} , but require solving a fifth-order equation for multiplication. Use of the weighted point model equations to correct for multiplication bias is described in Section 6.6.4 below.

6.6 MULTIPLICITY CALIBRATION AND MEASUREMENT CONTROL

6.6.1 Multiplicity Data Analysis Software

Multiplicity analysis software is required for computer control of the electronics, data analysis, calibration and measurement control, and all routine sample measurements. Current software includes the LANL International Neutron Coincidence Counting (INCC) and DEMING Least-Square Fitting codes (Ref. 16) and the Canberra Industries NDA 2000 codes. This software includes background and deadtime corrections, quality control tests, and statistical error analysis. They also can collect and analyze data for passive coincidence counting, active

coincidence counting, and active multiplicity counting. The calibration and measurement control procedures described in this section are based on the INCC code.

6.6.2 Detector Characterization Measurements

A series of detector characterization measurements are required to define the parameters used for data collection, calibration, and measurement control. The room background count rates should be measured. Typical singles, doubles, and triples count rates are 100 to 1000 cps, 1 - 2 cps, and 0.1 - 0.2 cps. Then a well characterized ^{252}Cf reference source should be measured. This provides a normalization measurement to be used for daily measurement control and calibration. The reference source can also be used to determine the detection efficiency, ϵ . The ^{252}Cf reference source is also used to determine the detector die-away time, τ , by making measurements at different gate widths. The coincidence gate width G is typically set to a value similar to the die-away time.

Lastly, multiplicity analysis requires that careful deadtime corrections be applied to the singles, doubles, and especially to the triples count rates. The singles and doubles rates can be corrected for deadtime according to the following equations:

$$S_0 = S_m e^{\delta S_m / 4} \quad (6-19)$$

$$D_0 = D_m e^{\delta S_m} \quad (6-20)$$

where $\delta = A + BS_m$, A and B are the deadtime coefficients, the subscript “m” means measured, and “0” refers to the quantity corrected for deadtime. The coefficients A and B depend on the multiplicity counter, particularly on the number of Amptek circuits. One way to determine A and B is to very carefully and reproducibly measure californium sources of different strengths and adjust A and B to obtain the same doubles/singles ratio for all sources.

The complex equations used for correcting the triples rate are built into the INCC code (Refs. 1 and 16) and use a constant parameter Δ called the multiplicity deadtime. The parameter Δ can be determined by measuring a weak and a strong ^{252}Cf source. The ratio of triples to doubles should be independent of the ^{252}Cf source strength after deadtime corrections, so the deadtime can be determined by adjusting Δ to give the same triples/doubles ratio. For multiplicity counters, typical values of the doubles deadtime coefficient are 0.1 to 0.6 μs , and typical values for the multiplicity deadtime coefficient are 25 to 170 ns. Examples of multiplicity deadtime coefficients are included in Table 6-1.

6.6.3 Multiplicity Calibration Procedure

Because multiplicity counters are used to assay a wide range of impure Pu items, representative physical standards are usually not available. It is possible to calibrate the counter directly by solving the singles, doubles, and triples equations for M , α , and m_{240} using three measured detector parameters: ϵ , f_d , and f_t . To the extent that the plutonium items satisfy the assumptions of the point model, this provides an accurate assay. However, whenever possible, traceable physical standards should be used to validate this procedure or to remove remaining biases caused by errors in the point model assumptions.

The initial determination of the detection efficiency using a ^{252}Cf reference source can be biased because of small uncertainties in the neutron yield of the source and slight differences in energy between ^{252}Cf and Pu fission neutrons. The efficiency should be corrected with Monte Carlo calculations. The magnitude of the adjustment will depend on the detector, but will typically be in the range of 1 to 2%. If Monte Carlo calculations show a significant difference in neutron detection efficiency between a point source and the actual plutonium items, the efficiency can be adjusted accordingly.

The doubles and triples gate fractions are calculated from the singles, doubles, and triples rates measured with the ^{252}Cf reference source using the following equations:

$$f_d = \frac{2v_{s1}D}{\varepsilon v_{s2}S}, \quad (6-21)$$

$$f_t = \frac{3f_d v_{s2}T}{\varepsilon v_{s3}D}. \quad (6-22)$$

In these equations v_{s1} , v_{s2} , and v_{s3} are the factorial moments of the ^{252}Cf source distribution from Ref. 1 or elsewhere. Typical values are $v_{s1} = 3.757$, $v_{s2} = 11.948$, $v_{s3} = 31.636$. If one or more physical standards are available, the calibration can usually be improved by adjusting f_t to obtain the best assays for the standards. This corrects for uncertainties in the nuclear data parameters of ^{252}Cf and plutonium and for differences between the actual items assayed and the point-model assumptions. The adjustment to f_t may be on the order of 10%.

If M or α for the physical standards is known, it may also be helpful to vary ε and obtain the best agreement with the known M , α , and mass values. This approach can only be helpful if M or α is well known. Otherwise, it will introduce a bias into the assays that will increase as M or α increases. In general, if there is no independent information on M or α for the standards, changes to ε are not advisable unless based on Monte Carlo calculations. An exception to this is where a number of well-known samples are available with characteristics very similar to the unknowns. In this case, all three detector parameters can be varied to provide the best assays.

After calibration, it is also helpful to verify the applicability of the multiplicity counting technique by measuring some items to which the technique is going to be applied. The measurements should be verified relative to calorimetry or some other traceable process. If new material categories need to be measured that may not be appropriate for multiplicity counting, some fraction of the measurements should undergo some verification process.

6.6.4 Multiplication Bias Correction

If large Pu metal or dense oxide items are to be measured, a variable bias correction is needed to correct multiplicity assays for the nonuniform probability of fission inside large multiplying items (Ref. 17). This correction is best added during the calibration procedure, but it can also be applied afterwards. The multiplicity software analysis code includes this correction in the form:

$$CF = 1 + a(M - 1) + b(M - 1)^2 \quad (6-23)$$

where M is the measured sample multiplication, and CF is a multiplicative factor that increases the calculated value of m_{240} in Equation 6-16. An empirical set of coefficients appropriate for metal items in different multiplicity counters is $a = 0.0794$ and $b = 0.1386$ (Ref. 17). These empirical coefficients were derived from known plutonium items and are not necessarily applicable to all Pu metal items and all counters. The correction factor approaches 1 as M approaches 1, so it can be left on even if the counter is only used to assay non-metallic items.

The weighted point model equations discussed in Section 6.5.8 provide a more robust bias correction within the framework of the multiplicity analysis equations. Fig. 6.14 compares the performance of the weighted point model on 232 metal samples measured in the Savannah River FB-Line multiplicity counter to the performance on 66 simulated metal cylinders, ranging in shape from pancakes to rods, that were used to determine the doubles and triples weighting factors (Ref. 15). The Savannah River bias correction coefficients are $a = 0.1460$ and $b = 0.1485$. Because the Savannah River and MCNPX curves in Fig. 6.14 are similar, these coefficients may be less dependent on Pu geometry, impurities, or counter cavity size.

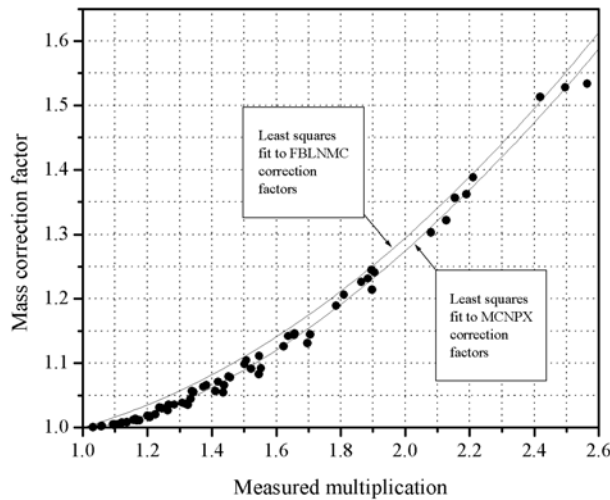


Fig. 6.14 $^{240}\text{Pu}_{\text{eff}}$ mass correction factor vs. measured multiplication for two sets of Pu metal samples. The measured set consists of 232 Pu metal items (upper curve). The computational set has 66 Pu metal cylinders used to determine the doubles and triples weighting factors set (lower curve).

Measurements of impure Pu metal show that the coefficients still vary with high levels of impurities and that the weighted point model can have an alpha-dependent bias for items with α values of 0.5 or more. Nevertheless, an added benefit of the weighted point model equations for impure Pu metals is that the actual α value is determined much more accurately. The standard point model overestimates α for large metal samples, often causing a Pu metal item to appear to be impure when it is actually pure (Ref. 15).

6.6.5 Ring Ratio Diagnostic

Monte Carlo calculations and laboratory measurement campaigns have shown that the ratio of the singles in the inner and outer rings is a good indicator for neutron energy spectrum shifts that may bias the assay. For example, Figure 6.4 plots the relative responses of the four ^3He tube rings in the Pyrochemical Multiplicity Counter as a function of energy. Low energy neutrons are preferentially detected in the inner ring, and high energy neutrons in the outer rings. The neutron energy spectrum affects the overall efficiency, the probability of induced fission, and the induced fission multiplicity distribution. These effects are usually not large, but can cause energy-dependent biases in Pu measurements of items with large amounts of (α, n) emitting impurities or neutron moderators.

Most multiplicity counter electronics packages provide separate measurements of the inner and outer singles count rates and can thereby provide an estimate of the average energy of the (α, n) neutrons. A correction procedure based on ring ratios has been developed for the standard point model, but not yet for the weighted point model. Even if the correction is not used, the ring ratio provides a useful diagnostic that can warn of high levels of impurities or moderators that may bias the assay.

6.6.6 Measurement Control Procedures

Measurement control is used by nuclear facilities to verify proper operation of their multiplicity counters. Quality control tests usually include a checksum test on the shift register electronics, the accidentals/singles test, an outlier test, a measurement control chi-squared limit, a declared-minus-assay limit, and a high voltage test limit (Ref. 16). For all measurements, the total count time should be split up into a series of smaller runs, such as 10 runs of 100 s for a total count time of 1000 s. This allows the outlier test to reject runs with unusually large double or triple coincidence bursts due to cosmic rays or other interferences.

Background runs should be done daily when the instrument is in use, or more frequently if there is reason to believe that the room background is changing significantly. Normalization runs should be done daily using a ^{252}Cf reference source or standard to ensure that the counter is operating correctly. Occasional measurements of a known item or representative standard is a good practice to verify system operation. Other recommended measurement control or assay procedures are described in Ref. 1.

6.7 PASSIVE MULTIPLICITY APPLICATIONS AND PERFORMANCE

6.7.1 Expected Assay Precision

Multiplicity counter precision is determined primarily by the statistical uncertainty in the singles, doubles, and triples counts and the reproducibility of sample placement. The dominant uncertainty is usually in the triples and is determined primarily by detector efficiency, die-away time, count time, count rate, neutron multiplication, and the (α, n) rate. The propagated uncertainty in the Pu mass is usually estimated by the analysis software in one of two ways: from the statistical scatter between the multiple runs that make up a single assay, or from theoretical methods that have been benchmarked against measurements of the observed scatter (Refs. 4 and 16). In either case, the quoted error is not a Total Measurement Uncertainty (TMU), but consists only of counting statistics and calibration uncertainties.

Figure 6.15 provides rough estimates of the predicted assay repeatability due to counting statistics for Pu metal ($\alpha=0$), oxide ($\alpha=1$), scrap ($\alpha=5$), and residues ($\alpha=20$) for a high-efficiency multiplicity counter (Ref. 4). The α values of such materials vary, but the values selected here are representative. Note that the repeatability due to counting statistics is always better for conventional coincidence counting than for multiplicity analysis.

6.7.2 Typical Assay Bias

Assay bias for multiplicity counting is very low for samples that meet the mathematical assumptions of the point model. However, in practice, container and matrix factors may yield noticeable biases. Table 6-3 provides a summary of typical performance for multiplicity assay of nuclear materials commonly found in DOE facilities and can be used to estimate performance for other similar applications. The observed repeatability and bias estimates include the uncertainties from neutron counting, gamma-ray isotopic analysis of the ^{240}Pu effective fraction, and reference values based on calorimetry/isotopics or destructive analysis.

One important question for safeguards personnel is when to use multiplicity counting versus conventional coincidence counting. Factors to be considered in selecting either conventional or multiplicity counting vary with material type. They include Pu mass, (α, n) reactions, available detector efficiency, self-multiplication, neutron energy effects, spatial distribution of fissile material, other matrix effects, available counting time/required precision, and container size and shape. For impure samples with unknown multiplication and α , the accuracy for multiplicity counting is usually much better. However, if the conventional coincidence and multiplicity results are the same within counting statistics, then it may be better to use the more precise conventional results. Other considerations for several major material types are given in the following sections.

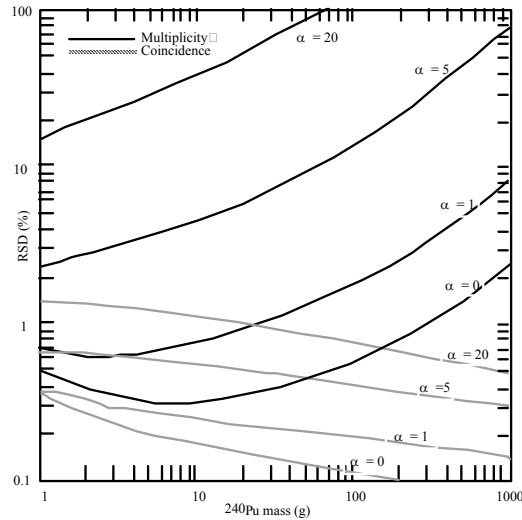


Fig. 6.15. Estimated precision for multiplicity and conventional coincidence assay using a multiplicity counter with a detector efficiency of 50%, a gate width is $64 \mu\text{s}$, a die-away time is $50 \mu\text{s}$, and a predelay of $3 \mu\text{s}$. The background is 100 cps, and the counting time is 1000 s.

6.7.3 Plutonium Metal

Pure plutonium metal has $\alpha = 0$, so conventional coincidence counting (known- α) will give assays with better precision. In reality most metal items contain some impurities, and their surface is usually oxidized. Actual α values range from 0.1 to about 1.0, which would produce unacceptable biases in conventional coincidence counting.

Plutonium metal buttons are dense, compact samples for which the theoretical point model does not correctly describe multiplication. The variable-multiplication correction described in Section 6.5.8 is usually needed to obtain good assay results, as illustrated in Figure 6.14 above. The weighted point model equations have also provided good results at Savannah River (Ref. 15). All of the metal measurements summarized in Table 6.3 have used a multiplication bias correction, except for those taken recently at Livermore with cans placed in the center of the 30-Gallon Multiplicity Counter. The Livermore metal data can be fit with small multiplication bias coefficients, or fit equally well without any correction.

6.7.4 Plutonium Oxide

Pure plutonium oxide yields neutrons from spontaneous fission and from (α, n) reactions on oxygen. Depending on burnup, α is in the range 0.4 - 0.8. If some impurities are present, it is conservative to estimate that $\alpha = 1$. Multiplicity information is not needed if the oxide is so pure that α can be calculated and the known- α approach can determine the mass and the multiplication from the singles and doubles rates. Most oxides in DOE facilities are impure, with α values between 1 and 4; for these multiplicity counting is significantly more accurate than coincidence counting because of induced fissions caused by (α, n) neutrons and changes in multiplication caused by density variations.

Table 6-3. Typical Multiplicity Counter Performance on Various Nuclear Materials.

Nuclear Material Category	No. Items/ Meas.	Pu Mass (g)	(α ,n)/sf Rate α	Count Time (sec)	RSD (%)	Bias (%)
Pu metal	13	200-4000	0 - 1.3	1800	4.6%	1.3%
	14	1500-5000	0	1800	2.7%	-0.1%
	283	5 - 2300	-0.2 - 1.7	1000	2.8%	0.4%
	32	2000	0		0.5%	-0.8%
	6	1100-2100	0 - 0.6		2.2%	2.0%
Calex Std.	8	398	1	1800	1.3%	0.3%
	150	398	1	1000	1.4%	0.8%
Pu oxide	45	500-5000	1	1800	2.2%	0.0%
Impure Pu oxide	12	20 - 875	0.7 - 4.3	1000	2-3%	0.8%
	261	4 - 1800	0.2 - 3.2	1800	2.3%	0.3%
	7	20 - 800	0.6 - 0.8	5000	0.7%	0.5%
Pu scrap	16	80 - 1175	1 - 6	3600	5.7%	-1.6%
	24	2000	1 - 6	1800	5.8%	-1.0%
	17	300 - 1400	2 - 30	1000	11.4%	-2.1%
Pu residues	10	40 - 300	13 - 29	3000	4.8%	0.9%
	8	160 - 340	7 - 34	3600	18.8%	-9.2%
	17	0 - 10	-1 - 82	1000	24%	-2.3%
	21	10 - 40	-4 - 29	1000	22%	1.8%
	16	40 - 400	0.4 - 13	1000	17%	-1.4%
Pu waste	est.	1	5	1000	10%	2 - 5%
U/Pu oxide	8	200-800g	1 - 2	1000	1 - 2%	1-3%
Pu inventory verification	106	1000-4000	1 - 6	1800	4.2%	0.6%
	67	300-1000	1 - 10	1200	8%	0.0%
	90	1 - 4000	1 - 6	6-12 h	10%	-0.5%
	150		0.8 - 4.5	1000	5%	0.2%

Figure 6.16 compares coincidence and multiplicity assays of 8 pure oxide samples and 7 impure oxide samples. The coincidence results include the known- α correction. The pure oxide samples have an average assay/reference ratio of $1.004 \pm 1.4\%$ by coincidence counting, and $1.006 \pm 0.66\%$ by multiplicity counting. The impure oxide samples are $1.039 \pm 8.2\%$ by coincidence counting, and $1.005 \pm 0.68\%$ by multiplicity counting.

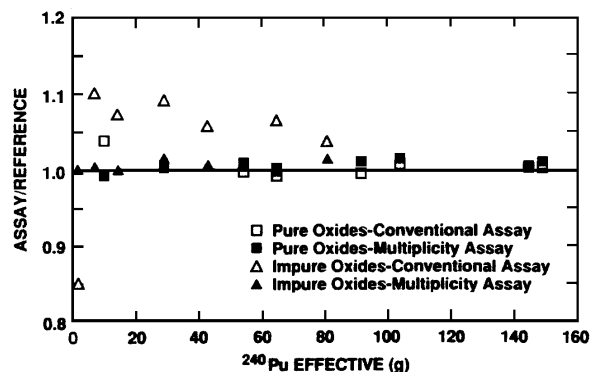


Fig. 6.16. Comparison of conventional coincidence and multiplicity assays of pure and impure plutonium oxide samples.

6.7.5 Plutonium Scrap

Scrap is plutonium-bearing material left over from processing activities that can be recycled. It can include relatively pure metal or oxide, or materials with large quantities of matrix elements like fluorine and beryllium. For multiplicity counting, we define scrap as items with α in the range of 1 to 6. The best assay technique for scrap depends on the nature of the item. An impure metal item is best assayed with multiplicity counting, but an item with very low multiplication and a very high (α, n) rate, like waste, is best assayed with coincidence counting. The selection of multiplicity or coincidence depends on whether the lower bias with multiplicity assay, which can correct for induced fissions, outweighs the loss of precision. Conventional coincidence counting usually provides an upper mass limit because it undercorrects for multiplication.

If the scrap contains moderating materials, or if it emits enough (α, n) neutrons with energies much different from fission neutrons, then the detection efficiency can vary from item to item. The ratio of count rates in the inner and outer rings is sensitive to neutron energy, and very impure scrap items display dramatic changes in their ring responses. Figure 6.17 illustrates the measured ratio of Ring 1 to Ring 4 as a function of mean energy in the In-Plant Multiplicity Counter. The ring ratio can be a valuable tool in identifying items that contain gross impurities and in distinguishing metal from oxide.

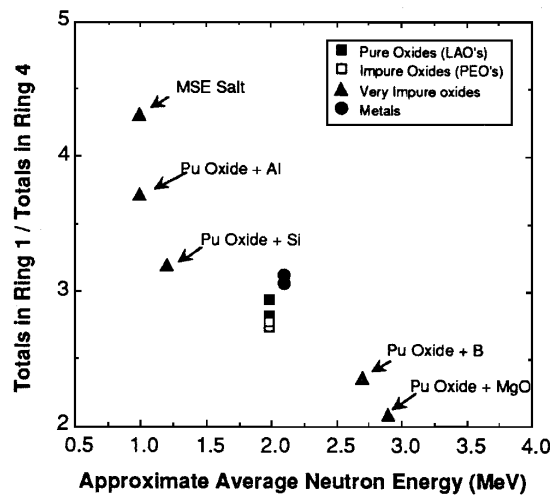


Fig. 6.17. The measured ratio of Ring 1 to Ring 4 as a function of mean neutron energy for various samples in the In-Plant Multiplicity Counter.

6.7.6 Plutonium Residues

Residues are plutonium-bearing materials retained during production operations. They can include ash, combustibles, inorganics, salts from pyrochemical processes, and wet items. Residues are often packaged in large cans, have significant quantities of SNM, and are very heterogeneous and difficult to measure. Because many residues contain large quantities of elements like fluorine and beryllium, they may exhibit α values of 10 to 30 or more. Multiplicity counting is the only feasible passive neutron option, but extremely long count times may be needed to get good triples precision. For samples with unknown (α, n) rates, multiplicity analysis is far less biased than conventional coincidence counting.

A recent example is the measurement of Pu-bearing salts at the Livermore Nuclear Materials Facility with a 30-Gallon Multiplicity Counter. The results are given in Table 6.3 for 3 mass ranges, depending on the success of the chemical separation process that yielded the salts. All multiplicity measurements were referenced to much slower but very accurate

calorimetry/isotopics measurements. Even at high α , the multiplicity results appear unbiased, but with a larger RSD.

For measurements of high- α Pu scrap and residue items, the Epithermal Neutron Multiplicity Counter (ENMC) is a good approach for obtaining much shorter counting times. At Hanford, the ENMC reduced assay times by factors of 8 to 40 relative to a thermal neutron multiplicity counter.

6.7.7 Plutonium Waste

Multiplicity counting can improve the assay of Pu waste in 208-liter drums or Standard Waste Boxes (~1900-liter) even though they contain only a few grams of plutonium. The additional information can flag the presence of shielding materials, detect highly multiplying items, or correct for (α ,n)-induced fissions or detector efficiency variations. The expected assay precision for multiplicity analysis of waste drums has been estimated using a Figure of Merit code and is included in Table 6-3. Multiplicity assay will have poor precision relative to conventional coincidence counting, but may be more accurate because the bias from (α ,n) induced fissions is corrected. (Note that when we use multiplicity analysis to solve for detector efficiency rather than sample multiplication, the RSD increases by a factor of 3 to 4 over the entire mass range and is 5% to 15% at best.) For screening at the TRU-waste detectability limit, multiplicity counting usually does not have sufficient precision.

6.7.8 Mixed Uranium/Plutonium Oxide

Mixed oxides do not meet the assumptions used in the multiplicity mathematics, and must be assayed with caution. The induced fission multiplicity distributions, fission cross sections, and capture cross sections in uranium are different from those in plutonium. If the calibration constants appropriate for plutonium are used to assay plutonium oxides that have a large uranium concentration relative to their plutonium content, the assay results tend to bias low (Ref. 17). If the coefficients are adjusted to fit a particular MOX material with a fixed U/Pu ratio, then the multiplicity performance can be good. Assay of high-burnup MOX items containing a few hundred grams of Pu with the Plutonium Scrap Multiplicity Counter in Japan gives 1% to 2% assay precision.

6.7.9 Plutonium Inventory Verification

Multiplicity counting can be used successfully for inventory verification. The technique provides a better verification than is possible with coincidence counting because it requires less initial inventory information. This section provides some general guidelines on inventory verification measurements, with past results as examples.

For inventory verification, it is helpful to segregate items into categories such as calibration and measurement control standards, Pu metal, low- α Pu (impure oxides and scrap), and high- α plutonium (residues with $\alpha > 6$). These categories can be defined by the observed sample multiplication, mass, α , or measurement precision. For low- α Pu, count times of 1000 to 1800 s are usually sufficient to eliminate counting statistics as a significant contribution to the overall precision. For high- α Pu, multiplicity counting may not be the preferred option because of the long count times required. The overall precision of multiplicity counting for total Pu mass has a lower limit of about 2% RSD once the error on $^{240}\text{Pu}_{\text{eff}}$ determined by gamma-ray spectroscopy is folded in. At some facilities, the use of stream-average isotopics may provide better results and eliminate the time required for gamma-ray isotopics.

For example, Figure 6.18 compares known- α coincidence assay and multiplicity assay for IAEA Physical Inventory Verification measurements at Rocky Flats using a 30-Gallon Multiplicity Counter (Refs. 15 and 16). Multiplicity improved the average agreement between declared and assay by nearly a factor of two over the best conventional approach. Multiplicity

verified 61% of the samples to within $\pm 3\%$, and 100% to within $\pm 18\%$. Overall, 1σ agreement between multiplicity and site declarations was 4.2 % for all items.

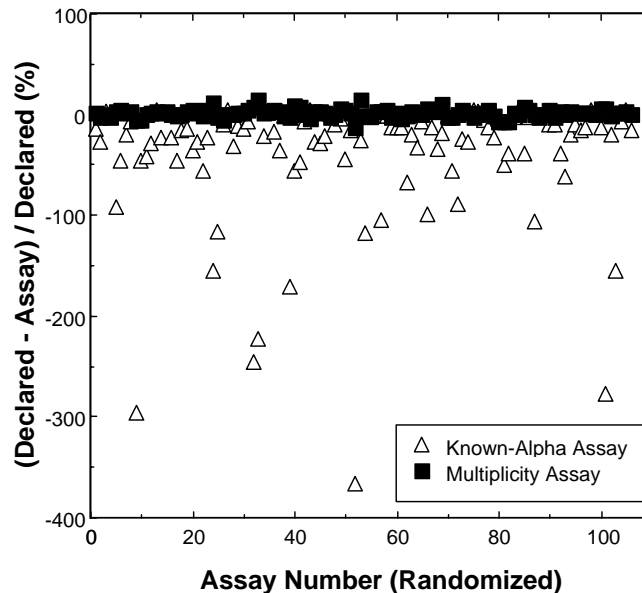


Fig. 6.18. Comparison of known- α conventional coincidence assay and multiplicity assay for some measurements using a 30-Gallon Neutron Multiplicity Counter.

For metal items, the data analysis should include a multiplication bias correction. Inventory verification may also require the assay of storage containers with more than one sample can. Although this does not satisfy the assumptions in the point model, experience does not indicate any observable biases in the multiplicity assay due to this effect. For example, the Physical Inventory Verification exercises at Rocky Flats (Refs. 15 and 16) required measurements of 10-gal. drums, with each drum containing two Pu oxide cans. There was no significant bias in the assay results due to the presence of two cans stacked one above the other.

A novel and difficult application of multiplicity counting to bulk inventory verification at Savannah River was reported in. The inventory consisted of cans of Pu or U/Pu stacked in varying configurations in sealed 18-, 36-, 110-, or 208-liter drums in metal, oxide, scrap, scrub alloy, or other forms. A californium shuffler with multiplicity electronics was used for active assay of uranium and passive assay of plutonium. The shuffler's relatively low efficiency of 18% required long count times, and Pu multiplicity assays were only possible for items with α values up to 6. A potential liability of multiplicity counting was that the sample cans were stacked two or more to a drum. Again, no additional biases were observed in the multiplicity assays that could be attributed to this effect. For this inventory, 73 of the 90 drums containing plutonium were verified to within 25%, and the average bias was only -0.5%.

Multiplicity measurements of bulk Pu items in shielded 9975 shipping containers have been carried out as part of Initial Physical Inventory Verification measurements at the Savannah River KAMS Facility (Ref. 20). The measurements were made with the drum-sized KAMS multiplicity counter, which is designed to correct for the presence of a thick layer of shielding material in each drum. The average assay over reference value for the entire measurement campaign was 1.002.

Experience suggests that a small fraction of the inventory has multiplicity assays well outside the reasonable expected limit of error because of unknown matrix effects that do not meet the point model assumptions. These outliers require calorimetry and/or gamma-ray isotopics to

resolve. However, multiplicity counting can substantially reduce the number of items that require these techniques and thereby allow an increase in measurement throughput.

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