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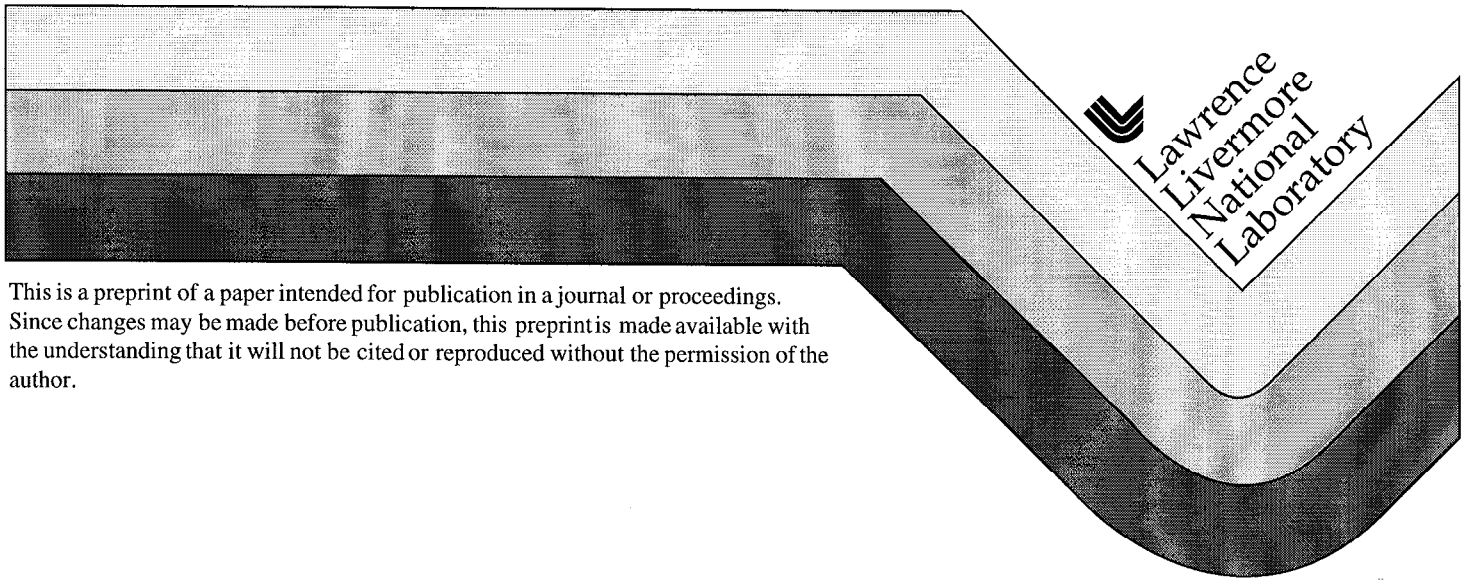

Portable NDA Equipment for Enrichment Measurements for the HEU Transparency Program

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Abstract:

In October 1996, the Department of Energy (DOE) and MINATOM agreed to use portable non-destructive assay (NDA) equipment to measure the ^{235}U enrichment of material subject to the HEU Transparency agreement. A system based on the "enrichment meter" method and high-purity germanium (HPGe) detectors had been previously developed for this application. Instead, sodium iodide (NaI) detectors were chosen to measure ^{235}U enrichment because HPGe systems might reveal sensitive information. Although the accuracy of the NaI systems is less than an HPGe system, it still satisfies the transparency requirements. The equipment consists of a collimated NaI detector, a Canberra Inspector Multi-channel Analyzer, and a laptop computer. The units have been used to confirm the enrichment of material at Russian facilities since January 1997. This paper compares the performance of the NaI systems with the HPGe system and discusses some significant differences.

Introduction:

The HEU Transparency Implementation Project uses portable NDA equipment to measure the ^{235}U enrichment of highly enriched uranium (HEU). Sodium iodide (NaI) detector systems using the "enrichment meter" method have sufficient accuracy, +/- 5%, to confirm that the material has an enrichment of 90%. Procedures for monitoring HEU with the NaI systems were finalized at the Transparency Review Committee meeting in December 1996. Subsequently, two NaI systems were shipped to each of the following plants in Russia:

- Siberian Chemical Enterprise (SChE), Seversk
- Ural Electrochemical Integrated Enterprise (UEIE), Novouralsk
- Electro Chemical Plant (ECP), Zelenogorsk

The systems were first used in January 1997 at the Chemical and Metallurgical Plant of the SChE. At this plant, the equipment measures containers of uranium metal components, metal shavings, and U_3O_8. In the following months, the equipment was used to measure containers of U_3O_8 and UF_6 at UEIE and ECP. The HEU Transparency Project expanded in early 1998 and additional sets of equipment were sent to the Fluorination and Blending facilities at SChE and the Mayak Production Association (MPA) facility in Ozersk. At the SChE Fluorination and Blending facilities, the equipment is used to measure containers of U_3O_8 and UF_6; at MPA, the equipment

measures containers of uranium metal components, metal shavings, and U_3O_8. More information concerning the use of the equipment at the various monitoring points in the Russian facilities can be found elsewhere in these proceedings.[1]

This paper will give a detailed description of the hardware and software used by the portable NDA equipment. A review of the enrichment meter principle will also be provided with a special emphasis on those factors that affect system performance. A more thorough discussion of the enrichment meter method can be found elsewhere[2]. We will also present the results of some laboratory experiments that compare the performance of the NaI-based system to that of the high-purity germanium (HPGe) system previously developed[3].

Review of the enrichment meter principle

In the "enrichment meter" principle, the ^{235}U enrichment is directly proportional to the detected count rate of the 186 keV transition. This method relies on an "absolute" measurement of this one gamma ray and is therefore subject to several sources of systematic error. A ratio method that compares the intensities of gamma rays from the decay of ^{235}U and ^{238}U, as is used by the analysis code MGAU[4], would not be prone to these errors. However, these ratio methods cannot be used for our application, because there are no suitable gamma rays in the decay of ^{238}U. The ratio methods compare gamma-rays from the decay of ^{234m}Pa, a decay product of ^{238}U, with those of ^{235}U. Unfortunately, ^{234m}Pa is the radioactive daughter of ^{234}Th (30 day half-life). Therefore, an equilibrium condition is only achieved between ^{238}U and ^{234m}Pa after several months. In our project, we need to measure the enrichment of newly processed material where the equilibrium condition has not been reached.

In general, the enrichment meter method can be used if the following conditions are met:

- The field of view of the detector is completely filled by the uranium material. This can be achieved for most samples by designing an appropriate collimator.
- The sample is "infinitely thick" for the 186 keV gamma rays. Because of the strong attenuating properties of uranium compounds, this condition can be achieved with rather thin samples. An infinite thickness for uranium metal is only 3 mm. For UF_6, it is 1.5 cm.
- Because of the short range of the 186 keV gamma rays in uranium, this method only measures the outermost layer of the material. Therefore, an accurate measure of the enrichment can only be obtained if the sample is homogeneous.

If the conditions of the method are fulfilled then the enrichment can be determined with the following equation:

$$Enrichment = KM_{CORR}ATT_{CORR}Rate(186),$$

where K is the calibration constant that can be determined by measuring an appropriate standard and M_{CORR} is the material correction factor. M_{CORR} accounts for the differing self attenuation properties of the uranium compounds to be measured. This correction is rather small. For example, if U_3O_8 has $M_{CORR} = 1.0$, then the factor for uranium metal is

.98 and that for UF_6 is 1.03. The correction for the attenuation of the container wall, ATT_{CORR}, can be quite significant. In the equation

$$ATT_{CORR} = e^{-\mu t},$$

where t is the container thickness and μ is the linear attenuation coefficient for 186 keV photons, a steel container wall that is 6 mm thick yields a correction factor of two. This is typical of the containers encountered in the HEU Transparency Implementation Project.

Given the simple equations above, the determination of the enrichment involves the extraction of the 186 peak area from the gamma ray spectrum to determine the rate. For high purity germanium detectors (HPGe) this is a rather straightforward process. The narrow peak shape (FWHM 1-2 keV) allows one to limit the analysis to a small region of the spectrum where a linear approximation to the Compton background is reasonable. Also, the high resolution allows one to determine the background on both the low-energy and high-energy sides of the peak. The low-energy resolution of the NaI detectors makes determination of the peak area more complicated. With the resolution being 15–20 keV FWHM, one must integrate over a much wider area, ~50 keV, where the shape of the background is changing. Furthermore, with this resolution, the 186 keV transition is not resolved from the gamma ray at 163 keV and there is only a small region where one could estimate the background at the low-energy end of the peak. One approach to solving this problem is to redefine the enrichment equation as:

$$Enrichment = M_{CORR} ATT_{CORR} \left[A(Area_1) + B(Area_2) \right],$$

where $Area_1$ is the count rate in an energy window about the 186 keV transition and $Area_2$ is the count rate of a wider window at a higher energy than 186 keV. This technique is incorporated into the IAEA standard method PMCN for uranium enrichment measurements with NaI detectors[5]. In the PMCN standard, the gamma ray spectrum is collected in 512 channels, the 186 keV peak is positioned in channel 300, "$Area_1$" is defined as the count rate in channels 260 to 340, and for $Area_2$ the channels are 350 to 445. The advantage of the two-window method is that the background region ($Area_2$) can be determined with high statistics in a fairly stable region of the spectrum. An obvious disadvantage is that this method requires two standards to determine the calibration coefficients A and B.

Description of the equipment

In the HEU Transparency Implementation Project the portable NDA equipment is used to measure uranium with a nominal enrichment of 90%. Most development projects for enrichment measurements use low-enriched uranium (LEU) since these sources tend to be more readily available. There are, however, some important considerations in making HEU measurements. Although the containers are of smaller volume and thinner than those found for LEU, they are often multi-layered. Furthermore, the outer containers can be rather large to ensure a criticality-safe geometry. This requires a greater degree of collimation to fulfill the requirements of the enrichment meter principle.

Our system is composed of commercially available components. The 186 keV gamma rays are detected by a 1.0-inch diameter by 1.0-inch long NaI detector with a 1.5-inch

diameter photo multiplier tube. This detector is shielded by a collimator whose length is 2 inches and whose aperture is 0.5 inch. This configuration ensures that the samples encountered in our project will completely fill the field of view of the detector.

The detector assembly includes a preamplifier tube base with sufficient gain so that no additional amplification is required. The preamplifier power and the high voltage for the detector are supplied by the Canberra Inspector. The multi channel analyzer (MCA) in the Inspector digitizes the signal and creates a 512-channel spectrum for the analysis. A laptop computer running OS/2™ and the Canberra GENIE-PC™ software controls the measurement and analyzes the data.

The enrichment determinations are performed by a menu-driven program written in the OS/2™ command language REXX™ . The code uses the graphical user interface tools provided by OS/2™ and GENIE-PC™ for input and output. This minimizes the amount of computer (and English) expertise required to operate the equipment. The operator only needs to "double-click" on the code's icon on the OS/2™ desktop to start the code. The program checks the communication link with the Inspector, turns on the high voltage, and creates the spectral display area on the screen. The operator is presented with a menu of options for the sample type that needs to be measured. Menus offer a full user-specified sample description or predefined values. The operator also selects the counting time. For this project, this is normally 60 seconds. The code displays the spectrum as the data are acquired. This allows the operators to verify that the equipment is working properly. When the acquisition time is completed, the operator is asked for additional information about the container. Once this is given, the code extracts the area of the 186 keV line, corrects this for material type and container wall attenuation, and calculates the enrichment. The enrichment is then displayed in a results window so that it can be recorded by Russian technicians and U.S. monitors. After the operator acknowledges the result, the program allows for additional measurements of the same type or returns to the main sample menu. An important feature of this software is that it has been customized for the particular sample types found at each of the Russian plants. This allows for many measurements to be made in a short time with a minimum amount of operator intervention.

System Performance Tests

We have performed many test measurements on laboratory standards of LEU and HEU to further demonstrate the performance of the system. These tests have been selected to contrast the characteristics of the system currently in use in the Russian facilities to a system using HPGe detectors. The measurements address the extrapolation of calibrations based on LEU standards to measurements of HEU. In particular, this issue is studied using the "two-window" analysis technique for the NaI-based system. In a similar manner, we investigate the applicability of the large attenuation corrections with the two different systems.

The test measurements were performed using laboratory uranium enrichment standards. The LEU standards each consist of 200 grams of U_3O_8 in sealed aluminum containers.

The ^{235}U enrichment values for these LEU sources are 0.71%, 1.94%, 2.95%, and 4.46%. The source geometry was designed specifically for these "enrichment meter" measurements. The characteristics of these sources are described in great detail elsewhere[6]. In terms of the measuring geometry, the U_3O_8 is a cylinder with a diameter of 70 mm and a height of 15–20 mm. The detector views the material through a 2-mm-thick aluminum wall. We also used HEU sources with enrichments of 20.06%, 52.56%, and 93.18%. These sources were produced at New Brunswick Laboratory with the same geometry as the LEU sources.

For the high-resolution system, we used a planer HPGe detector. This detector has a diameter of 16 mm and a thickness of 10 mm. The resolution is about 520 eV FWHM at 122 keV. The detector is shielded by a tungsten cup with a 1-cm wall thickness. The collimator in front of the detector is 16-mm thick with an aperture of 10 mm. The detector electronics and data analysis were performed with the same Inspector and laptop computer used with the NaI detector.

The two systems were used to collect data on both the LEU and HEU standards. In addition, some of these measurements were made with stainless steel absorbers for attenuation correction tests.

Results

Figure 1 shows the HPGe gamma ray spectrum in the energy region near the 186 keV gamma ray. One can see that it is a straightforward task to extract net area of this peak. The 186 keV peak is well separated from any other lines in the spectrum and a linear approximation to the background can give reasonable results. In contrast, Figure 2 shows the spectrum for the NaI case. Here, the 186 keV peak is not clearly resolved from the other lines in the decay of ^{235}U and the structure of the background is far more complex.

One important characteristic of the HPGe-based systems is the ability to extrapolate the enrichment calibration. HPGe systems can be calibrated on available LEU standards and then used in the field to measure HEU. The solid triangles in Figure 3 show the measured enrichment versus the declared enrichment for all sources measured with the HPGe system. The calibration was determined using only the LEU data. The extrapolation method works quite well. We also analyzed the NaI data on these sources using the "two-window" method described above. We calibrated using the LEU standards (2-4.5%) and then calculated the enrichment for the HEU standards. The results are plotted as filled circles in Figure 3. A bias can be introduced by this method, probably because the shape of the background above 200 keV is quite different for LEU and HEU. The calibration based on LEU standards with the "two-window" method assumes that the shape is constant.

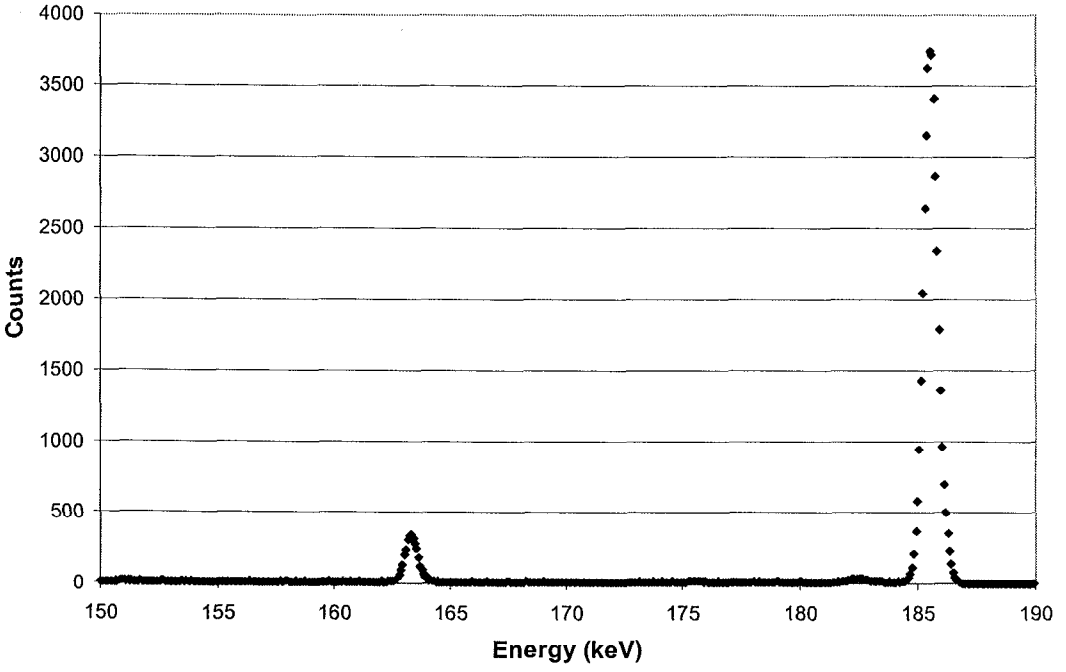

Figure 1: High Purity Germanium Spectrum of Uranium

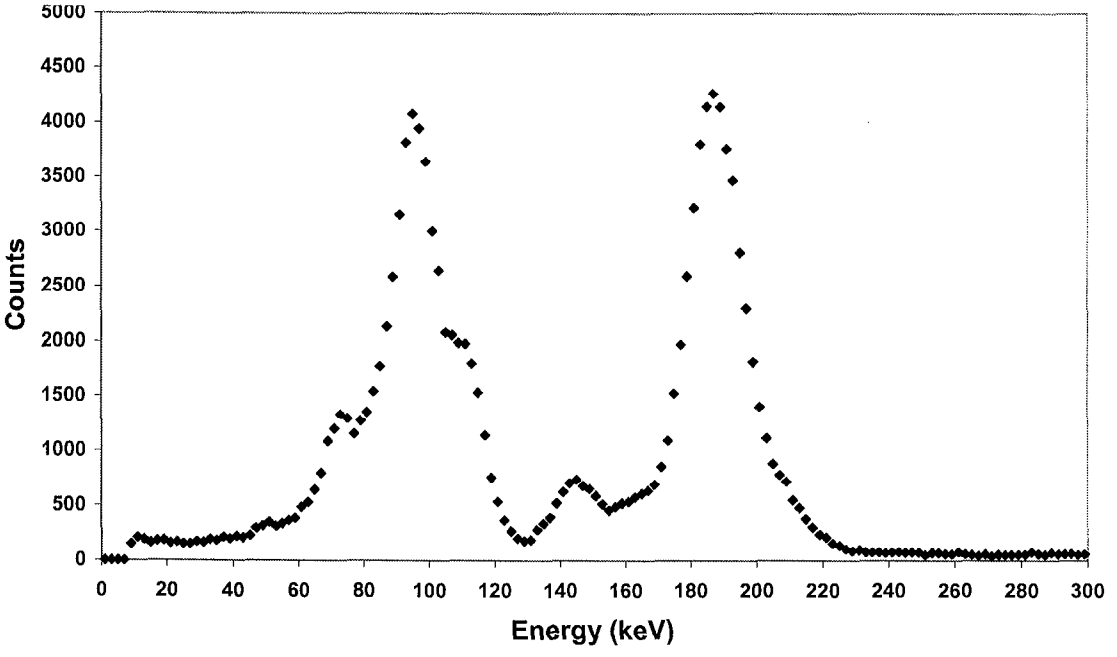

Figure 2: NaI - Gamma Ray Spectrum of 93% Enriched Uranium

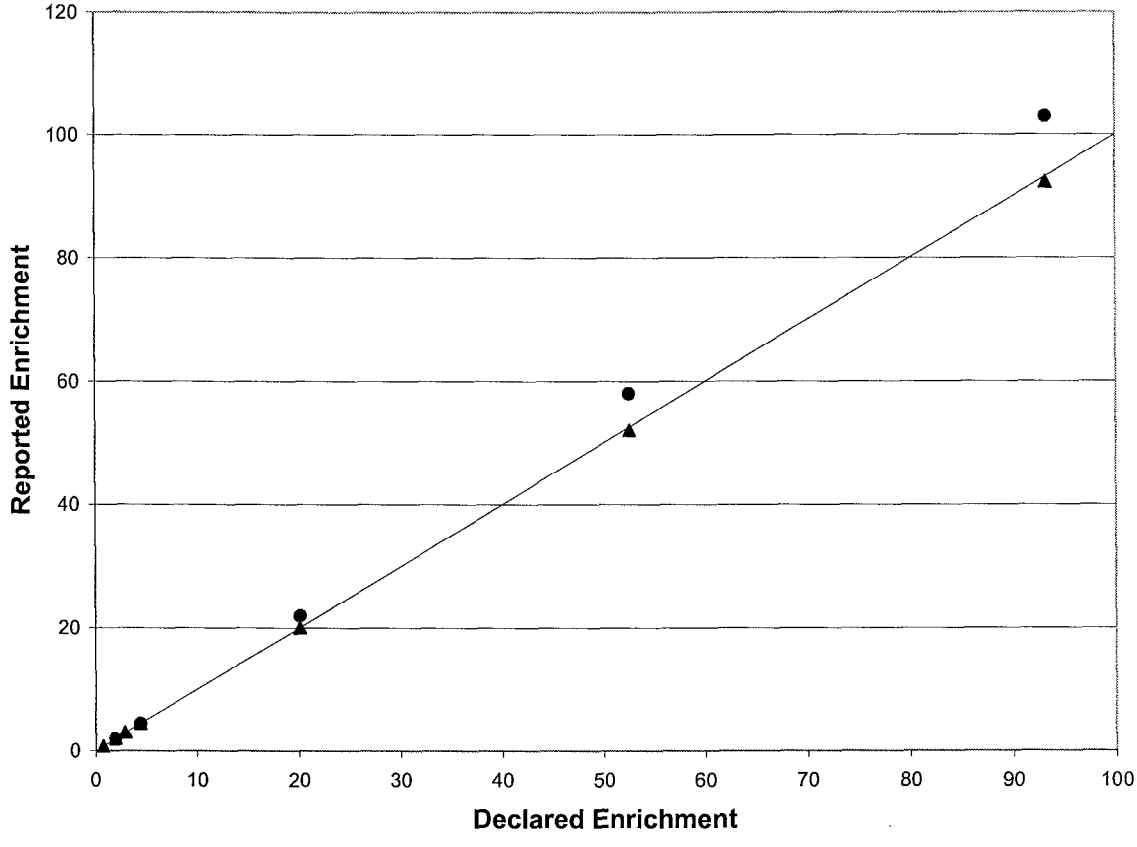

Figure 3: Enrichment calibration data for high purity germanium (triangles) and NaI (circles) enrichment systems. (The statistical error in the counting data is less than 1%.)

The NaI-based system can also be more susceptible to errors with the attenuation correction. Table 1 lists the results of measurements on the 93% HEU source with varying stainless steel attenuation. One can see that the measured attenuation factor is in agreement with the calculated value for the HPGe data. However, the attenuation correction is too large for the NaI "two-window" results. The broad energy window used to measure the 186 keV peak in the NaI analysis includes a significant portion of the gamma rays scattered by the stainless steel. Therefore, by using the standard value of the absorption coefficient (which includes cross sections for scattering over all angles), one overcorrects for the attenuation.

Table 1 : Attenuation corrections for HPGe and NaI detector systems with various amounts of stainless steel. (The statistical error in the counting data is less than 1%.)

Thickness (mm)	Calculated Attenuation	Corrected Enrichment (Ge)	Corrected Enrichment (NaI)
0	1.00	93.30	93.2
1.8	1.23	92.23	99.55
3.6	1.51	92.48	105.64
5.3	1.85	-	110.93

Summary

The HEU Transparency Implementation Project uses portable NDA equipment, described in this paper, to confirm the enrichment of uranium in containers at four Russian plants. In over two years of operation, all results of these measurements have been consistent with the declared enrichment.

We have also presented laboratory test data that shows some of the limitations in using NaI detectors instead of HPGe systems. The low resolution of the NaI detectors limits the range of some aspects of these measurements. However, by calibrating the equipment with standards that resemble the samples to be counted in the field, one can still achieve sufficient accuracy to meet the needs of this program.

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