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Analysis of Historical Delta Values for IAEA/LANL NDA Training Courses

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ABSTRACT

The Los Alamos National Laboratory (LANL) supports the International Atomic Energy Agency (IAEA) by providing training for IAEA inspectors in neutron and gamma-ray Nondestructive Assay (NDA) of nuclear material. Since 1980, all new IAEA inspectors attend this two week course at LANL, gaining hands-on experience in the application of NDA techniques, procedures and analysis to measure plutonium and uranium nuclear material standards with well known pedigrees. As part of the course the inspectors conduct an inventory verification exercise. This exercise provides inspectors the opportunity to test their abilities in performing verification measurements using the various NDA techniques. For an inspector, the verification of an item is nominally based on whether the measured assay value agrees with the declared value to within three times the historical delta value. The historical delta value represents the average difference between measured and declared values from previous measurements taken on similar material with the same measurement technology. If the measurement falls outside a limit of three times the historical delta value, the declaration is not verified. This paper uses measurement data from five years of IAEA courses to calculate a historical delta for five non-destructive assay methods: Gamma-ray Enrichment, Gamma-ray Plutonium Isotopics, Passive Neutron Coincidence Counting, Active Neutron Coincidence Counting and the Neutron Coincidence Collar. These historical deltas provide information as to the precision and accuracy of these measurement techniques under realistic conditions.

1. INTRODUCTION

The International Atomic Energy Agency (IAEA) maintains records of the results of Nondestructive Assay (NDA) measurements performed during Agency inspections around the world. These results are analyzed for each method and material at each facility to determine the level of agreement that is routinely achieved. The average difference between the measured values and the declared values is the *historical delta* or *performance value* for that measurement at that facility [1].

IAEA inspectors must accurately compare their gamma-ray spectroscopy and neutron coincidence counting measurement results with the known historical delta values as part of the process of verifying the facility declarations for the inspected items. If the measurement result agrees with the declared value to within plus or minus three times the historical delta value, the declaration for the item is verified. If the result lies outside this range the item declaration is not verified. The historical delta of a measurement technique should accurately reflect the precision and accuracy that can be achieved with the measurement technique. An historical delta that is too small relative to the actual accuracy and precision of a given measurement technique would result in too many false rejections of the operator declaration simply from statistical variation. This would create unnecessary work for the facility personnel and Agency inspectors. This is not as problematic, however, as a larger historical delta parameter. If an item is falsely rejected due to smaller constraints, more tests can be conducted to remedy the declaration. However, if the historical delta is much larger than the actual accuracy or precision of a technique, the probability that an inspector would verify a declaration that should have been rejected would increase. This would greatly reduce the effectiveness of using that given measurement technique to detect incorrectly declared items.

All IAEA inspectors since 1980 have attended training courses on NDA presented at Los Alamos National Laboratory. In this course, inspectors are taught the science underlying the measurement techniques they learned during the Introductory Course on Agency Safeguards. They also have the opportunity to vary the conditions under which the measurements are performed (i.e., count rate, shielding) to determine the limitations of the standard measurement techniques. The course ends with a verification exercise in which the inspectors apply their skills to decide whether a list of declared items should be considered verified or not.

In previous training courses, the inspectors have verified the NDA measurements using a historical delta value provided by LANL instructors. The value approximated a realistic historical delta taking into consideration similar facility and nuclear material constraints, but was not based on measurement data. Utilizing the previous five years of IAEA inventory verification data, historical delta values have been calculated that reflect the LANL facility conditions and nuclear material measured during the courses. The historical delta values are the standard deviation of the cumulative percent errors for each NDA method. The percent errors were calculated using the following formula:

$$\frac{(\text{Measured} - \text{Declared})}{\text{Declared}}$$

Not all items measured in the course were included in the calculation of our historical delta calculations. There are also items that inspectors may encounter but are significantly different in composition from the pure items. These are Mixed Oxide (MOX) and impure plutonium items. Therefore, separate neutron analysis strata have been included for MOX items and impure plutonium items.

2. RESULTS:

Uranium Isotopics

Uranium Enrichment employs Thallium doped Sodium Iodide (NaI) detectors, Cadmium Zinc Telluride (CZT) detectors, and high purity Germanium (Ge) detectors to measure ^{235}U fraction. The most commonly used measurement technique is called the infinite thickness method, or the enrichment meter technique [2]. The ^{235}U fraction (enrichment) of an item is directly proportional to the intensity of the 185.7-keV gamma-ray peak observed in a spectrum from a collimated gamma detector. The NaI and CZT detectors are calibrated by taking spectra from two or more items of known enrichment.

The peak areas from a region of interest around the 185.7 keV peak (ROI_1) and from a background area at slightly higher energy than 185.7 keV (ROI_2) are used to determine the calibration constants A and B shown in the formula below.

$$E(\% \text{ enrichment}) = A(ROI_1) + B(ROI_2)$$

For Ge detectors, a much more accurate net peak area can be obtained for the 185.7 keV gamma ray. This enables us to use a simpler calibration formula; the percent enrichment is directly proportional to the peak area. The calibration constant, K, is the proportionality ratio.

$$E(\% \text{ enrichment}) = K\,(ROI_{net})$$

Both of these calibration formulas must be corrected for differences between the calibration standards and verification items for parameters such as the container wall thickness and item composition.

The uranium enrichment historical delta value was created from two IAEA courses worth of data. The historical delta value is calculated by the average, absolute value of the percent errors of the measured enrichment compared to the declared enrichment value. Measurements were taken with NaI detectors, CZT detectors, and Ge detectors. Many of the same items were measured with all of these detectors to produce the source data, so the characteristics of the individual sources being measured should not affect the different detector results. The NaI results are shown in Table 1.

Historical Delta Average	3.47%
Historical Delta StDev	5.78%

Table 1. NaI Enrichment Measurement Historical Delta results.

Given this Historical Delta, the verification criteria for these items would be plus or minus three times the historical delta average, or ±10.5%. Note that this percentage is not enrichment, it is the relative agreement between the measured and declared enrichments.

The NaI detectors used in the course are 51 x 12_mm crystals. When comparing the historical delta percentages for the three types of detectors, the NaI detector results are the most accurate and precise. This is most likely due to the high efficiency of the NaI detectors, which gives excellent statistics in a short measurement time (typically less than 300 seconds and often less than 100 seconds). However, because of the poor resolution of NaI detectors, the regions of interest used to analyze the spectrum must be very broad. This increases the risk of other radioactive materials in the item or the environment introducing undetected peaks into the spectrum, which could bias the results. If such materials are expected in the environment, a background spectrum should be taken. If there is reason to suspect the presence of such materials in the items to be analyzed, then higher resolution detectors must be used.

The results for CZT detectors are shown in Table 2.

Historical Delta Average	6.16%
Historical Delta StDev	6.62%

Table 2. CZT Enrichment Measurement Historical Delta results.

The principal reason for the higher historical delta of CZT is the lower efficiency of these detectors. The crystals are 0.5cm^3 in volume. Even with long count times (as much as 1800s are used, usually for natural enrichment items), the CZT detectors do not acquire as many counts as the NaI detectors. The Agency typically uses CZT detectors to measure large items where the count rate is high enough to compensate for the low efficiency of the crystals, i.e., even smaller CZT detectors inserted into the guide tubes of fresh fuel assemblies. For CZT detectors, inspectors have the choice of using the same ROI setup as for NaI, or narrowing the ROIs to avoid interference from other radioactive materials.

Enrichment measurements using CZT detectors show very-low bias when properly performed. However, the precision suffers due to resolution constraints. Although the resolution of the CZT detector is better than the resolution of the NaI detector, there are circumstances in which the Agency still requires the higher resolution provided by Germanium detectors.

The historical delta values calculated for Germanium detectors are found in Table 3.

Historical Delta Average	6.80%
Historical Delta StDev	11.00%

Table 3. Ge Enrichment Measurement Historical Delta results.

Germanium has the best resolution of the gamma spectroscopy systems used by the Agency. The detectors used in the course have a frontal area of 1000mm^2, and are 15mm thick. These detectors produce good statistics with reasonable count times. This enables inspectors to measure the enrichment of items contaminated with other radioactive materials, such as in aged UF6 cylinders. However, Ge detectors are only used when other detectors cannot be used because Ge detectors require liquid nitrogen to operate. The need for liquid nitrogen and the time required for detector cooling present operational challenges for inspectors in the field, but enable inspectors to make measurements that would not be possible with lower-resolution systems.

In addition to infinite thickness measurements, Germanium detectors can also be used to make ratio-based measurements, as implemented in the FRAM [3,4], MGA [5,6], and MGAU [7,8] software packages. Analyses performed with these techniques and codes have many advantages. They do not require calibration, and are self-correcting for variations in sample composition, wall thickness, and detector efficiency. These advantages are clearly shown in the calculated historical delta values in Table 4.

Historical Delta Average	2.02%
Historical Delta StDev	1.84%

Table 4. MGAU Historical Delta results.

MGAU uses the x-ray region of the gamma spectrum to determine the isotopic composition of the item. Instead of directly correlating the 185.7 keV peak intensity to the enrichment, MGAU uses the intensity ratio between nearby peaks from different isotopes. This makes the measurement relatively insensitive to changes in container wall thickness, chemical composition of the item, and detector efficiency. The detector does not have to be calibrated as for the other enrichment measurements. This technique does require isotopic homogeneity, and that the uranium be old enough that the ^{238}U daughter products are in secular equilibrium with ^{238}U (a few months). The robustness of this technique is evident from the Historical Delta value and excellent precision of the results.

Plutonium Isotopics

Measuring **plutonium isotopic composition** requires a high-resolution Germanium (Ge) detector. Although Ge detectors are bulky and must be cooled to liquid nitrogen temperatures (77K), IAEA inspectors use Ge detectors when the best possible resolution is needed. The data reported here are from MGA [5,6] analysis, using the 60-300 keV region of the spectrum. The software uses peak intensity ratios to determine the isotopic composition from the spectrum. Like MGAU, which was developed from MGA, the MGA analysis system does not require calibration, and is self-correcting for source/detector geometry, container wall thickness, and source composition. Three courses worth of data have been analyzed to calculate the historical delta value. The Historical Delta value reported in Table 5 is for the $^{240}Pu_{eff}$ fraction. Historical Delta values could be calculated for any single isotope. The $^{240}Pu_{eff}$ fraction was chosen because that is the value that is used, in combination with Neutron Coincidence or Multiplicity counting results, to calculate the total Pu mass.

Historical Delta Average	3.21%
Historical Delta StDev	4.28%

Table 5. Historical Delta for Plutonium Isotopics results.

Neutron Coincidence Counting

Passive Neutron Coincidence Counting is used to measure radioactive material that undergoes spontaneous fission [9]. The even-numbered nuclides, ^{238}Pu, ^{240}Pu, ^{242}Pu, are all significant sources of neutrons from spontaneous fission. The neutrons from spontaneous fission are strongly correlated in time. By measuring the coincident neutron emission rate, we can determine the mass of fissioning material. Neutron coincidence counting is a very SNM-specific assay technique. There are very few naturally occurring neutron sources that produce time-correlated neutrons. Only fission and cosmic ray interactions in the detector will produce real coincidence events. The typical background rates for neutron coincidence measurements are therefore low. Other than pure plutonium, plutonium can be found in other forms, i.e. impure and Mixed Oxide (MOX). Because these forms have different neutron coincidence rates and are encountered in the field, we have calculated separate Historical Delta values for pure plutonium, impure plutonium, and MOX.

Los Alamos National Laboratory utilizes four High Level Neutron Counters (HLNCs) [10] for IAEA training courses. They are descriptively labeled to identify one from the other: 10 atmosphere (labeled 10 atm in the table), Blue and Tan, Silver, and White. The HLNC detector employs ^{3}He tubes embedded in polyethylene. The ^{3}He tubes are generally filled to a pressure of three atmospheres. One detector has 10-atmosphere tubes, giving it higher efficiency. The polyethylene in the HLNC and most other neutron detectors serves as a moderator which thermalizes the neutrons from a high energy (1-2 MeV) to thermal energy (0.025eV). Helium-3 tubes are much more efficient at thermal energies. The interaction of the thermal neutrons in the tubes yields an electronic pulse that is counted. A shift register counts the electronic pulses and analyzes their time correlation to determine a singles rate and a doubles rate for the source/detector combination. This data is then analyzed by the IAEA Neutron Coincidence Code (INCC) [11] software developed by Los Alamos National Laboratory.

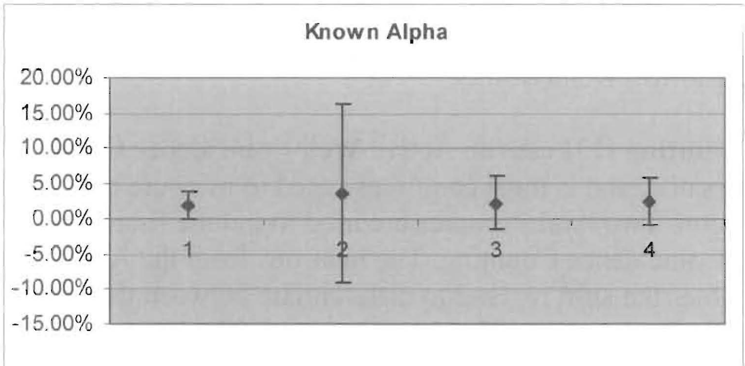
INCC uses two analysis methods: Known Alpha and Passive Calibration Curve. The Known Alpha method is best used for pure materials because it is more sensitive to perturbations such as impurities in the sample and any changes in the singles background count. The Known Alpha method works well where multiplication and density variation are present; while the Passive Calibration Curve method is best for the impure materials and items that are similar in characteristic to the calibration standards. INCC is programmed to output the result from either a primary "normal analysis method" or a secondary "backup method." If the results are within three standard deviations of the combined error, INCC recommends that the normal analysis be used; else INCC recommends the backup method. In these training courses, INCC is set up with the Known Alpha as the normal analysis method, and Passive Calibration Curve as the backup analysis method. Data from seven courses were analyzed to calculate the Historical Delta values for the pure plutonium mass data.

Table 6 shows the historical delta results for each detector, as well as the historical delta value for all Passive Calibration results and Known Alpha results. Individually, the Historical Delta value for the Known Alpha method is significantly more precise than the Passive Calibration Curve Historical Delta value.

Passive Calibration	10 atm	Blue + Tan	Silver	White	Average
Historical delta	8.43%	7.21%	5.73%	5.69%	6.77%
Uncertainty	16.59%	16.27%	5.02%	3.34%	10.31%
Known Alpha					
Historical delta	2.00%	3.63%	2.33%	2.41%	2.59%
Uncertainty	2.03%	12.83%	3.89%	3.36%	5.53%
Combined					
Historical delta	2.71%	7.21%	2.95%	4.35%	4.31%
Uncertainty	4.79%	16.27%	3.51%	5.81%	7.60%

Table 6. Pure Plutonium Mass Historical Delta results.

The results from each detector were analyzed to determine a detector-specific Historical Delta value. These values and their standard deviations are shown in Graph 1. The data show that the different HLNC detectors are all giving comparable results, therefore the results were combined into a single HLNC historical delta value.



Graph 1. Known Alpha Historical Delta values for HLNC detectors.

Impure Plutonium Mass materials must be identified through knowledge of the facility or process, or by operator declaration. As discussed previously, the Passive Calibration Curve analysis typically produces better results for impure items because the impurities increase the neutron production rate by facilitating (α, n) reactions. The Known Alpha method is therefore extremely sensitive to impurities. The large Historical Delta value for impure items, 10.71%, reflects these facts. Although impure items can be measured using a HLNC and Passive Calibration Curve analysis, a multiplicity counter is the best choice for measuring impure items. Multiplicity analysis [12] provides three data points: the singles rate, the doubles rate, and the triples rate. This allows us to calculate the (α, n) rate for the item, enabling more accurate measurements of impure items.

Coincidence measurements are the main focus of the neutron techniques taught during the IAEA NDA Techniques Course. Multiplicity techniques are taught in other courses. The historical delta results for neutron coincidence counting are shown in Table 7.

Passive Calibration	10 atm	Blue + Tan	Silver	White	Average
Historical delta	8.73%	11.96%	11.00%	11.15%	10.71%
Uncertainty	10.40%	13.51%	13.02%	10.99%	11.98%

Table 7. Impure Plutonium Mass Historical Delta results

A **PuO_2 or Mixed Oxide sample (MOX)** should always be measured passively because of the Pu content. For most MOX, which has negligible impurities, the Known Alpha technique should give the best results. The MOX used in these training courses has impurities that affect the neutron measurements, so the Passive Calibration Curve technique gives slightly better results, as shown in Table 8.

Passive Calibration	10 atm	Blue + Tan	Silver	White	Average
Historical delta	12.44%	11.06%	9.98%	9.11%	10.65%
Uncertainty	4.31%	3.58%	4.94%	5.12%	4.49%
Known Alpha					
Historical delta	12.26%	12.00%	12.17%	10.54%	11.74%
Uncertainty	3.96%	2.39%	5.47%	3.98%	3.95%

Table 8. Mixed Oxide (MOX) Historical Delta results.

Active Neutron Coincidence Counting [13] uses an Active Well Coincidence Counter (AWCC). The AWCC is very versatile, and is most commonly used to measure cans of uranium, in an upright configuration. Two AmLi sources are used to induce fission in ^{235}U sources for Active Neutron Coincidence Counting. The neutrons from the AmLi source are uncorrelated. This enables the shift register to differentiate between the AmLi source neutrons and the induced fission neutrons. The detection rate of the induced fission neutrons indicates the fissile mass in the detector. The AWCC detector is larger than the HLNC detectors and uses roughly twice the amount of ^{3}He tubes, to achieve higher efficiency. Thus, the AWCC can assay large quantities of Highly Enriched Uranium (HEU) to a precision of 1-5% in 1000s. Data from seven IAEA courses were used to determine the Historical Delta value shown in Table 9

AWCC	Aquila	Canberra	Average
Historical Delta	10.03%	14.00%	12.02%
Uncertainty	10.26%	10.92%	10.59%

Table 9. Uranium Mass Historical Delta results.

The Uranium Neutron Coincidence Collar (UNCL) [14] measures low- or highly-enriched uranium using four polyethylene sides to thermalize neutrons. Three of these sides contain ^{3}He tubes to detect the neutrons. The fourth side contains an AmLi source. The AmLi source in the HLNC serves the same function as the AmLi sources in the AWCC. Like the HLNC and AWCC, the UNCL also employs the INCC software to analyze the data. The UNCL measures fissile material to an accuracy of 2-4% in 1000s. Data from six IAEA courses were used to calculate the Historical Delta value shown in Table 10.

Historical Delta	3.21%
Uncertainty	3.20%

Table 10. Uranium Neutron Coincidence Collar Historical Delta results.

3. DISCUSSION

These initial values will be refined as additional courses are held in the future. This will add to the realism and training value of these courses for inspectors, and potentially for students of other training courses held at LANL.

The historical delta values reported here are unique. They are based on unusually wide-range calibrations. For example, infinite thickness measurement calibrations often cover a range of 3% to 4% ^{235}U enrichment, while those in these training courses typically cover a range of 60% to 90% ^{235}U enrichment. These are also measurements performed in a training environment, where count times may be shorter or much longer than those typically used in actual safeguards inspections. We have done our best to exclude measurements that were not realistic representations of the inspectors' and measurement systems' capabilities. Some such measurements may have been included, which would artificially increase the reported historical delta values. We expect that a physical inventory-style measurement campaign by experience NDA practitioners would produce lower historical delta values for some or all of these measurement techniques.

4. CONCLUSION

We have done an initial analysis of the historical delta values for most of the measurement techniques taught in the IAEA NDA training courses at Los Alamos National Laboratory. These values will be recalculated after each course, providing an increasing level of realism and training benefit to the inspectors.

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