7. ACTIVE NEUTRON MULTIPLICITY COUNTING

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7.1 INTRODUCTION

7.1.1 Definition of Active Neutron Multiplicity Counting

A nondestructive assay technique for bulk, highly-enriched uranium (HEU), called active neutron multiplicity counting, has been developed as an extension of passive neutron multiplicity counting. Development of this technique began about fifteen years ago (Ref. 1, 2, and 3), but there is still only limited literature and field experience. The purpose of this chapter is to document what is known to date and to give the reader a feel for the expected measurement performance of this technique.

As described in Chapter 6, passive multiplicity analysis usually uses neutron singles, doubles, and triples count rates to solve for plutonium mass, multiplication, and alpha. Active multiplicity analysis was developed to provide a similar capability for uranium. The doubles and triples count rates are used to solve for the sample multiplication and the induced fission rate. An Active Well Coincidence Counter (AWCC) or similar instrument is used to induce fissions in the ^{235}U, so the singles rate is not used because it is dominated by the large background from the AmLi interrogation sources. However, active multiplicity counting is more complex than passive multiplicity counting because there is an additional new parameter: the interaction of the AmLi source neutrons with the assay sample which is called the "coupling." The ^{235}U mass depends on both the induced fission rate and the "coupling," and this parameter must also be determined, as described in Section 7.2.4.

This chapter will describe the equations used for active multiplicity, how measurements are made, and what measurement performance can be expected. The rest of this introductory section will describe the historical reasons for the development of this technique, its advantages and disadvantages, and its areas of application.

7.1.2 Historical Reasons for Active Multiplicity Counting

Active neutron coincidence counting using an AWCC equipped with AmLi interrogation sources is a standard technique for assay of ^{235}U, as described in Chapter 17 of Ref. 4. Calibration curves are normally obtained by counting uranium standards and plotting the double coincidence rate vs. ^{235}U mass. Because of neutron multiplication and absorption in the uranium, the calibration curves are nonlinear and are sensitive to the geometry and ^{235}U density of the item. The calibration curve approach works extremely well for many situations, and often yields nearly linear calibrations because of cancellation between multiplication and absorption effects.

To obtain accurate results with the calibration curve approach, the calibration standards must be representative of the assay items. Some of the important sample characteristics are the enrichment, density, geometry, and material composition. However, appropriate calibration curves are not always available, either because suitable standards are not available, or because the important characteristics of the items to be assayed are not well known. For example, uranium metal and uranium oxide items cannot be assayed with the same calibration curve, so representative physical standards of both material types are needed. For large mass HEU metal items, variations in the geometry of the items, or how they are stacked in the can, will change the

self-multiplication dramatically and cause large biases in the assay result. In general, the measurement bias will increase as the characteristics of the calibration standards and assay items diverge.

The goal of active neutron multiplicity counting is to use both the double and triple coincidence rates to provide a new measured parameter that solves for sample self-multiplication and thereby removes most of the bias caused by irregular sample geometry and density. This technique is much less sensitive to differences between the standards and the unknowns, and has a number of potential areas of application.

7.1.3. Areas of Application for Active Multiplicity Counting

Active neutron multiplicity counting has potential applications in shipper/receiver confirmation, inventory verification, or accountability measurements of uranium in difficult material forms. DOE facilities contain many metric tons of uranium in varied materials forms, including mixed oxides and metals, weapons components in shielded storage drums, high-density scrap/waste, remote-handled waste, and non-self-protecting irradiated fuels. The capability for direct accountability, inventory verification, or shipper/receiver verification of the ^{235}U is often not available because of the difficult measurement geometries involved. The use of active multiplicity counting can reduce inventory differences, expedite materials consolidation, and help reduce security and storage costs.

Some specific material forms for which active multiplicity counting may help provide less biased measurements, or provide ancillary information on sample self-multiplication or uranium enrichment, are listed below (Ref. 5). The technique could often be most useful if larger volume counters, or especially faster neutron counters (as described in Chapter 8) were available.

1. Measurement of kilogram quantities of HEU in metal, irregular metal scrap, or very impure oxide form. Uniform metal pieces of constant geometry are best assayed with conventional coincidence counting because of the better precision of the doubles rate.
2. Verification of uranium weapons components or weapons assemblies.
3. Assay of 5A UF_6 cylinders containing 10 to 20 kg of HEU. Measuring the self-multiplication may make it possible to correct for variations in the UF_6 shape that occur during freezing.
4. Accountability or verification of the HEU mass in U/Pu metal scrap, U/Pu oxide, high-density U/Pu waste, or HEU metal contaminated with Pu.
5. Measurement of alternative nuclear materials such as U-233 oxide, Np metal, or Np oxide. This may require development of additional NDA techniques such as combined neutron/gamma-ray/calorimetry or new active/passive neutron-based approaches.
6. Shipper/receiver verification or inventory verification of HEU stored in shipping containers. These are often shielded for personnel dose reduction, making the measurements very difficult. This may require development of new capabilities utilizing neutron generator-based NDA with associated particle imaging.
7. Accountability, verification, or outlier resolution of high-density HEU scrap/waste, low-density HEU scrap/waste, and remote-handled waste for safeguards termination. Better active/passive techniques that provide matrix and geometry-independent results may be required.
8. Accountability or verification measurements of un-irradiated, non-self-protecting U/Pu fuel assemblies, HEU fuel rods, and U-233 fuel assemblies.

7.1.4. Advantages and Disadvantages of Active Multiplicity Counting

The advantage of active multiplicity counting is the ability to provide a more accurate measurement for many of the HEU material forms described above. This is because information on sample self-multiplication is obtained without prior knowledge. As a result, calibration for some material types will require far fewer representative standards, at the cost of reduced accuracy. Fabrication costs for physical standards will be reduced, and assay biases associated with poorly defined items that lack representative calibration standards will be lessened. The measurement time, typically 1000s or more, is still relatively short compared to calorimetry or sampling and analytical chemistry.

The disadvantages of active multiplicity counting include the longer measurement times required to provide good precision on the triple coincidences (at least 1000s compared to the 100 to 300s counting times used for most conventional active coincidence assays). This can be reduced by the use of an active multiplicity counter, but this instrument costs more than a conventional AWCC. Also, the determination of the sample mass is not straightforward, requiring determination of a new parameter, the coupling, as described later.

7.2 ACTIVE MULTIPLICITY EQUATIONS

7.2.1 Overview

The analysis equations for active multiplicity counting are derived from those for passive multiplicity counting, as given in Chapter 6, and are similar in form. They are again based on the measured neutron singles, doubles, and triples count rates. The most important difference is the use of an active well counter equipped with AmLi sources to induce fissions in the ^{235}U. Thus, the fission rate to be solved for is not a passive rate, but an induced one. Also, the measured singles are dominated by the AmLi sources, and are usually not useful for assay. For the same reason, the parameter "alpha," a measure of (α, n) -induced neutrons, is not relevant. The equations are no longer self-contained, but now require additional information to determine the coupling between the AmLi source neutrons and the assay sample.

The active multiplicity analysis equations are also based on the assumptions described in Chapter 6 for passive multiplicity analysis. If actual measurement geometries and samples do not meet all of these assumptions, we can expect to encounter some biases or limitations. Specifically, the "point model" assumption that the neutron detector efficiency and the probability of fission are uniform over the sample volume is not always valid. The neutron counters used for active multiplicity analysis, like the AWCC, were not designed to provide a completely flat efficiency profile across the sample volume. More importantly, the assumption of constant fission probability across the sample volume is not valid for bulk HEU, and a multiplication bias correction has not yet been developed for active multiplicity analysis. The relationships for coupling as a function of multiplication that are described below tend to compensate for these effects, but not completely.

7.2.2 Singles, Doubles, and Triples Count Rates

The neutron multiplicity electronics and software yield the first three reduced factorial moments of the measured neutron multiplicity distribution, which are the singles, doubles, and triples counts, respectively. Theoretically, the singles, doubles, and triples count rates (S, D, T) are given by

$$S = S_0 + B + S_s + FM\varepsilon_f \nu_{s1} \tag{7-1}$$

$$D = \frac{F\varepsilon_f{}^2 f_d \nu_{s2}}{2} \bullet C_d \tag{7-2}$$

$$T = \frac{F\varepsilon_f{}^3 f_t \nu_{s3}}{6} \bullet C_t \ , \tag{7-3}$$

where S_o = singles count rate from the AmLi sources without a sample present,
B = background singles rate,
S_s = change to S_o due to scattering and absorption of AmLi neutrons by the sample,
F = induced fission rate in sample,
M = neutron multiplication
ε_f = efficiency for detecting induced fission neutrons,
ν_{s1}, ν_{s2}, ν_{s3} = first, second, and third reduced factorial moments for AmLi-induced fissions in ^{235}U. (Note that these symbols have a different definition for active versus passive multiplicity counting.)
C_d = a correction factor for self-multiplication of doubles,
C_t = a correction factor for self-multiplication of triples,
f_d = doubles gate fraction, and
f_t = triples gate fraction.

For a neutron detector with a die-away time characterized by a single exponential with a time constant τ, the doubles gate fraction f_d is given by

$$f_d = e^{-P/\tau}(1 - e^{-G/\tau}) . \tag{7-4}$$

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

The triples gate fraction is given approximately by

$$f_t = f_d{}^2 \tag{7-5}$$

but is usually determined experimentally. Note that ε_f is the efficiency for detecting induced fission neutrons. There is also a different (lower) efficiency ε_a for detecting AmLi source neutrons that pass through the shielded endcaps and still get detected.

In principle, the induced singles count rate could be used to determine the ^{235}U mass. In practice, the singles are dependent on AmLi neutron source scattering, neutron absorption, and background fluctuations. For large ^{235}U masses of well-defined geometry, useful assays have been obtained by singles counting if varying background is not a problem. But in general the active multiplicity approach described below, based on doubles and triples count rates, is much more accurate.

7.2.3 Calculation of Sample Self-Multiplication

Expressions for the correction factors C_d for doubles self-multiplication and C_t for triples self-multiplication can be derived from the passive multiplicity equations. In terms of the self-multiplication M, the expressions are as follows (Refs. 1 and 3):

$$C_d = M^2\left[1+\frac{(M-1)\nu_{s1}\nu_{i2}}{\nu_{s2}(\nu_{i1}-1)}\right] \tag{7-6}$$

$$C_t = M^3\left[1+\frac{(M-1)(3\nu_{s2}\nu_{i2}+\nu_{s1}\nu_{i3})}{\nu_{s3}(\nu_{i1}-1)}+\frac{(M-1)^2 3\nu_{s1}\nu_{i2}{}^2}{\nu_{s3}(\nu_{i1}-1)^2}\right] \tag{7-7}$$

where ν_{i1}, ν_{i2}, ν_{i3} = first, second, and third reduced factorial moments for subsequent generations of fission neutron-induced fissions in ^{235}U.

Using Eqs. 7-6 and 7-7 for the self-multiplication correction factors and Eqs. 7-2 and 7-3 for D and T, it is possible to solve for sample self-multiplication without knowing the fission rate F, which cancels out in the ratio T/D. The self-multiplication M can be obtained by solving the following cubic equation (Ref. 1):

$$M^3+M^2\left[\frac{3\nu_{s2}(\nu_{i1}-1)\nu_{i2}+\nu_{s1}(\nu_{i1}-1)\nu_{i3}-6\nu_{s1}\nu_{i2}{}^2}{3\nu_{s1}\nu_{12}{}^2}\right]$$
$$+M\left[\frac{\nu_{s3}(\nu_{i1}-1)^2+3\nu_{s1}\nu_{i2}{}^2-3\nu_{s2}(\nu_{i1}-1)\nu_{i2}-\nu_{s1}(\nu_{i1}-1)\nu_{i3}-(3T/D\varepsilon_f)(f_d/f_t)\nu_{s1}(\nu_{i1}-1)\nu_{i2}}{3\nu_{s1}\nu_{12}{}^2}\right]$$
$$+\frac{3T}{D\varepsilon_f}\frac{f_d}{f_t}\left[\frac{\nu_{s1}(\nu_{i1}-1)\nu_{i2}-\nu_{s2}(\nu_{i1}-1)^2}{3\nu_{s1}\nu_{12}{}^2}\right]=0. \tag{7-8}$$

The triples/doubles ratio provides a good measure of the neutron multiplication, as documented in Ref. 6. A value for the multiplication is needed to perform active multiplicity assays and is useful by itself to help authenticate uranium samples.

7.2.4 Definition of Sample Coupling

Equations 7-1 through 7-3 are similar to those used in passive multiplicity counting. However, for active multiplicity counting, F is the rate at which neutrons from the AmLi interrogation

sources induce first generation fissions in ^{235}U, rather than the ^{240}Pu-effective spontaneous fission rate. Therefore, active multiplicity analysis requires a new parameter called the "coupling" which describes the induced fission rate, F, in terms of the AmLi source strength, Y, and the mass *m* of ^{235}U in the assay sample:

$$F = CmY \tag{7-9}$$

where C = coupling,
F = induced fission rate in sample from the AmLi neutrons,
Y = total output of both AmLi sources (neutrons/s), and
m = mass of ^{235}U in grams.

The coupling depends on the item's geometry, ^{235}U density, chemical and isotopic composition, and location in the assay chamber. In broader terms, it depends on the solid angle between the sample and the AmLi sources, the neutron multiplication in the sample, and other neutron moderation, scattering, and absorption effects. For these reasons, the coupling will not be linear with sample mass.

If we substitute Eq. 7-9 into Eqs. 7-2 and 7-3 for the doubles and triples count rates, the product *Cm* appears together in both equations. Thus we cannot solve for the sample mass *m* without using some additional information to obtain an equation for the coupling. During the development of active multiplicity counting, several different expressions for the coupling have been derived.

The original uranium metal measurements at Savannah River Site and Y-12 (Ref. 2) used a coupling equation of the form

$$CY = a + \frac{b}{m^{1/3}}, \tag{7-10}$$

where *a* and *b* are calibration constants. This equation was selected so that the fission rate F in Eq. 7-9 would have the form $am + bm^{2/3}$. One term is proportional to sample mass, as expected for high-energy interrogation neutrons, and one term is proportional to the sample's surface area, as expected for low-energy interrogation neutrons.

Later work revealed an empirical relationship between the coupling and the multiplication (Ref. 3 and 6). This was an important discovery because the multiplication can be obtained from the triples/doubles ratio independent of sample mass or coupling, as described in the previous section. Monte Carlo calculations for a series of metal and oxide standards (Ref. 7) showed that the coupling was inversely related to the multiplication because of the increasing penetration of the AmLi source neutrons at lower ^{235}U densities. The coupling was also found to be nearly independent of ^{235}U mass and density. A good fit to the data was obtained with the following equation:

$$C = a - \frac{b(M-1)}{1 + c(M-1)}, \tag{7-11}$$

where *a*, *b*, and *c* are calibration coefficients. This relationship between the coupling and the multiplication is illustrated in Figure 7.1 (Ref. 6).

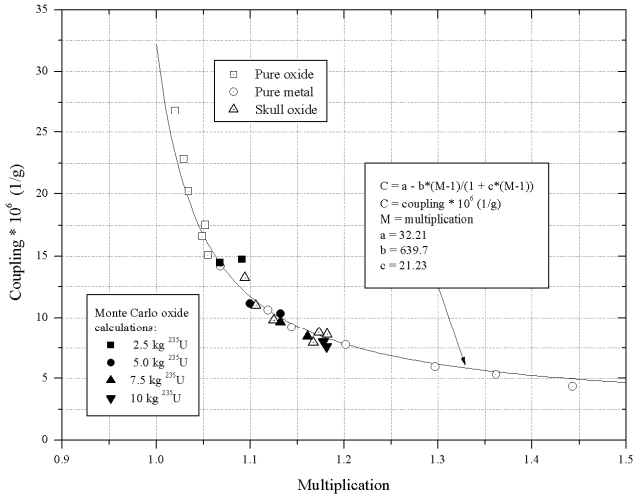

Fig. 7.1 Coupling as a function of multiplication for three categories of uranium samples (Ref. 6).

With the coefficients shown in Figure 7.1, Eq. 7-11 was applied to a series of very impure "skull oxide" items measured at Y-12 (Ref. 6). Because these items had known masses, fill heights, enrichments, and diameter, and were relatively uniform, it was possible to calculate their multiplication by Monte Carlo. The calculated values compared very well with those obtained using the triples/double ratios. The coupling and the multiplication correction factor tend to compensate each other, so that the product is insensitive to ^{235}U mass over a relatively large range. This provides a quantitative explanation for why an almost linear calibration curve of double coincidences versus ^{235}U mass works well for AWCC measurements of many metal and oxide items. For active multiplicity analysis, the use of calibration curves of coupling versus multiplication looks promising for the assay of uranium samples whose detailed characteristics are not known, such as irregular pieces of metal and impure oxide.

A mathematical relationship for the coupling as a function of the solid angle between the sample and the AmLi sources and the sample multiplication has also been developed (Refs. 8 and 9). This relationship can be written as

$$C = k\varphi(\Omega)\varphi(M), \tag{7-12}$$

where $\phi(\Omega)$ represents the dependence of the coupling on the source-sample geometry, $\phi(M)$ represents the dependence of the coupling on the flux depression within the sample, and k is a scaling factor determined from calibration standards. To obtain an expression for $\phi(M)$, this approach derives the "loss-to-fission ratio" of first fission generation neutrons that are leaking out of the sample as

$$\frac{P(loss)}{P(\,fission)} = \frac{\nu_{i1} M}{M-1} - 1\,. \tag{7-13}$$

If leakage is the main loss mechanism, then this ratio is the probability of a fission neutron leaking from the sample divided by the probability that it will induce further fission. And if fission is the main absorption mechanism, then the flux depression of the AmLi interrogation

neutrons is just the ratio of the probability that they will pass through the sample to the probability that they will be absorbed in fission. If a similar functional form is assumed to apply to AmLi neutrons entering the sample, the coupling can be written as

$$C = k\varphi(\Omega)\left(\frac{\nu_{i1}M}{M-1}-1\right). \qquad (7\text{-}14)$$

Once the coupling is determined from calibration curves of coupling versus multiplication using physical standards and/or Monte Carlo calculations, the product of the scaling constant *k* and $\phi(\Omega)$ is calculated by dividing the coupling by the $\phi(M)$ term. The approach works well for disks placed on top of each other in different configurations, as described in Section 7.5.4.

Monte Carlo calculations have also been used to study the coupling for uranium geometries and matrices for which standards are not available (Ref. 10). The calculations used the MCNPX code, which is capable of modeling the induced fission coincidence rate in uranium from an AmLi interrogation source (Ref. 11). The MCNPX results were benchmarked to measurements made with an AWCC, and then the code was used to model one set of metal items and two sets of oxide items in a cylindrical geometry. For cases where sample diameter was constrained, so that solid angle effects were minimized, the relationship between coupling and multiplication is nearly collapsed to a single curve, as shown in Figure 7.2.

The relationship shown in Figure 7.2 is almost independent of item characteristics, with significant divergence only at multiplications below 1.05. The fit shown through the data was determined with a complex combination of linear and exponential functional forms, and is a specific empirical fit to the detector and sample geometries selected for the study. The authors recommend that this approach be used by each facility undertaking active multiplicity measurements until such time as a more generic approach becomes available. Each facility should re-calculate the curve of coupling versus multiplication with their active well counter and their available physical standards, and supplement with MCNP calculations if necessary, in order to obtain the curve needed to analyze their data.

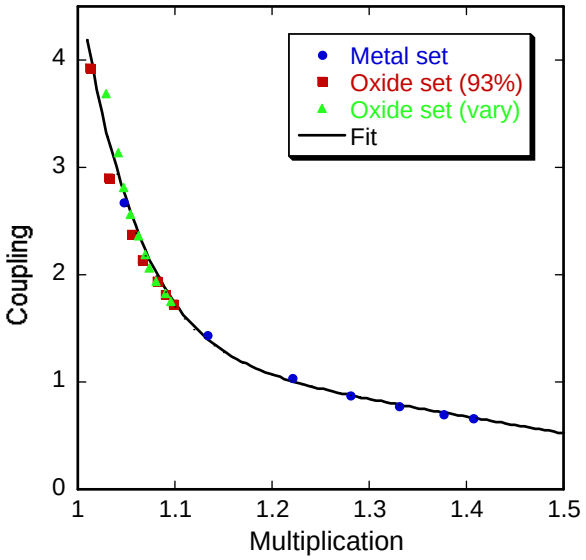

Fig. 7.2. The Monte Carlo modeled relationship between the multiplication and the coupling for three material types: uranium metal, uranium oxide of constant enrichment, and uranium oxide of variable enrichment. The black line is an empirical fit to all the data points (Ref. 10).

7.2.5 Determination of Uranium Mass

Once the sample multiplication M is obtained from the ratio of T/D using Eq. 7-8, the rate *F* of AmLi-induced fissions in the sample is obtained from the doubles count rate (Eq. 7-2), corrected for self-multiplication using Eq. 7-6. A relationship for coupling as a function of M and/or *m* must be selected, using one of the approaches described in the preceding section. The ^{235}U mass *m* is then given by

$$m = \frac{F}{CY}. \tag{7-15}$$

If the uranium enrichment is known, the total uranium mass is the result *m* divided by the enrichment. An alternative analysis method is explained in Ref. 12.

7.3 ACTIVE MULTIPLICITY MEASUREMENT PROCEDURES

7.3.1 Measurement Hardware

Most active multiplicity measurements to date have been carried out with conventional AWCCs that contain AmLi sources to induce fission events in the uranium. The AWCC measurement cavity is typically configured to have a detection efficiency of about 27% to 30% and a neutron die-away of about 52 μs. The only hardware change is the use of a multiplicity shift register to collect neutron single, double, and triple coincidence events. The AmLi sources do not produce a very high count rate, so a variety of existing multiplicity shift registers (including those without derandomizer circuits) or list mode modules can be used for this application. However, the AmLi neutron sources do produce a large number of accidental coincidence events that limit the assay precision, as described in more detail in Section 7.4.

The Epithermal Neutron Multiplicity Counter (ENMC) has also been used for active multiplicity measurements by designing and constructing a new set of active end plugs (Ref. 13). Several different design ideas were studied, including varying the end plug material, recessing the interrogation neutron sources, and shielding the interrogation sources with reflector materials. The optimum solution was to construct the endplugs from polyethylene without the interrogation sources recessed or shielded. Because the ENMC uses higher pressure ^{3}He tubes than the AWCC, the MCNP-calculated efficiency for the active mode was near 55%, with a die-away time around 19 μs. This large reduction in the die-away time relative to the AWCC reduces the number of accidental coincidences from the AmLi interrogation neutron source. Running the ENMC in active mode will reduce the assay time for uranium samples by a factor of approximately ten over the standard AWCC.

In the future, neutron counters designed specifically for active multiplicity counting may become available. There are several design goals for reducing the accidental coincidence background from the AmLi sources. The first goal is to design a counter that minimizes the interrogation neutron detection efficiency and maximizes the induced fission neutron detection efficiency. This will result in less random interrogation neutrons accidentally falling within the

coincidence gate. The second goal is to build a counter with as small a die-away time as possible. This will allow for a reduction in the coincidence gate width that reduces the accidental coincidence rate proportionally. Both goals may be realized by the development of a liquid scintillator-based neutron counter (Refs. 14 and 15). This counter could have a good efficiency of about 25% and a die-away time and coincidence gates as short as 30 ns. This would yield another order of magnitude reduction in assay time for HEU, and allow much smaller samples (about 100g or more) of HEU to be measured by active multiplicity counting. The current status of liquid scintillator counters is described in Chapter 8.

7.3.2 Measurement Software

Active multiplicity analysis requires the use of a software package to acquire and analyze data from the multiplicity shift register. Measurement control options, quality control tests, and calibration and least-squares fitting options are also needed in the software. These tasks can be accomplished with the INCC and Deming codes (Ref. 16), or several commercial software packages that utilize the same analysis algorithms. These codes include data collection and analysis algorithms for passive coincidence counting by several different algorithms, passive multiplicity counting, active coincidence counting, active minus passive coincidence counting, and active multiplicity counting. At this time the active multiplicity option in the INCC code only determines the neutron multiplication from the triples/doubles ratio, but does not determine the uranium mass. The calculation of sample coupling and ^{235}U mass requires the use of a separate spreadsheet. Complete active multiplicity analysis will be added to the INCC code in the near future.

7.3.3 Detector Characterization

Detector characterization measurements and software setup procedures for active multiplicity counting are similar to those described in Chapter 6 and in Ref. 17 for passive multiplicity counting. These measurements will determine the background singles, doubles, and triples count rates, the detector efficiency and die-away time, and the multiplicity shift register gate width and gate fractions. Because of the low count rates encountered in active multiplicity analysis, the coincidence and multiplicity deadtime coefficients can be set to the detector manufacturer's recommended values without further measurements. An exception might be if the detector is to be used for active/passive measurements of U/Pu, which will require very careful determination of the deadtime corrections.

Because active measurements are involved, it is important to fix and document the position and orientation of the AmLi sources in their holders and the position of all cadmium liners and metal reflectors in the assay chamber. Most importantly, the position of the uranium item must be carefully controlled and documented. For example, the shape and magnitude of the coupling curve will depend on whether the items are always placed on a fixed stand, independent of their fill height, or whether they are positioned at different heights to keep the uranium itself in the center of the assay chamber.

7.3.4 Measurement Control and Calibration

Measurement- and quality-control procedures for active multiplicity counting are again similar to those described in Chapter 6 and in Ref. 17 for passive multiplicity counting. These will

usually include a checksum test on the shift register electronics, the accidentals/singles test, an outlier test which rejects runs that lie outside a limit, a measurement control chi-squared limit, a declared-minus-assay quality check limit, and a high voltage test limit. For all measurements, the count time should be split up into a minimum of 10 runs, with an individual length of 10 to 100s. This makes it easier to diagnose electronic noise or instrument drift problems, and makes it possible to use quality control outlier tests. The outlier tests can reject runs with unusually large double or triple coincidence bursts due to cosmic rays. 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. Also, normalization runs should be done daily with a neutron source or physical standard to ensure that the counter is operating correctly.

The calibration process for active multiplicity counting consists of several distinct steps.

1. First one must determine the fission neutron detection efficiency ε_f, the doubles gate fraction f_d , and the triples gate fraction f_t as part of the process of detector characterization. These values are used by the software code to calculate the correct multiplication M for each item using the doubles and triples count rates (Eq. 7-8). Representative physical standards of known multiplication should be used to validate this calculation if possible.
2. A relationship for coupling as a function of multiplication M and/or sample mass *m* must be selected. The best approach to date (Ref. 10) is to develop a new curve of coupling as a function of M with each facility's actual neutron counter and the physical standards or working reference materials available at that site. If necessary, the coupling curve can be supplemented with Monte Carlo modeling of the containers to be measured. However, one cannot model the singles, doubles, and triples count rates accurately because of uncertainties in the AmLi source spectra, the neutron source strength Y, the amount of source shielding in the detector, the exact detector geometry, etc. So the modeling study can provide the relative shape of the curve, but one or more representative physical standards will be needed to provide absolute values.
3. In principle, the multiplication correction and the coupling curve contain enough information to provide calibrated assays using Eq. 7-15. This approach should be validated against representative physical standards or working reference materials if at all possible.
4. The active multiplicity calibration procedure does not need to be repeated unless there is a significant change to the physical configuration of the unknown items to be measured or to the detector, new electronics are installed, or measurement control limits cannot be maintained.

7.3.5 Item Measurement Procedure

Active multiplicity measurements should be made with the uranium content of the item centered in the assay chamber. For example, a 15% difference in response was reported in Ref. 2 by misplacing cans of HEU metal by about 5cm vertically. The item should also be centered radially in the counting chamber to minimize position effects caused by efficiency variations. This counting geometry should be maintained for all standards and assay items.

The count time should be sufficient to provide the desired measurement precision. One percent RSD on the triple coincidence counts is desirable, which typically requires 1000 seconds or more of counting time, as described in Section 7.4.

Item measurement data should be corrected for the small double and triple coincidence background from the AmLi neutron interrogation sources by the multiplicity counter's data acquisition and analysis software. This background is due to electronic pulse pileup and/or source impurities. It may also be necessary to correct for the double and triple coincidence backgrounds from cosmic-ray induced spallation neutrons. This correction is proportional to the total mass of uranium present in the chamber, and can be large at high uranium masses because uranium is a very high-Z material with a good cross section for producing spallation neutrons. It is also good to correct the measurements for spontaneous fission in ^{238}U.

The final solution for the desired ^{235}U mass *m* then requires the following steps:

1. The active multiplicity option in the software code can determine the neutron multiplication from the triples/doubles ratio using Eq. 7-8.
2. Using a separate spreadsheet, the sample coupling is obtained from the previously determined curve of coupling as a function of multiplication M. This curve should incorporate the AmLi neutron source strength Y.
3. The rate F of AmLi-induced fissions in the sample is obtained from the doubles count rate (Eq. 7-2), corrected for self-multiplication using Eq. 7-6.
4. The ^{235}U mass *m* is obtained from Eq. 7-15. If the relationship for coupling involves the uranium mass *m* as well as the multiplication M, then the solution for *m* can be obtained by iteration or by developing a new analytical equation that solves for *m* in closed form. (The small corrections for cosmic-ray induced spallation and ^{238}U spontaneous fission may also require iteration.)
5. The total uranium mass is the result *m* divided by the enrichment.
6. The measurement precision in the triples/doubles ratio and the resultant multiplication M is provided by the software analysis package, but the final uncertainty in the ^{235}U mass is not currently available. The dominant source of counting uncertainty is usually the triples, so this can be estimated and propagated in the final spreadsheet analysis. This would provide a value for the final measurement precision, but not a Total Measurement Uncertainty (TMU) that includes all possible sources of error. Additional information on measurement uncertainties is given in the next section.

7.4 ACTIVE MULTIPLICITY COUNTING PRECISION

7.4.1 Calculation of Expected Counting Precision

As mentioned earlier, active multiplicity counting involves relatively low count rates compared to passive multiplicity counting. Most of the singles events are just due to the AmLi sources, and there are not many doubles and triples from induced fissions. Indeed, the AmLi neutrons are the major source of accidental double and triple coincidences that limit assay precision. This leads to an important question: are enough real triple coincidences detected to make active multiplicity analysis a useful analysis technique for realistic uranium items and practical counting times?

To answer this question, calculations of the expected measurement precision due to counting statistics were carried out with an active version of the Figure of Merit code used for the design of passive multiplicity counters (Refs. 1, 18, 19). The code predicts the expected single, double, and triple count rates and their estimated precision by calculating the factorial moments of the expected neutron multiplicity distribution. The multiplicity distribution does not need to be measured, but is predicted from pre-selected sample and detector design parameters. The code

also computes the triples/doubles ratio and its uncertainty in order to obtain an estimate of the multiplication and its uncertainty. For active multiplicity counting, the Figure of Merit code requires an estimate for coupling. The empirical relationship between coupling and self-multiplication from Figure 7 in Ref. 6 was used. The code was benchmarked by comparison with measurements of bulk uranium metal and oxide samples.

Calculations of the expected measurement precision were done for three detector options: a thermal neutron counter like the AWCC, the Epithermal Neutron Multiplicity Counter (ENMC), and a future liquid scintillator-based fast neutron counter (Ref. 19). The properties of the three detector options are summarized in Table 7-1. All three options assumed a cylindrical sample cavity of about 20 cm in diameter and 25cm high, plus 2.5 cm of lead shielding between the cavity and the neutron detectors for the scintillator-based option. The selection of upper and lower end plug material (polyethylene, graphite, or nickel) affects fission neutron detection efficiency, AmLi neutron detection efficiency, and die-away time. The selection shown in Table 7-1 is intended to optimize active assay precision while still retaining a relatively penetrating AmLi neutron interrogation spectrum in the sample cavity.

Table 7-1. Detector parameters used for performance calculations (Ref. 19).

Active Detector System Name	No. of Neutron Det.	AmLi Source Strength (n/s)	End Plug Material	Fission Neutron Detection Efficiency	AmLi Neutron Detection Efficiency	Die-Away Time (micros)	Coinc. Pre-Delay (micros)	Coinc. Gate Width (micros)
Active We	42	20000	poly	26.7%	6.94%	52	3	64
Epitherma	121	40000	poly	55.0%	16.00%	19	3	28
Liquid Sci	24	1000000	nickel	25.0%	0.60%	0.030	0.005	0.050

7.4.2 Results for Thermal Neutron Counting

Assay precision for a thermal neutron counter was calculated using a standard AWCC configuration with 42 ^{3}He tubes (4-atm) to detect thermal neutrons (Ref. 19). The AWCC was assumed to be in the fast mode, with polyethylene end caps and a cadmium-lined sample cavity. Although the AWCC usually employs two AmLi neutron sources that each emit about 50,000 n/s, the calculations assumed a total yield of 20,000 n/s. This is because Figure-of-Merit calculations for active multiplicity show a minimum in the precision curve with an optimum source yield in range of 20,000 to 50,000 n/s (Ref. 1).

Figure 7.3 illustrates the calculated relative standard deviation (RSD) for active multiplicity assay for the AWCC, the ENMC, and the liquid scintillator, assuming 1000-s counting times (Ref. 19). Counting precision for the AWCC does not achieve 1% unless the item contains 4 kg or more of ^{235}U. By comparison, the curves for conventional coincidence counting can achieve 1% at 2 kg of ^{235}U. The counting precision is always better for conventional coincidence counting than for multiplicity analysis, but in this example it is not much better. But we can conclude that 1% active multiplicity measurements cannot be achieved with an AWCC in 1000-s count times except for large metal items.

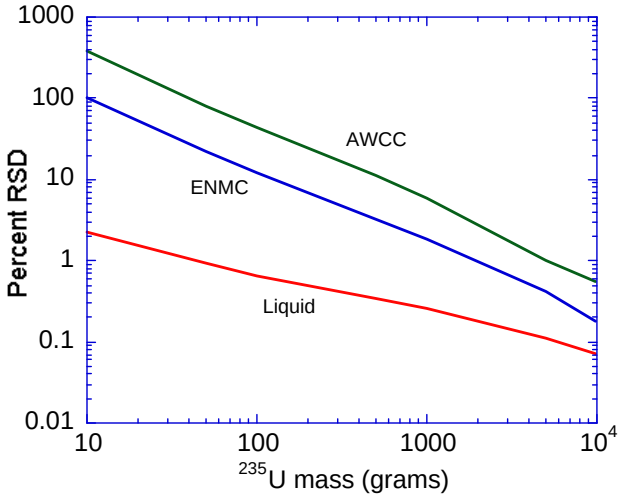

Fig. 7-3. Estimated RSD for the active multiplicity assay as a function of ^{235}U mass (g) in uranium metal for the three detector options summarized in Table 7.1 with 1000-s counting times (Ref. 19).

Benchmark measurements showed that the calculated precision curves were usually within 15 to 25% of the actual observed uncertainties. In-plant measurements at Savannah River and Y-12 confirmed a useful active multiplicity assay precision of 1 to 2% RSD in 1000s counting time for 4 kg or more of ^{235}U metal using a conventional AWCC (Ref. 2).

7.4.3 Results for Epithermal Neutron Counting

The new ENMC (Ref.13) contains more ^{3}He tubes, at 10-atm pressure, and less moderator material than the AWCC, allowing it to detect epithermal neutrons before thermalization. For active multiplicity counting with the ENMC, the predicted counting precision is illustrated in Figure 7.3 (Ref. 19). All calculations again used the parameters summarized in Table 7-1. For the ENMC, the basic cavity design and geometry are similar to the AWCC, and the coupling values should be similar. Over the mass range from 10 to 10,000 g ^{235}U, the ENMC is predicted to have roughly 2 to 4 times lower RSD than the conventional AWCC, which would imply 4 to 16 times shorter counting times for active multiplicity analysis. Stated differently, the ENMC should be able to do active multiplicity analysis of 1 kg or larger samples in 1000 s to within 1% RSD, whereas the AWCC needs 4 kg or larger samples to obtain the same precision.

These predicted count time factors for the ENMC relative to the AWCC were benchmarked by measuring a series of ^{235}U oxide standards, each with about 1 kg of uranium but with different enrichments (Ref. 13). Each standard was counted to a 1% doubles precision in both counters. The counters were configured in a similar manner with the same cavity height and interrogation sources. Shown in Table 7-2 are the count times and the count time ratio (AWCC/ENMC) for each standard. This factor ranges from 5.1 to 10.7 for the doubles count rate over the measured mass range. The triples count rate will have even better ratios.

Table 7-2. Count time factors required to reach a fixed 1% precision for each standard (Ref. 13).

^{235}U Mass (g)	AWCC count time(s)	ENMC count time(s)	Count time factor
116.8	52800	8220	6.4 ± 1.1

128.4	47400	6540	7.3 ± 1.4
170.5	33840	3150	10.7 ± 1.7
265.0	11250	1980	5.7 ± 1.1
372.1	12180	1480	8.2 ± 1.3
515.4	5910	830	7.1 ± 1.1
658.8	3690	720	5.1 ± 0.9
904.0	3500	355	9.9 ± 2.0

7.4.4 Results for a Future Fast Neutron Counter

If faster active neutron assay systems could be developed to increase facility throughput on uranium inventory verification measurements, this would provide large benefits in measurement time and operating cost. For example, wherever the RSD for a measurement is reduced by a factor of 3, the count time required to obtain that RSD is reduced by a factor of 9.

The third detector option in Table 7-1 is a proposed array of very fast NE213 or BC501 liquid scintillators assembled into a fast neutron coincidence counter (Refs. 14 and 15). This option can provide good total neutron detection efficiency if enough liquid scintillator cells are used. Each cell requires pulse shape discrimination electronics to separate neutron recoil events in the scintillator from the Compton-scattered gamma rays. Because fast fission neutrons are detected very quickly in the scintillators, the "die-away time" is really just the time-of-flight of the neutrons from the sample into the detector cells, roughly 30 ns or less for a typical can-sized counting chamber. The coincidence resolving time can then also be extremely short, leading to very low accidental coincidence counting rates. Also, the ability to discriminate against the lower energy AmLi neutrons makes it feasible to use very large interrogation sources and increase the induced fission rate.

For active multiplicity counting, the performance of a liquid-scintillator-based fast neutron counter in Figure 7.3 is predicted to be good enough for 1% or better assay precision over nearly the entire ^{235}U mass range (Ref. 19). If this can be realized in practice, then a liquid scintillator-based counter could provide active multiplicity results for almost all DOE facility inventory materials within 1000-s counting times or less.

7.5 ACTIVE MULTIPLICITY MEASUREMENT PERFORMANCE

7.5.1 Factors that Affect Measurement Performance

This section summarizes the information available to date on active multiplicity measurement performance in DOE facility or laboratory campaigns. As described in the previous section, the limited precision of the triple coincidences in a reasonable counting time provides a limit on the attainable performance. However, other factors may bias the measurement, and their effect may exceed that of counting statistics.

Active multiplicity assays can be biased by a number of usually unknown, or incompletely known, sample or detector properties. These can include spatial variations in neutron self-multiplication or detection efficiency resulting from matrix effects such as neutron moderation, absorption, scattering, or capture. The item's size, shape, and density can affect the coupling parameter, and its radial and axial placement in the well counter can also affect coupling and neutron detection efficiency. If the calibration procedure provides a relationship for coupling as a function of multiplication that is based on some representative physical standards, then many of

the potential sources of assay bias are already folded into this relationship. Other samples with larger variations in matrix effects may still lead to measurements biases. But in most cases the ability to measure sample multiplication and the use of a representative coupling curve makes active multiplicity assays more bias-free than conventional coincidence assays, as described in this section.

7.5.2 Uranium Metal Field Measurements

In-plant active multiplicity measurements of bulk, highly-enriched uranium metal have been carried out at the Savannah River Fuel Tube Fabrication Facility and the Y-12 Uranium Processing Facility (Ref. 2). The measurements were made with standard AWCCs outfitted with multiplicity shift registers. The sample cans contained HEU metal chunks, plates, and cylinders, which typically filled the cans to within several centimeters of the top. The mass of ^{235}U varied from about 0.4 to 17 kg.

Figure 7.4 illustrates one set of Y-12 data on a series of HEU metal cans (and one can of impure oxide) with the observed doubles response plotted as a function of ^{235}U mass. The data show the non-linear effect of the coupling between the AmLi sources and the sample, which causes the response curve to bend down, and the effects of sample self-multiplication, which causes a very high and variable response at a ^{235}U mass of 16 kg. One 16 kg ^{235}U sample was measured three times: as received, with the can tamped down so that the metal pieces would have a closer packing and higher self-multiplication, and with the pieces loosened. This yielded the cluster of the three highest data points on the graph. Also note that the oxide data point does not lie on the same response curve as the metal samples. The scatter in the conventional AWCC assay of these samples, which is based on the observed double coincidence rate, is 25% (RSD).

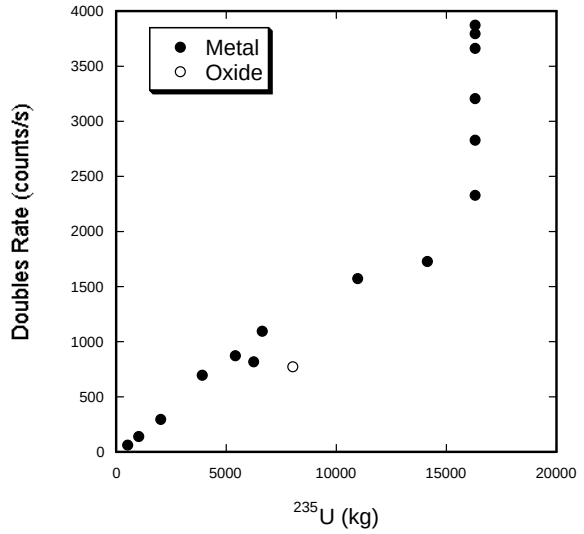

Fig. 7.4. Y-12 data on HEU metal and oxide samples measured in an AWCC. The observed doubles are plotted as a function of ^{235}U mass (Ref.2).

The active multiplicity results are plotted in Figure 7.5, which illustrates the multiplication-corrected double coincidence rate as a function of ^{235}U mass. The corrected rates provide a smooth fit through all of the metal data and the one oxide sample. The non-linear shape of the curve is attributed to the non-linear dependence of the coupling. The active multiplicity assay is consistent with the tag values to within 2% (RSD). This is remarkable agreement, considering the wide variety of samples and the initial, uncorrected scatter of 25% (RSD) (Ref. 2).

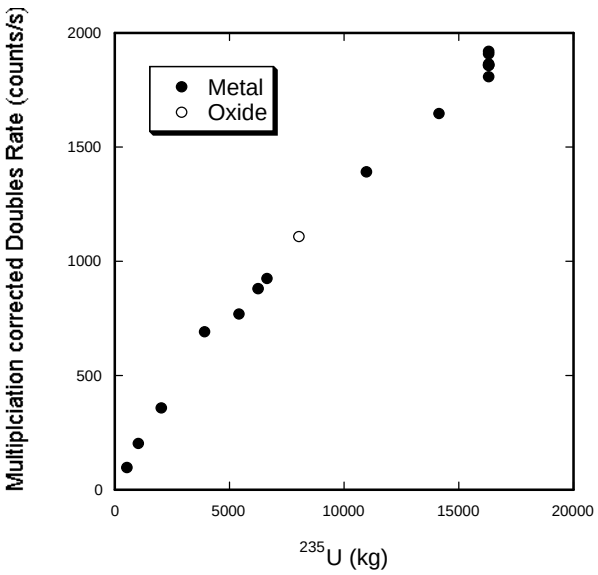

Fig. 7.5. Y-12 data on HEU metal and oxide samples measured in an AWCC. The triples-to-doubles ratio is used to correct the doubles response, and the self-multiplication-corrected doubles are plotted as a function of ^{235}U mass (Ref.2).

7.5.3 Uranium Oxide Field Measurements

Seven skull oxide (very impure U_3O_8) samples were measured at the Y-12 Uranium Processing Facility (Ref. 6). These contained 55% to 84% uranium by weight and also contained erbium, a strong thermal neutron absorber. The measurements were performed in a standard AWCC, but with an erbium liner to reduce the sensitivity of the measurements to that impurity. For the 7 skull oxide containers, the conventional calibration curve fit is 9.1% RSD with a doubles precision of about 1%. The active multiplicity analysis used Eq. 7-11 for the coupling (Figure 7.1) and yields a 4.4% RSD. The standard deviation from counting statistics is about 2%, so the dominant error is from the coupling estimate in Eq. 7-11.

These oxide measurements show a correlation between sample density and the observed triples-to-doubles ratio that is good enough to provide a quantitative correction factor rather than just a diagnostic flag. After the self-multiplication correction is applied, a smooth non-linear response curve is obtained, suggesting that the effects of sample geometry on coupling may be tolerable.

7.5.4 Laboratory Measurements of Uranium Metal Standards

A laboratory measurement campaign with HEU metal disks to evaluate active multiplicity analysis in a controlled setting is reported in Ref.9. An AWCC was used to measure the disks in several different configurations to allow a comparison of conventional calibration curve analysis and active multiplicity analysis. Three measurements were taken with a different number of disks (different mass) in compact geometry for use as calibration points. The rest of the measurements were performed with the pucks arranged in 15 different configurations using aluminum metal spacers, as illustrated in Figure 7.6. The count times ranged from 5 to 10 minutes which generally resulted in a precision in the triples count rate of better than 3%.

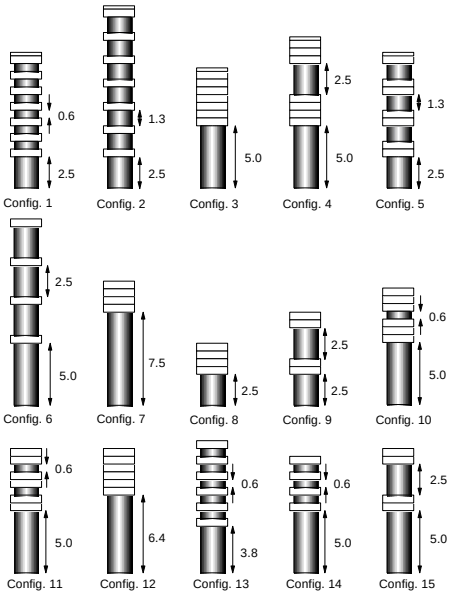

Figure 7.6. The 15 different configurations of uranium disks. The disks are represented by rectangles, and the aluminum spacers are shown as cylinders, with their heights given in cm. Each uranium disk has a mass of approximately 528 grams except for one which has about half the mass. The configurations labeled 3, 7, and 12 were used for calibration.

A conventional calibration curve of doubles count rate as a function of ^{235}U mass was determined from the three calibration configurations using a polynomial expression (Ref. 9). Using that calibration curve, the assays for the remaining 12 configurations are biased low by 6.5% with a scatter of 4.4% (RSD).

For active multiplicity analysis, the three calibration data points were used to solve for the scaling constant in the coupling relationship in Eq. 7-14, using the multiplication values obtained from the triples/doubles ratios. The solid angle factor $\phi(\Omega)$ was assumed to be constant and the actual scaling factor derived from the three calibration standards was the product $k\phi(\Omega)$. This scaling factor was constant to within 1%, suggesting that this approach works well for disks placed on top of each other in different configurations in the center of the assay chamber with no strong radial variations. The ^{235}U masses for the other configurations were then calculated from Eq. 7-15. The results are biased low by only 0.3%, with a scatter of 2.9% (RSD).

The active multiplicity results are in better agreement with the known mass than the calibration curve results, as summarized in Figure 7.7. This improvement is the result of using both the doubles and triples count rate to solve for the multiplication, which is dependent on the uranium material configuration. The current work used the same material form for both the determination of the scaling factor and the assay measurements. Experimental work to determine the sensitivity of the scaling factor to other materials forms is needed.

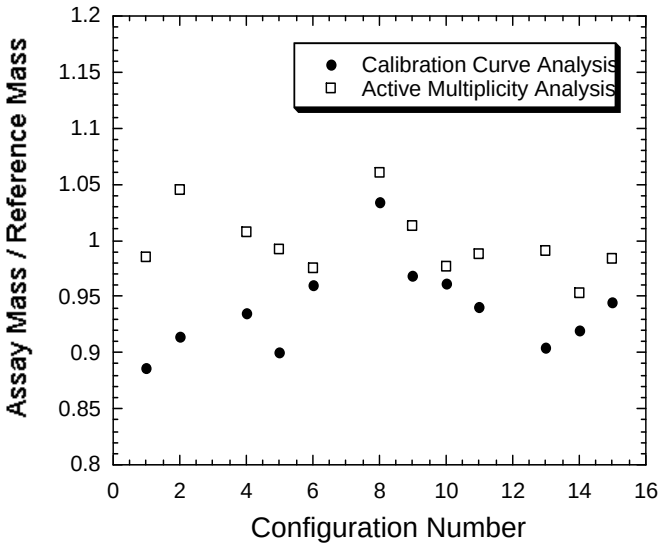

Figure 7.7. Comparison between the calibration curve analysis results (solid circles) and the active multiplicity analysis results (open squares) for the different HEU metal disk configurations. An unbiased result will have an Assay Mass / Reference Mass value of unity (Ref. 9).

7.5.5 Uranium Metal and Oxide Modeling Results

A Monte Carlo study undertaken to better understand the relationship between the multiplication and the coupling as the geometry of the sample varies is reported in Ref. 10. The modeling allows a wide range of material characteristics to be explored. In this case, uranium metal and oxide items in the range of 1 to 3 kg were modeled in a variety of different cylindrical geometries. To benchmark the Monte Carlo calculations, a uranium metal sample was measured in an AWCC. This step is essential because of uncertainties in the AmLi neutron source strength, the AmLi energy spectrum, the exact position of the AmLi sources within their containers, and other spatial variations that affect the doubles and triples gate fractions. These uncertainties limit the absolute agreement between the Monte Carlo calculations and measurements to about 10%, but the calculations can provide good relative comparisons.

The Monte Carlo study modeled a set of metal items, a set of oxide items with a constant enrichment of 93%, and a set of oxide items with constant mass but variable enrichment. The results were analyzed with both the conventional calibration curve and active multiplicity techniques. Calibration curves for the conventional analysis were obtained by fitting the modeled doubles rate versus mass for the three material types. Ten uranium masses (metal or oxide) were chosen randomly, the neutron count rates were modeled, and the results were analyzed with the three conventional calibration curves and an active multiplicity analysis based on a single curve of coupling versus multiplication (Fig. 7.2).

The results of the analysis are shown in Table 7-3. The active multiplicity analysis technique results have a bias 2% with a scatter of 5% (RSD). This technique did much better than all 3 of the conventional calibration curves. For two items, the active multiplicity assays were more than 5% different from the true value. These items were oxide samples with a low multiplication (less than 1.05). The relationship between the multiplication and the coupling at small values of multiplication changes very rapidly with multiplication, and is not as well defined. Any small errors in the multiplication can lead to large errors in the coupling and the mass.

7.5.6 Modeling of Inhomogeneous Uranium Items

A modeling study of inhomogeneous items is reported in Ref. 10. There were situations where facilities needed to measure oxide mixed with metal pieces, or cans of metal chunks. To simulate this, the modeling study distributed 10, 20, or 50 metal spheres throughout a can of uranium oxide. The sphere radius was randomly chosen and varied up to a maximum of either 0.5 or 1 cm. For each combination of the number of spheres and maximum radius, five randomly generated samples were modeled.

Table 7-3. Ratio of the assay mass divided by the true mass used in the Monte Carlo m for 10 randomly generated samples. Shown are the mass ratios for the 3 conventional calibration curves (the metal, constant enrichment, and variable enrichment material typ and the active multiplicity analysis technique.

Item mass (g) and type	Metal Calibration Curve	Oxide Calibration Curve (93%)	Oxide Calibration Curve (variable)	Active Multiplicity Analysis
580 (oxide)	1.59	1.17	1.02	1.05
342 (metal)	1.04	0.70	0.53	1.03
1184 (oxide)	1.34	1.08	1.02	1.00
1366 (metal)	1.00	0.78	0.72	1.00
308 (oxide)	1.68	1.16	0.94	1.07
149 (metal)	1.06	0.68	0.41	0.98
88 (oxide)	1.55	0.98	0.54	1.10
126 (oxide)	1.59	1.04	0.68	0.99
2346 (metal)	1.00	0.89	0.89	0.99
483 (oxide)	1.48	1.05	0.89	0.94
Average	1.33	0.95	0.77	1.02
RSD	0.28	0.18	0.22	0.05

Table 7-4 summarizes the results of the 5 Monte Carlo calculations for each of the different sphere characteristics (Ref. 10). For these inhomogeneous items, the oxide calibration curve modeled with the constant enriched material and the active multiplicity results generally did best. The metal calibration curve leads to large biases for materials that are still relatively dilute. The Monte Carlo study showed how the calibration curve results become biased when the standards do not represent the assay items. The study also showed that the active multiplicity analysis has the potential to overcome some of the limitations of the calibration curve analysis.

Table 7-4. Ratio of the assay mass divided by the true mass used in the Monte Carlo model for inhomogeneous U oxide items with embedded metal spheres.

No. of spheres	Max sphere radius (cm)	^{235}U mass(g)	M	Metal	Oxide (93%)	Oxide (variable)	Active multiplicity
0		1227	1.07	1.25	1.00	0.94	0.96
10	0.5	1248	1.07	1.24	0.99	0.94	0.96
10	1.0	1423	1.09	1.19	0.97	0.93	1.00

20	0.5	1262	1.07	1.24	0.99	0.93	0.96
20	1.0	1505	1.09	1.17	0.96	0.92	1.01
50	0.5	1337	1.08	1.22	0.98	0.93	0.98
50	1	2144	1.14	1.07	0.94	0.95	1.09
Average				1.20	0.98	0.93	0.99
RSD				0.06	0.02	0.01	0.05

7.5.7 Expectations for Other HEU Materials

The measurement results reported in this section show that active multiplicity counting provides a more robust assay than conventional coincidence counting because it provides additional information, the sample self-multiplication. For material types where it is possible to construct a curve of coupling as a function of multiplication and/or mass, active multiplicity analysis can provide useful shipper/receiver or inventory verification measurements without a large number of representative standards, like passive multiplicity counting. For example, active multiplicity will give relatively unbiased results between uranium metal and oxide, even if the counting precision is reduced. Like passive multiplicity counting, it may be appropriate to calculate each item's mass by both conventional and multiplicity analysis, and compare the results. The conventional analysis will have better precision, but if the two approaches differ by much more that the expected scatter, then the active multiplicity results should be less biased.

Future work on active multiplicity counting will include the study of additional geometries and material types and the development of better relationships between coupling, multiplication, and mass. Equally important, the future use of epithermal or fast neutron counters will allow the technique to be extended to the wide variety of low or medium mass uranium scrap that is present in DOE facilities. As new high efficiency, large volume passive neutron counters go into service, it will be possible to add active end plugs that enable verification of excess weapons materials stored in drums. And, future fast multiplicity counters with very low accidental coincidence backgrounds may be able to do both passive and active multiplicity measurements of mixed HEU/Pu oxides in cans, or inventory verification of mixed HEU/Pu items in large drums.

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