VALIDATION OF IQ3 MEASUREMENTS FOR HIGH-DENSITY LOW-ENRICHED-URANIUM WASTE DRUMS AT PELINDABA

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ABSTRACT

The U.S. Department of Energy Office of International Safeguards and the Nuclear Energy Corporation of South Africa (NECSA) are collaborating on a measurement campaign to complete an inventory of nuclear materials contained in waste drums at Pelindaba. Containers with reportable quantities are declared to the International Atomic Energy Agency (IAEA). All known containers of high-enriched-uranium waste have been measured and declared. However, some 20,000 drums containing low-enriched-uranium (LEU) waste are currently being evaluated by the IQ3 drum waste assay scanner. The LEU inventory includes a large quantity of material that is composed of a high-density waste matrix that contains high concentrations of uranium. In some cases, the density or uranium quantity contained in the waste drums exceeds the current measurement calibration limits. Calibration of the IQ3 for these high-density, high-uranium-concentration containers is complicated by the fact that suitable standards are not available. Because this instrument is a dual-use system (i.e., both NECSA and IAEA

personnel perform measurements using the IQ3), the calibration standards and techniques must be acceptable to both organizations. This paper provides an overview of the project, including techniques devised to compensate for the lack of suitable standards, as well as reconfiguration of the IQ3 to facilitate measurement of high-density, high-uranium-concentration drums to ensure that an accurate declaration of the LEU inventory can be completed.

Introduction

In 2001 the USDOE Office of International Safeguards (OIS) provided a nondestructive assay (NDA) system to the NECSA under a bilateral agreement between the United States and South Africa. The purpose of the system was to provide assay capability to help to resolve an anomaly in the safeguards inventory that had been declared to the IAEA and to characterize the legacy waste that had been generated by a uranium enrichment program [1].

The system was delivered to the PelStore facility at the Pelindaba Nuclear Institute, the location of the former uranium enrichment program. As a part of the bilateral agreement, OIS provided a comprehensive support program, which included an upgrade to the system (originally built in 1995). Oak Ridge National Laboratory and Canberra Industries provided installation, setup, calibration, training, and development of measurement techniques. The system was an IQ3 drum waste assay system manufactured by Canberra Industries. The IQ3 has the capability of assaying 208-L drums using a six high-purity germanium (HP6e) detector configuration. The three planar detectors in a vertical plane on the rear are used for enrichment analysis, and the remaining three coaxial detectors in a vertical plane on the right-hand side of the counting chamber are used for quantification, with the capability of performing ^{133}Ba transmission-corrected assays. The IQ3 drum-handling fixture was adapted to handle a variety of drum sizes and diameters encountered at PelStore [2].

Prior work has involved developing calibrations and measurement techniques for a large number of container sizes, waste matrices ranging from very low density (< 0.1 g/cm^3) up to moderate densities (about 0.6 g/cm^3). Enrichments range from depleted to 93% enriched uranium, and mass loadings rang from less than 1 g of uranium up to more than kilogram quantities of uranium. The current measurement campaign focuses on high-density, LEU containers. These containers have densities that can exceed 1.1 g/cm^3, with enrichment between depleted and <20%, and may contain kilogram quantities of uranium.

Because this is a safeguards application, the primary criterion of interest is ^{235}U mass. In addition, the system is intended to be a shared-use system between NECSA and the IAEA. Each organization has configured its own computer to control the IQ3 and has collaborated with U.S. experts to develop calibration and measurement methods that are acceptable to both organizations [3].

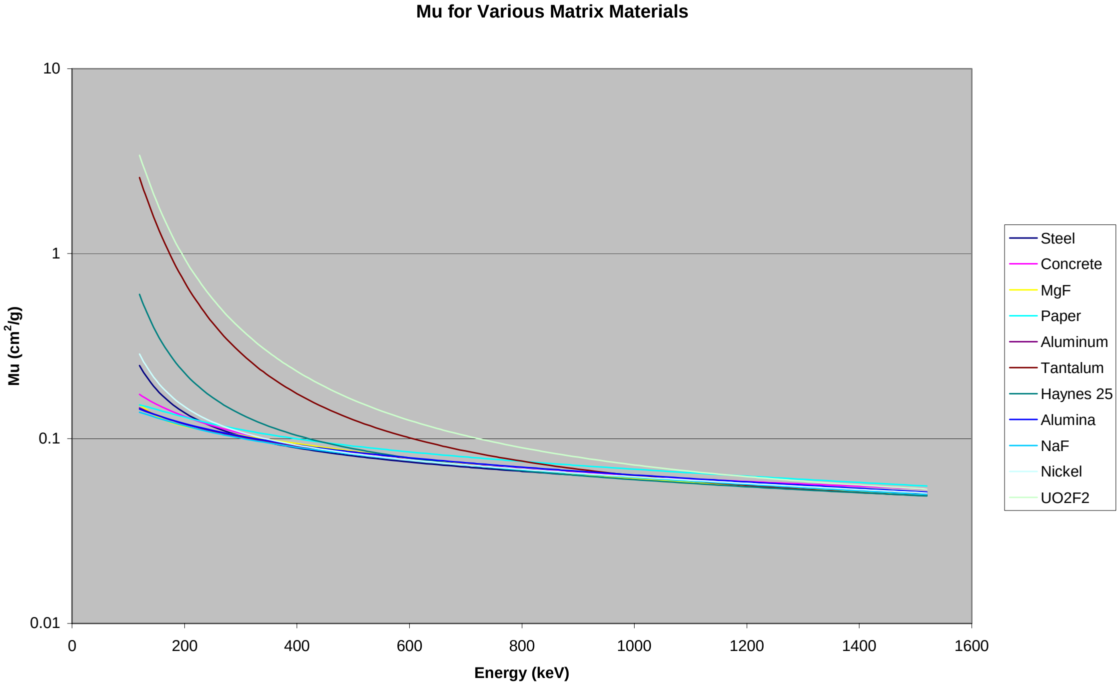
IQ3 Drum Scanner at PelStore 1

PelStore Facility

Description of the Problem

The IQ3 system was designed primarily to measure lower uranium mass values and low- to medium-density waste. The range of matrices includes densities of up to greater than 1.1 g/cm^3 and mass ranges from less than 1 g to over 1000 g of uranium. As a result, for the higher-density matrix, the ^{133}Ba transmission source energies are too low to effectively penetrate the containers. In addition, attenuation correction factors surrounding the 186-keV gamma-ray signal produced by ^{235}U are highly dependent on the composition of the matrix materials and the uranium concentration, neither of which is known at the start of the assay for most of the containers.

The selected solution for this problem is to use a twofold assay. At gamma-ray energies of 1001 keV and higher, attenuation correction is primarily dependent on the mass of material through which the gamma rays must pass, not on the type of material. As is shown in Figure 1, the mass attenuation coefficients (units of cm^2/g) vary greatly for



different materials at low energies (e.g., 186 keV). At high energies, however, the mass attenuation coefficients for most materials are show little variation. As a result, when a high-energy gamma ray is used for assay, the transmission energy and assay energy do not need to be as close to one another as is the case when lower-energy peaks are used. Generalized corrections for the energy difference can be used without regard for the type of material contained.

The first part of the assay, then, is to determine the quantity of ^{238}U using the 1001-keV gamma-ray signal produced by its daughter ^{234m}Pa. In order to obtain transmission-corrected assays at this energy, the ^{133}Ba source cannot be used. The primary energy of this source is 356 keV, and no gamma rays are produced at energies above 400 keV. To resolve this problem, a ^{60}Co source was added to the transmission source holder. The ^{60}Co source provides peaks at 1173 and 1332 keV that can be used for transmission correction. In addition, a mass calibration can be performed that uses the net weight, volume, and fill height of the assayed container to estimate a matrix density. This matrix density can then be used to estimate the attenuation correction factor and derive a second estimate of the mass of ^{238}U for purposes of comparison. This approach can be used only where a sufficient quantity of ^{238}U is present. The lower limit of detectability for the system is substantially larger for ^{238}U than for ^{235}U because of the smaller number of 1001-keV gammas per second per gram of ^{238}U (about 100) relative to the number of 186-keV gammas per second per gram of ^{235}U (about 43,000). For LEU, however, the approach will provide a sufficient lower limit of detectability for safeguards purposes.

Because the primary isotope of interest, however, is ^{235}U, the estimate of the mass of ^{238}U is not sufficient to satisfy the measurement goals. In order to estimate the mass of ^{235}U present in containers, enrichment analysis must also be used. The enrichment is measured by analyzing spectra from three low-energy germanium detectors using the UPu (or MGAU) software package. This software package makes use of the gamma and X-rays in the region of 90 to 116 keV to accurately estimate uranium enrichment. The ^{235}U mass can then be estimated as follows:

$$^{235}\text{U (mass)} = {}^{238}\text{U (mass)} * \text{enrichment} / (1 - \text{enrichment})$$

where enrichment is expressed as a fraction.

One weakness of this approach is that inhomogeneity in enrichment will produce biases. Homogeneity can be checked, however, by comparing the results from each of the three detectors, which are placed at different heights on the container.

Calibration Confirmation

Because this planned operation far exceeds (in both drum density and uranium mass) the tested range of the system to date, plans were developed to make working standards for testing the system performance of the IQ3 system. To do this, it was decided to perform testing with actual contaminated alumina gel (Algel) material. This ensures that all of the measurement characteristics (matrix density, enrichment, and uranium mass) are similar to the measurements planned for the measurement campaign.

Because these drums do not represent calibrated standards, a protocol was developed to homogenize the material; extract small measurement volumes (1-liter bottles), which can be more accurately quantified; perform NDA and enrichment measurements on the small bottles; and perform destructive analysis on a random sampling to validate the NDA and enrichment measurements.

Once these 1-L working standards have been fully evaluated, several configurations can be developed using mixtures of bottles of clean Algel and the 1 –L bottle working standards to develop a variety of test configurations for the 100-, 160-,and 200-liter drums that have been used for storage of the contaminated Algel.

The development of the working standards is currently in process, with the expectation that the confirmation testing of the IQ3 system will occur in the fall of 2005.

References

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