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# CALIBRATION OF THE HB-LINE ACTIVE WELL NEUTRON COINCIDENCE COUNTER FOR MEASURE OF LANL 3013 HIGHLY ENRICHED URANIUM PRODUCT SPLITS

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## ABSTRACT

In this paper, the set-up, calibration, and testing of the F-Area Analytical Labs active well neutron coincidence counter (HV-221000-NDA-X-1-DK-AWCC-1) in SRNL are described for use in the Savannah River Site (SRS) trans-uranium metal production facility to enable assay of mixed uranium/plutonium metal product. The instrument was required within a three-month window for availability upon receipt of LANL uranium oxide samples into the SRS facility. Calibration of the instrument in the SRNL nuclear nondestructive assay facility in the range 10 – 400 g HEU is described. We also report qualification and installation of the instrument for assay of the initial suite of product samples.

## INTRODUCTION

In this paper we describe set-up, calibration, and testing of the F-Area Analytical Labs active well neutron coincidence counter (HV-221000-NDA-X-1-DK-AWCC-1)[1] in Savannah River National Laboratory (SRNL) for use in the Savannah River Site trans-uranium metal production facility (HB-Line) to enable assay of mixed uranium/plutonium metal product. The instrument was required within a three-month window for availability upon receipt of Los Alamos National Laboratory (LANL) national defense highly enriched (Category IV HEU) uranium oxide samples into the SRS HB-Line facility.[2] SRNL Analytical Development Section (ADS) field nuclear scientists were tasked with calibration and qualification of the instrument prior to installation in HB-Line.[3]

Instrument calibration was required prior to installation to meet the DOE guidelines for readiness.[4] The receipted HEU canisters are projected to contain U-235 in quantities up to nearly 2 kg. HB-Line processing included splitting the contents into two to four cans with highly enriched uranium (HEU) content of up to 1200 g in order to conform to the range of the intended calibration standards. The guidelines of reference 3 required qualification of the instrument by the Site Nuclear Material Protection, Control, and Accountability and subsequent acceptance by the DOE customer after evaluation versus International Target Values for safeguarding nuclear material.[5]

In the event, the U-Al calibration standards would not be available in a timely manner, and could not be received into SRNL anyway due to Material Control and Accountability (MC&A) constraints. The SRNL ADS was able to resolve the issue by providing a calibration for the instrument using composite working standards available in SRNL in the range 1 – 388 g HEU. This calibration would allow calendar year 2007 verification measurements in HB-Line for product splits in that mass range until the instrument could be subsequently calibrated in HB-Line using the desired standards. The subsequent calibration would allow verification measurements for splits with contents up to 1200 g HEU.

The ADS field nuclear nondestructive assay (NDA) group has a great deal of experience using active well coincidence counters (AWCC) for HEU activation analysis,[6,7,8] and the SRNL nuclear NDA facility is located in a suitably low neutron background area with three distinct MC&A mass control zones (MCZ). The three adjacent MCZ's allowed dispersal of existing material to accommodate receipt of up to 388 g of HEU and of the active well calibration sources so that we were able to perform the experimental work required within criticality and MC&A guidelines. Our AWCC experience has been with the identical LANL INCC 5.02 software planned for the HB-Line active well development and qualification.[9] Therefore our expertise and infrastructure made the ADS nuclear NDA group uniquely capable to perform the calibration work within the Site timeline and criticality guidelines.

International Target values for Category IV HEU material project uncertainties near 6%for active well measurements of uniform HEU metal. However the HB-Line mission would confront samples that are neither uniform HEU metal nor in the mass range near kilogram quantities for which the target values are projected. The U-Al calibration standards used at SRNL are described in the Experimental section of this paper and represent fairly large extremes of non-uniformity. While we recognized that use of such calibration standards does not represent an ideal scenario, it was agreed that the SRNL standards would be suitable to satisfy the time frame requirements of references 3 and 4 to demonstrate the operability of the active well instrument. Qualification and installation could proceed with the plan of subsequent calibration with the more suitable standards.

**EXPERIMENTAL**

The HV-221000-NDA-X-1-DK-AWCC-1 instrument uses two AmLi($\alpha$,n) neutron sources to activate fast fission in the HEU samples placed in the activation cavity. The AWCC instrument is shown in the photograph of Figure 1. It contains an assembly of 48 $^{3}$He detector tubes mounted in two concentric circles in polyethylene moderator blocks. The neutron signals are summed using an AMSR-150 shift register coincidence counting circuit with the detector parameters shown in Table 1. The AWCC has an AC operated chain hoist to lift and expose the activation chamber to insert and remove samples and to insert and remove the activation sources. The cylindrical chamber has dimensions r = 17 cm and h = 42 cm and is almost ideally suited to accommodate a five-gallon pail. The two sources N071 and N072 have Am-241 $\alpha$-activities of $4.4 \times 10^{10}$ dps and yield ($\alpha$,n) neutron rates of approximately 50000 n/sec and are notched for custom fit into the top and bottom activation positions.[10] The sources are inserted and removed with custom wrenches.

The AWCC assay technique uses a source of activation neutrons to induce fission of the sample U-235 and counts the fission neutrons in coincidence mode. The coincidence count rate is then taken to be proportional to the U-235 content as describedby Dewberry et al.[9] The method is commonly accepted at SRS and by the LANL Nuclear Safeguards staff.[11] Data acquisition and analysis are performed using existing internationally-accepted software, INCC 5.02, installed on a stand-alone PC dedicated especially for this instrument.

Table 1. Active well acquisition parameters employed with the AMSR-150 shift register.

| | | |
|---|---|---|
| Predelay: | 4.50 | µsec |
| Gate length: | 64.00 | µsec |
| 2nd gate length: | 64.00 | µsec |
| High voltage: | 1650 | v |
| Die away time: | 50.0000 | µsec |
| Efficiency: | 0.2359 | |
| Multiplicity deadtime: | 2.8000 | nsec |
| Coefficient A deadtime: | 0.6660 | µsec |
| Coefficient B deadtime: | 0.3810 | nsec |
| Coefficient C deadtime: | 0.0000 | |
| Doubles gate fraction: | 0.6660 | |
| Triples gate fraction: | 0.3710 | |
| Normalization constant: | 1.0000 +- | 0.0000 |
| Active singles bkgrnd: | 16353.907 +- | 1.490 cps |
| Active doubles bkgrnd: | -0.649 +- | 1.451 events/secs (eps) |
| Active triples bkgrnd: | 0.212 +- | 1.426 eps |

The dead-time parameters of Table 1 were not measured using two Cf-252 sources as prescribed in the Nuclear Safeguards Technology Training Program[12], but rather represent our best judgment based on experience with four other neutron counting instruments in the NDA facility. At the low coincidence rates expected for HEU samples in the range 10 – 1200 g, the coincidence dead-time corrections are not crucial factors. Likewise the doubles and triples gate fractions were not determined experimentally, but were calculated from the theoretical equations presented in the LANL Training program.[12] These two items are not crucial parameters for activation analysis either, but can become important when assaying mixed U/Pu items using neutron multiplicity counting.

With no sample in place, the activation sources yield an active background of approximately 16354 neutron singles events/sec (eps) and an active coincidence background of (< 1 eps). Thus the instrumental singles efficiency is approximately 16% for the activation sources, with a corrected random coincidence rate of < $1x10^{-5}$. With an N175 activation source[13] placed in the center of the sample cavity, the detection efficiency was measured to be 23.6(3)% as described in detail below. This is in very good agreement with the efficiency of 23% stated in reference 14.

A cadmium absorber between the sample chamber and the $^{3}He$ detectors reduces the thermal neutron component of the activation spectrum and prevents re-entry of thermal neutrons into the activation chamber. The fast fission neutrons that escape the chamber are then *thermalized* by the polyethylene moderators surrounding the two concentric banks of 48 $^{3}He$ detectors. Thus the $^{3}He$ detectors capture the residual thermal component of the fast fission neutrons from activation of the HEU sample inside the chamber. Therefore the characterization, calibration, and verification measurements were all performed in the fast fission mode in this instance. Removing the Cd liner would shift the neutron spectrum toward lower energies, and the resulting data would be acquired in the thermal mode. This option remains available to investigate. Previous HB-Line verification measurements on Desicooler waste were performed in the thermal mode.[7]


**Figure 1. Photo of AWCC installed in HB-Line with chamber open and in the "up" (loading) position.**

**Chamber Characterization Activities**

The activation chamber was characterized for the overall singles counting efficiency and for the sensitivity of all regions of the activation chamber to counting neutrons. This was achieved by using the instrument in the passive mode (the activation sources removed) to obtain twelve counts with an Am(Li) source with a known neutron activity.[13] Source number N175, which produces approximately 50000 n/sec with no doubles events, was placed in the sample chamber, and the number of singles events was recorded. With these data we were able to demonstrate an axial detection efficiency profile in nearly exact agreement with the original characterization of Figure 3 in reference 14.

The acquisition cavity was laid out in cylindrical coordinates (r, h, θ) with outer dimensions $r_{max}$ = 17 cm and with a height $h_0$ = 0 and $h_{max}$ = 42 cm. The first acquisition, labeled *Bottom 1*, was obtained by placing the source on the bottom of the outer perimeter of the cavity. This point was designated as the starting point with arbitrary θ = 0. So our starting point has coordinates ($r_{max}$, 0, 0). From there the source was moved around the outer bottom of the cavity to θ values of π/2, π, and 3π/2.

The next four acquisitions were obtained at r = $r_{max}$, h = 18 cm and at the four values of θ denoted in Table 2. The last four acquisitions were obtained at at r = $r_{max}$, h = 35 cm and at the four values of θ denoted in Table 2. With these twelve acquisitions we obtained good coverage and characterization of the cavity. The singles rates demonstrate there are no deep minima in the detection efficiency and indicate that each of the $^3$He tubes appears to be operating well. Comparing the four characterization measurements at (17, 18, θ) to the one efficiency measurement we performed at approximately (0, 18, θ) – the cavity center – demonstrates a very flat radial profile for the cavity detection efficiency.

The reader should note the large scatter in the doubles rates observed in Table 2. The doubles rates, which should be zero, are dominated by accidentals. This is completely independent of the quality of the standards and is generally true of all active well acquisitions. The overall doubles average from the 13 characterization measurements is very near zero (0.6 ± 4.2), which demonstrates our electronic dead-time corrections and software accidentals corrections are appropriate. However these doubles measurements also demonstrate a one-sigma uncertainty of approximately 4.2 eps, and so the scatter in our doubles calibration measurements can not be far superior to that limit.

**Table 2. Cavity Characterization Measurements.**

| acquisition | (r,h,q) | S (cps) | s (Singles) (cps) | Doubles (events/s) | s (Doubles) (events/s) |
|---|---|---|---|---|---|
| background | N/A | 37.6 | 0.4 | 0.13 | 0.03 |
| Bottom 1 | 17, 0, 0 | 11672 | 5 | 1.5 | 4.7 |
| Bottom 2 | 17, 0, p/2 | 11650 | 3 | -3.29 | 2.65 |
| Bottom 3 | 17, 0, p | 11703 | 5 | 3 | 3.34 |
| Bottom 4 | 17, 0, 3p/2 | 11604 | 3 | -1.75 | 2.28 |
| Middle 5 | 17, 18, p/4 | 12067 | 3 | 7.9 | 2.7 |
| Middle 6 | 17, 18, 3p/4 | 11991 | 3 | 6.3 | 3.5 |
| Middle 7 | 17, 18, 5p/4 | 12042 | 2 | 3.6 | 3.4 |
| Middle 8 | 17, 18, 7p/4 | 11909 | 4 | -0.5 | 3.4 |
| Top 9 | 17, 35, p/8 | 11422 | 3 | 1.7 | 2 |
| Top 10 | 17, 35, 5p/8 | 11356 | 3 | 0.5 | 1.6 |
| Top 11 | 17, 35, 9p/8 | 11407 | 3 | -4.4 | 1.8 |
| Top 12 | 17, 35,13p/8 | 11349 | 3 | -6.8 | 3 |

**Calibration Acquisitions**

The instrument was efficiency calibrated using twelve U-Al standards whose individual HEU content ranged from 1 g – 102 g. Using these standards, multiple groupings were obtained whose HEU content varied over diverse increments between 1 g – 372 g. Total neutron count rates (singles) and coincidence neutron count rates (doubles) were acquired as described in Dewberry, et al. [6,7,8] The calibration data are displayed in Table 3, and the measured doubles rates (D) versus known mass are shown in Figure 2. The plot of Figure 2 displays the 1-σ scatter of the observed D rates. The standards are described in Table 4.

The data of Table 3 are fit with the cubic calibration curve of equation (1), which we constrained to pass through the origin. The least square fitting was performed by Deming Least Squares Curve Fitting for Windows 1.13. In considering the scatter in the observed doubles rates, it is very important to recognize that the standards are not uniform and were used in a composite manner. That is, each calibration measurement was composed of multiple standards.

**Table 3. Calibration Measurements as described in text.**

| Acquisition | HEU mass | Singles (cps) | Doubles (eps) | sigma (eps) | Triples (eps) |
|---|---|---|---|---|---|
| Active BG | 0 | 16256 | -2 | 1.7 | -0.07 |
| U285Al | 285 | 4098 | 25.5 | 3.3 | 3.1 |
| U177Al | 177 | 689 | 7.6 | 2.2 | -1.5 |
| U30Al | 30 | 3689 | 3.3 | 2.1 | 0.6 |
| U78Al | 78 | 3735 | 11 | 2.6 | -1.4 |
| U126Al | 126 | 3954 | 11.3 | 2 | -2.8 |
| U232Al | 232 | 3957 | 14 | 3.1 | 1.3 |
| U127Al | 127 | 3545 | 9.8 | 2.1 | 0.4 |
| U372Al | 372 | 4270 | 34.8 | 2.4 | -0.2 |
| U363Al | 363 | 4242 | 26 | 1.8 | 1.8 |
| U307Al | 307 | 4118 | 18.3 | 2.6 | 1 |
| 33UAl | 33 | 3807 | 5.8 | 2.2 | -0.8 |
| 285UAl | 285 | 4277 | 22.5 | 1.9 | 0.4 |
| 118UAl | 118 | 3871 | 8.9 | 1.9 | 0.8 |
| 308UAl | 308 | 4255 | 15.8 | 2.4 | -0.7 |
| 372UAl | 372 | 4302 | 30.4 | 1.9 | 0.2 |
| Active BG | 0 | 16351 | -2.5 | 1.3 | 0 |

The measured real doubles rates from the calibration acquisitions were fit with the cubic equation

$$\text{Doubles rate (eps)} = a + b(\text{mass}) + c(\text{mass})^2 + d(\text{mass})^3,$$

$$= 0 + 0.137m - 0.000611m^2 + 1.25\times10^{-6}m^3, \quad (1)$$

where m is the declared HEU mass in units of grams. The uncertainty in the observed doubles rates is dominated by the accidental coincidences, which are easily calculated and subtracted from the reals plus accidentals by equation (2).

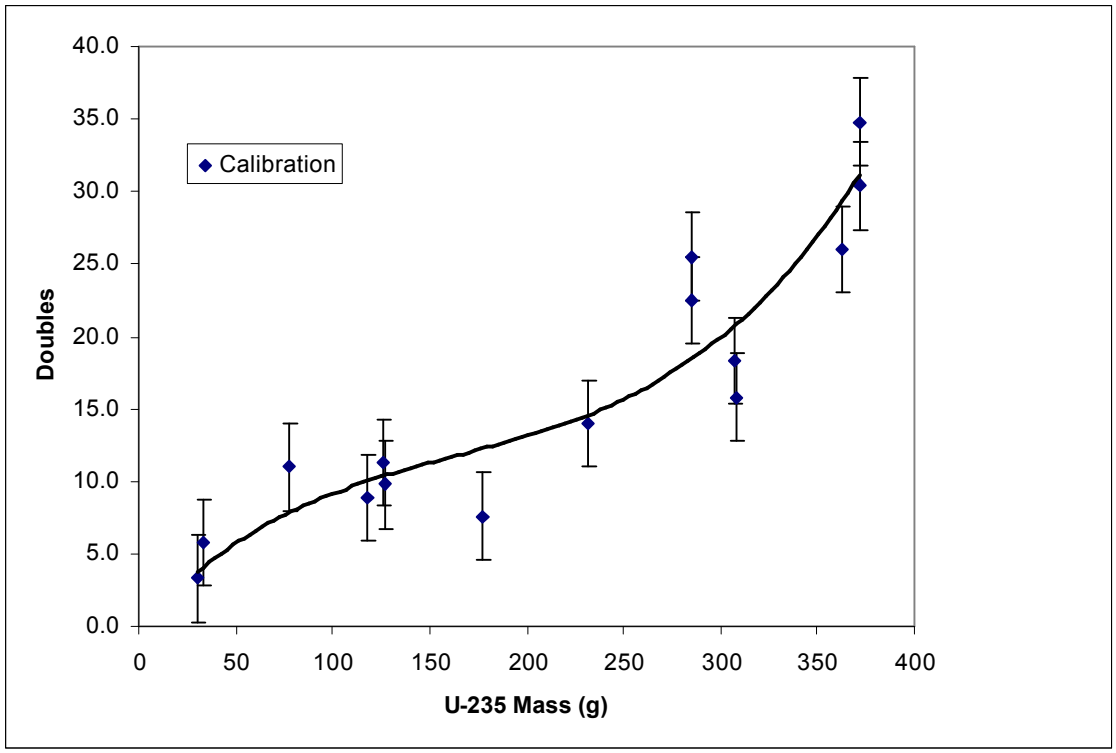


**Figure 3. Doubles calibration versus known mass of HEU.**

$$\text{Accidental D} = S^2 \times \text{gate} = 16000 \times 16000 \times 64 \times 10^{-6}$$

$$= 16000 \text{ events/sec,} \qquad (2)$$

where S is the measured singles rate including the active background. Thus measurement variability is governed primarily by statistical variation in the accidental coincidence rates obtained. Counting times in the range of 1000 seconds to 10000 seconds generally reduce statistical uncertainties down to near 20% for the standards counted. Clearly the low mass standards yield the greatest uncertainty for a given count time, however since the efficiency curve has been constrained to pass through the origin (i.e. zero mass should yield zero coincidence counts) statistical uncertainty from the lowest mass standard is significantly mitigated by the least squares curve fitting. We observed statistical uncertainty of approximately 7% in the doubles rate from the two highest mass calibration standards of 363 and 372 g. We observed statistical uncertainty of approximately 50% in the doubles rate from the two lowest mass calibration standards of 30 and 33 g. Though we tabulate it, we have not used the observed triples rates in the calibration or verification measurements.

**Table 4. Description of the twelve standards used in the calibration and verification measurements.**

| Standard # | U-235 Mass (g) | Total U (g) | Verbal Description |
|---|---|---|---|
| wt2014 | 1.01 | 2 | 2″x8″ planar array of chips |
| wt2016 | 39.11 | 59 | "pillow" of chips |
| wt2017 | 0.99 | 2 | 1″x10″ planar array of chips |
| wt2018 | 5.96 | 9 | "pillow" of chips |
| wt2019 | 15.78 | 24 | "pillow" of chips |
| wt2020 | 0.71 | 1 | 5-inch rods |
| wt2021 | 4.20 | 5 | 3″x8″ slab |
| wt2022 | 9.91 | 12 | 5-inch rods |
| wt2024 | 2.74 | 3 | 5-inch rods |
| wt2026 | 98.31 | 155 | "pillow" of chips |
| fl001 | 98.53 | 170 | 5″x8″ slab |
| 20015420 | 88.00 | 90 | cylinder 1.5-cm base with 10-cm height |

Factors that contribute to the non-linearity of the curve of equation (1) are

- sample orientation and configuration,
- ($\alpha$,n) reactions that increase the neutron singles rate,
- neutron absorption in the sample or container,
- spontaneous fission from U-236 or Pu-240
- neutron multiplication from "chain reaction" type fission.

None of ($\alpha$,n) reactions, spontaneous fission, or neutron multiplication contributes significantly to the non-linearity of (1). These phenomena are not significant for HEU. At the rates we observed, dead-time losses are also insignificant. However it is clear from the verbal description in Table 4 that the standards are not uniform. Thus orientation and configuration non-uniformity can be significant contributors to the calibration non-linearity.

The data of Table 2 demonstrate that there are no large top to bottom variations in neutron detection efficiency. The detection efficiency of the cavity was more thoroughly characterized in reference 14. The rigid nature of many of our standards, and in particular the slab items wt2021 and fl001 made it impossible to approximately center all of the items in the cavity during the calibration activities. While a correction for orientation would have allowed us to more finely tune the calibration curve, such a process would have served to make the curve less general. This might well have disqualified it for the use intended.

There are no known neutron absorbers present in the standards. Since the calibration acquisitions all came from combinations of standards to generate the known U-235 masses, any neutron absorber contribution in the curve should be "washed out" anyway by the composite nature of each pseudo-standard in the acquisition designation. The combinations of standards used for the calibration are shown in Table 5. We were not able to observe a neutron absorber contribution to any single data point in Figure 3. Clearly the possibility still exists. Spontaneous fission Pu contributions and chain reaction type fission are discussed below.

**Table 5. Combinations of Standards used in calibration runs.**

| Acquisition Designation | Standards Used |
|---|---|
| Active BG | None |
| U285Al | All except item 20015420 |
| U177Al | 2014, 2016, 2017, 2019, 2020, 2022, 2026 |
| U30Al | 2017, 2019, 2020, and 2022 |
| U78Al | 2016, 2017, 2018, 2019, 2020, 2022, 2024 |
| U126Al | 2019, 2022, 2024, 2026 |
| U232Al | 2017, 2018, 2019, 2022, 2024, 2026, fl001 |
| U127Al | 2017, 2018, 2019, 2022, fl001 |
| U372Al | All items |
| U363Al | 2016,2017,2018,2019,2020,2022,2026,fl001,20015420 |
| U307Al | 2018, 2020, 2022, 2026, fl001, 20015420 |
| 33Ual | 2014, 2017, 2019, 2022 |
| 285UAl | All except item 20015420 |
| 118UAl | 2019, 2024, fl001 |
| 303UAl | 2019, 2024, 2026, fl001, 20015420 |
| 372UAl | All items |
| Active BG | None |

## Verification Acquisitions

After calibration, we performed 35 verification measurements by re-acquiring the doubles rates using the same group of composite samples treated as unknowns. The INCC software then fits the observed doubles rates onto equation (1) to determine HEU content. The verification measurements are listed in Table 6 and are plotted in Figure 4. The values in the last column of Table 6 list the measured value with 1-σ uncertainty. With the exception of the data taken at mass 88, the verification measurements lie within experimental error of the calibration curve. These data are also reported in reference 11 and are evaluated in reference 15 to declare the instrument qualified for HEU assay.

| Table 6. Verification Measurements. | | | | | | |
|---|---|---|---|---|---|---|
| Acquisition | Known HEU mass (g) | Singles (cps) | Doubles (eps) | sigma (eps) | Triples (eps) | Measure Mass (g) |
| 102U | 98 | 364 | 5.8 | 1.7 | 0.3 | 64+/-19 |
| 102U1 | 98 | 363 | 7.4 | 2.4 | 0.7 | 96+/-31 |
| 102U2 | 98 | 363 | 6.2 | 3.7 | 0.7 | 71+/-42 |
| 102U3 | 98 | 367 | 7.8 | 2.8 | 1 | 107+/-38 |
| 102U4 | 98 | 364 | 8.6 | 2.2 | 2.4 | 129+/-33 |
| 190U | 186 | 399 | 9.5 | 2.9 | 1.1 | 157+/-48 |
| 190U1 | 186 | 399 | 9.5 | 3.4 | 0.2 | 159+/-57 |
| 190U2 | 186 | 396 | 9.1 | 2.7 | 0.7 | 146+/-43 |
| 190U3 | 186 | 399 | 11.6 | 2.7 | -1.1 | 167+/-39 |
| 190U4 | 186 | 382 | 13.2 | 2.5 | 1.5 | 207+/-39 |
| 345U | 307 | 544 | 25 | 2.5 | 1 | 341+/-34 |
| 345U1 | 307 | 535 | 20.4 | 2.8 | 2 | 306+/-42 |
| 345U2 | 307 | 531 | 24.5 | 2 | 4.2 | 337+/-28 |
| 345U3 | 307 | 530 | 23.4 | 3.6 | -1.7 | 330+/-51 |
| 345U4 | 307 | 531 | 22.2 | 1.8 | 3.2 | 321+/-26 |
| 372U | 366 | 573 | 26.7 | 2.7 | -2.3 | 351+/-35 |
| 372U1 | 366 | 576 | 29 | 2.4 | -0.6 | 363+/-30 |
| 372U2 | 366 | 574 | 25.2 | 2.3 | 1.2 | 342+/-31 |
| 372U3 | 366 | 581 | 31.4 | 2.1 | 1.2 | 376+/-25 |
| 39U | 39 | 210 | 4.2 | 2.5 | 0.7 | 36+/-21 |
| 39U1 | 39 | 202 | 5.0 | 2.9 | -0.5 | 45+/-26 |
| 88U | 88 | 35 | 3.1 | 2.9 | 0.5 | 26+/-24 |
| 88U1 | 88 | 36 | 3.7 | 2.7 | 0.7 | 31+/-23 |
| 88U2 | 88 | 40 | 6.8 | 2.6 | -2.5 | 67+/-26 |
| 88U3 | 88 | 37 | 5.3 | 2.9 | 0 | 48+/-26 |
| 88U4 | 88 | 48 | 5.5 | 2.5 | -0.5 | 50+/-23 |
| 289U | 285 | 456 | 14.7 | 2.6 | -1.9 | 237+/-42 |
| 289U2 | 285 | 461 | 14.7 | 2.6 | -0.6 | 237+/-42 |
| 289U1 | 285 | 459 | 15.8 | 2.3 | -0.1 | 254+/-37 |
| 289U3 | 285 | 410 | 19.4 | 1.7 | 2.9 | 297+/-26 |
| 289U4 | 285 | 415 | 15.9 | 2.9 | -1.0 | 255+/-47 |
| 240U | 236 | 592 | 15.7 | 1.9 | 0.2 | 253+/-31 |
| 240U1 | 236 | 585 | 13.8 | 1.4 | -0.6 | 220+/-22 |
| 240U2 | 236 | 587 | 13.7 | 2.0 | 3.3 | 218+/-32 |
| 240U3 | 236 | 481 | 12.0 | 2.1 | -1.1 | 178+/-31 |

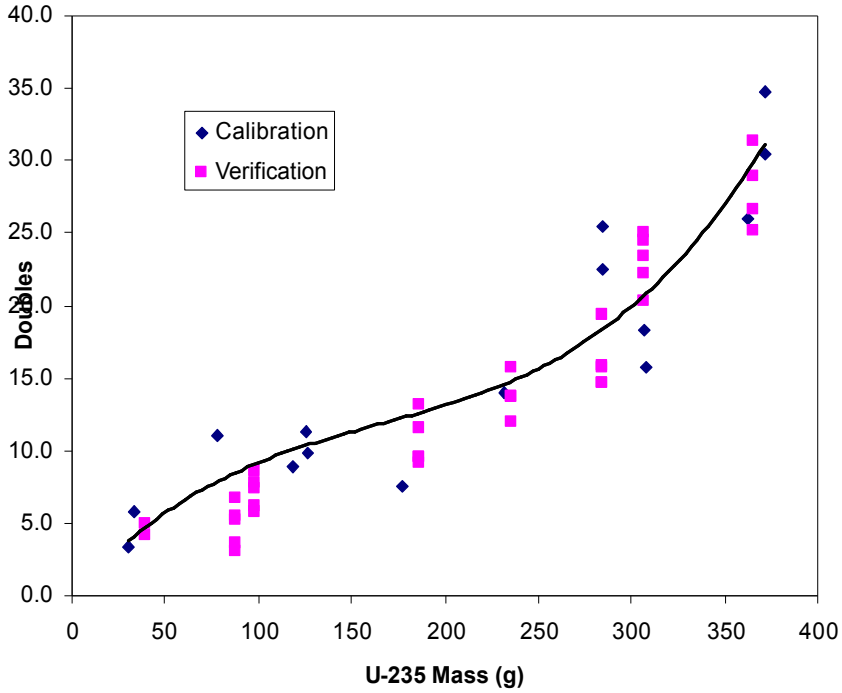


**Figure 4. Verification Measurements. The smooth curve is the calibration curve,the blue diamonds represent the calibration points, and the pink squares represent the verification points. The uncertainty for each point is not indicated on the plot.**

**Pu Spontaneous Fission Contributions**

Spontaneous fission contributions from items of mixed U/Pu content cause significant problems in active well measurements of HEU content. These problems are synergistic and can not be solved with a first order perturbation treatment. In addition to spontaneous fission neutrons predominantly from Pu-240 that occur in coincidence, the Pu-239,241 components are fissile. These two species then contribute strongly by the same (n,f) activation as U-235 to the flux of coincidence neutrons in the cavity. Thus the coincidence rate is enhanced. The multiplicity ν for Pu-239 is slightly larger than for U-235, and the (n,f) cross-section is slightly larger for Pu-239 than for U-235. Thus Pu-239 is more sensitive toward activation than U-235. For samples with sufficient fissile material content, there are second order enhancements from the fission neutrons themselves causing activation. Therefore minor components of weapons grade (WG) Pu can cause significant increases in the rate of detected coincidence events. The authors of reference 7 observed these very important effects assaying several HEU Desicooler waste samples.

We attempted to gain a measure of the positive bias Pu would introduce in mixed U/Pu samples in the acquisitions of Table 7. Acquisitions (UAl99 and UAl99Pu7) and (UAl127 and UAl127Pu) represent two sets where we compare a measurement of standard UAl99 and UAl99 combined with a sample of 7.18 g of WG Pu and a measurement of standard UAl127 and UAl127 combined with 7.18 g of WG Pu. Both comparisons in Table 7 demonstrate that including the 7.18 g of Pu in the activation cavity raises the doubles rate by approximately 7 events/sec.

| Table 7. Mixed U/Pu Acquisitions. | | | | |
|---|---|---|---|---|
| **Acquisition** | **HEU mass** | **Singles (cps)** | **Doubles (eps)** | **sigma (eps)** |
| Active BG | 0 | 12666 | -2 | 1.7 |
| UAl99 | 99 | not noted | 4.3 | 1.5 |
| UAl99Pu | 99+Pu7.2 | 16500 | 11.5 | 2.3 |
| U127Al | 127 | 3545 | 9.8 | 2.1 |
| U127AlPu7 | 127 +Pu7.2 | 3648 | 16.5 | 3.2 |
| 50411 Pu | 0.78 g Pu | 12803 | 1.6 | 0.9 |

These measurements are approximately equivalent to causing a verification measurement of a 99-g item of HEU to result in a measured mass of near 220 g and to causing a verification measurement of a 127-g item of HEU to result in a measured mass of near 250. Because of the curvature of the calibration equation, both biases depend on where on the curve the item occurs. But it is clear from these two simple comparisons that WG Pu yields a greater rate of events/sec per gram than does HEU.

These two simple tests do not adequately demonstrate the synergy of the effect. That is, we have not demonstrated the magnitude of the mutual (n,f) activation "crosstalk" of the U/Pu interaction. Use of the 7.18-g Pu standard is fairly representative of the total Pu contamination we expect in the HB-Line LANL 3013 campaign, and so it might seem reasonable to accept a first order perturbation of approximately 1 eps/g of Pu. However the "crosstalk" effect will become significantly greater when items of near 1 kg of HEU are introduced with Pu contamination into the activation chamber.

The method of active neutron multiplicity assay is under development at SRNL and at LANL.[16,17] This technique of analysis takes advantage of the minor difference in neutron multiplicity $\nu$ between Pu-239 and U-235 fission. Combining a passive measure of a sample's neutron doubles, triples, and multiplicity rate with an active measure in theory allows a second order perturbation treatment to determine both HEU and total Pu content of a sample. Another option would involve a passive neutron multiplicity measure of total Pu content combined with a $\gamma$-PHA measure of Pu-isotopics and U/Pu ratio. We believe the HB-Line facility should collect the important passive/active neutron and $\gamma$-PHA nuclear data from the samples identified as mixed U/Pu to support this kind of international research and development program.

## CONCLUSION

We have calibrated an active well neutron coincidence counter in the SRNL nuclear NDA facility for use in HB-Line for HEU verification measurements. The instrument is calibrated in the range 30 – 400 g of HEU and has been qualified and installed for use in HB-Line for the LANL Category IV HEU 3013 split-and-verify campaign. The measured neutron coincidence activation data were fit with a cubic equation of doubles rate versus known mass to relate measured real coincidence rate to HEU content in unknown samples.

We have thoroughly characterized the activation chamber of the instrument with regard to sensitivity of neutron counting, and we have acquired control chart singles data with the two activation sources to define subsequent quality control measurements for the facility. We have conducted several passive and active measurements of mixed U/Pu samples. The instrument can represent a valuable opportunity for collection of important nuclear data using real samples for development of passive/active second order U/Pu perturbation treatment of active well verification measurements. The facility has a similar opportunity for acquisition of γ-ray isotopics data for subsequent analysis of mixed U/Pu samples as discussed in the text.

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