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# Absolute Measurements for Uranium Verification Content in Radiographic Containers

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**Abstract**

Depleted Uranium (DU) is used for its very high density in civilian uses include radiation shielding in medical radiation therapy, industrial radiography equipment, containers used to transport radioactive materials. Absolute measurements have been performed for verification of uranium mass content in gamma radiography by using detector's model developed with MCNP in nuclear safeguard inspection for these samples. Both the experimental results obtained as well as MCNP results are used to estimate the $^{238}$U mass content. The determined and the declared $^{238}$U masses values are found in an agreement with accuracy from -1.74 % to 1.80 %.

***Keywords:*** Radiographic Containers; Uranium Verification.

## 1. Introduction

Radiographic sealed sources are special form, stainless steel capsules containing a high activity. Their gamma emissions are continuous and to be transported and carried the sources need to be housed in special portable containers. These exposure containers (in some countries also called cameras) totally surround the source with shielding such as lead or, more effectively, uranium. Depleted uranium has a number of peaceful applications: counterweights or ballast in aircraft, radiation shields in medical equipment used for radiation therapy and containers for the transport of radioactive materials. The DU is universally defined as uranium (U) in which the percentage weight of the uranium isotope U- 235 is less than 0.711 %. The DU has a higher percentage of the uranium isotope U-238 (around 99.6%) than naturally occurring uranium (NU) (about 99.3% U-238). The DU is produced as a by – product of low enrichment (LEU, i.e. with U-235 enrichment of less than 20%) processing for U-235 enrichment, and out of reprocessing of spent nuclear fuel in the black end of the nuclear fuel cycle, i.e., it is a manmade nuclear (NM) [1-4].

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System for verification of DU used for non- nuclear application has been described by A. A Hamed and his colleagues [5], measurement of DU has been performed by employing non- destructive assay techniques. Results show that the used techniques can detect the presence of DU in the investigation material. Also, quantitive values of U-235 enrichment and mass content of the assayed material could be obtained with a precision of better than 7%. During nuclear safeguards inspection on nuclear facilities or location outside facility, the main target measurements is either to verify the declared nuclear material or to characterize the unknown one. Measurement techniques are categorized into destructive or non-destructive methods, based on whether the measured material is destroyed or kept as it is after assay, so gamma radiography could be measured in non- destructive method. In nuclear safeguards inspection on location outside facility (LOF), gamma radiography which content DU as shielding material from radioactive sources have been measured for verification activities. The detection systems most widely used for gamma spectrometry are NaI(Tl) and HPGe based detectors [6]. One of the most important characteristics of a detector is its efficiency. Important advantage of NaI(Tl) is high detection efficiency and operate at room temperature [7]. The Monte Carlo method has been used to calculate the photon detection efficiency and energy resolution curves for a 1.5" X 1" NaI(Tl) scintillator detector by C.M. Salgado and his colleagues [8]. The detector has been exposed to gamma rays in the energy range from 20 keV to 662 keV. The results showed good agreement with the experimental data. The variation of the intrinsic efficiency of the NaI(Tl) detector against the source-detector distance has been calculated by A. A. Mowlavi and his colleagues [9] for different gamma ray energy by using MCNP code. The absolute efficiency of the gamma detector system will be used at Turkish Accelerator and Radiation Laboratory at Ankara (TARLA) is simulated using MCNPX code [10]. The results have been obtained for NaI(Tl) detector system and compared with the experimental results. A good agreement was found between calculation and experiment. In this work, the gamma radiography has been measured by NaI detector without need to reference material for calibrating the measuring system, MCNP5 simulation has been used for molding the set up configuration to estimate the absolute full energy peak efficiency of measuring system. Combination between the experimental work and the simulation results to determine the mass of DU content in the measured samples which used as shielding material. This work has been performed for verification purposes during nuclear safeguards inspection.

## 2. Measurement Setup

Two common types of Gamma radiography have been measured, the first type is the model 660 series exposure devices are portable industrial radiography exposure devices and are identical expect for capacity and locking mechanisms with dimensions of approximately 47 cm in length, 21 cm in width and 37 cm in height and total weight is 24 kg and DU weight is 16.8 kg. The model 660 series radiographic exposure device is a horizontally oriented "S" tube design exposure device consisting a cast depleted uranium (DU) shield encased in a steel housing. The outer shape illustrate in figure (1) [11].


**Figure 1:** Gamma radiography, the model 660

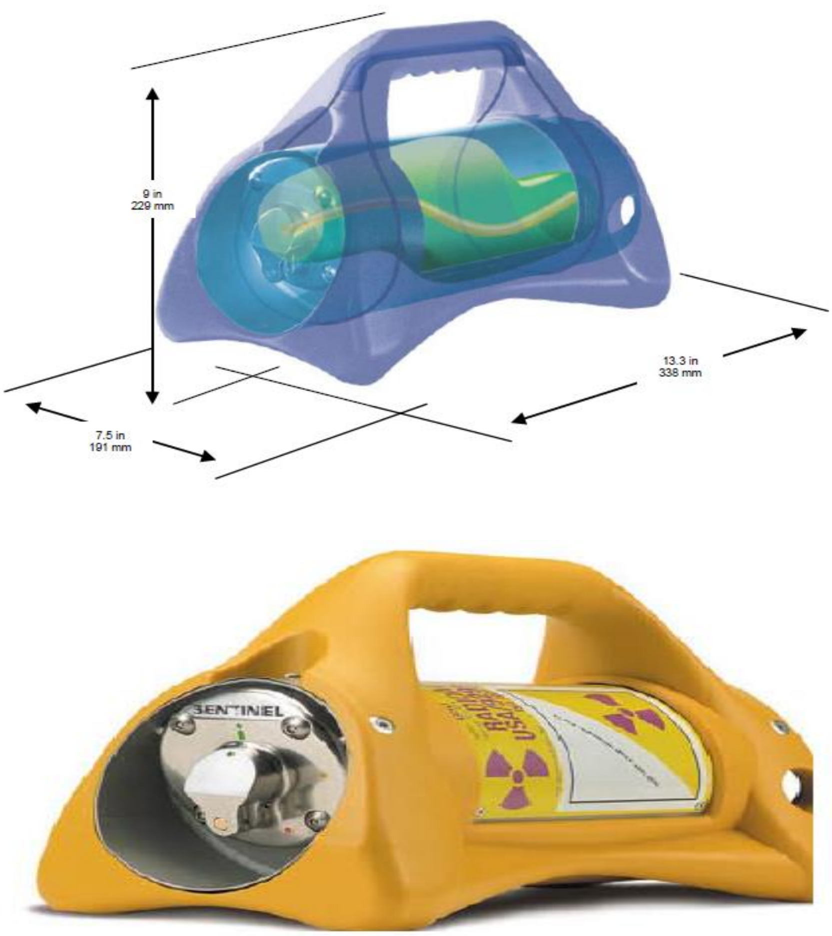


**Figure 2:** Gamma radiography, the model 880

The second type is the Model 880, physically small, lightweight, portable industrial radiographic exposure devices. The ‘S’ tube design exposure device consists of a cast depleted Uranium (DU) shield contained and secured within a 300 series stainless steel tube with stainless steel discs welded at each end to form a cylinder shaped housing. Both discs are recessed into the stainless steel tube to provide protection for the locking mechanism at the rear side and the outlet port at the front side. The outer shape and outer dimensions illustrate in figure (2) with total weight is kg and DU weight is 15 kg [12].

MCNP is a computerized mathematical technique that is useful especially to solve complicated three-dimensional problems. The input file created by the user is subsequently read by MCNP. This file comprises information about the materials specification, the characteristic of geometry, the location and features of the photon, electron or neutron source, the kind of answers or tallies desired and any variance reduction methods used to increase efficiency [13]. In this work, MCNP5 was used to simulate NaI(Tl) detector efficiency. The efficiency was obtained by using F8 tally. F8 is the pulse height tally without any variance reduction. The detector geometry definition about cells and surfaces given in MCNP5 input file. The data obtained from the manufacturer for the detector specifications and DU samples are used for Monte Carlo simulation to design of MCNP input files. MCNP calculations have been performed based on the above mentioned data. The detector model has been applied and checked in absolute measurements for validation purposes. The gamma ray spectrometer used in this work is a portable scintillation model IPROS-3 (serial number 13000115) [14]. The detector has a NaI crystal with dimensions (76.2 x76.2 mm) and an Aluminum housing of 1mm.Each NM sample was measured in such a way that its axis of symmetry is perpendicular to the extended axis of symmetry of the detector, with 65 cm distance separate between the sample and the Al cap of the detector. The setup arrangement for the measured DU samples illustrate in figure (3).

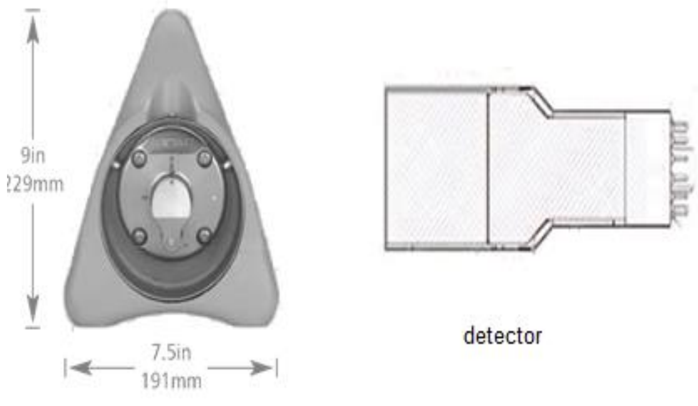


**Figure 3:** the setup arrangement for the measured DU samples

All measurements were carried out by adjusting the experimental setup in such a way that errors due to electronic losses were minimized (dead time did not exceed 2 %). Also, the measuring life time was optimized to achieve good statistics (statistical errors are always kept below 1%). Both the experimental results obtained as well as MCNP results are used to determine the $^{238}$U mass content in the verified samples. Monte Carlo calculations also were performed to estimate the absolute efficiency of the detector at the samples locations with respect to the detector. $^{238}$U mass content in NM samples can be determined by measuring the $C_R$ of the 1001 keV emitted from such NM from experimental work and calculating the absolute efficiency of the detector ($A_t$ , $\Omega_f$ and $\varepsilon_i$) at that energy from MCNP simulation.

## 3. Results and discussion

The basic objective of nuclear safeguards inspection on facilities or location outside facility is characterization and verification for inventory of NM by different method. The passive gamma ray emitted at the most signature of U-238 at 1001.2 keV is measured by a gamma – ray spectrometry system. Table (1) indicates the measured $C_R$ values with associated uncertainties for the measured DU nuclear material samples.

**Table 1:** The measured $C_R$ values with associated uncertainties for the measured DU nuclear material samples.

| Sample ID | $C_R$ | ± $б_{cr}$ |
|---|---|---|
| 1 | 47.54667 | 8.83333E-6 |
| 2 | 45.43833 | 9.25E-6 |
| 3 | 45.14714 | 1.16567E-5 |
| 4 | 47.62667 | 2.5E-5 |
| 5 | 47.59041 | 6.55205E-6 |

MCNP5 input files have to contain detailed characteristics about the detector's as well as the experimental setup configuration. The characteristics of the verified samples and detector obtained from manufacture data were used to design the MCNP input file. The MCNP5 code has been used for modeling the detector response, since it contains a tally, F8, which is specific for detector pulse height determination. The fraction of gamma-ray with certain energy absorbed in the detector active volume represents its absolute full energy peak efficiency at that energy. MCNP5 input files are designed for NM samples at the measuring distance to perform the calculations. The number of histories was selected to keep the relative standard deviation due to MCNP calculations less than 2%. Each run of calculation was performed using $10\times 10^6$ number of histories. Table (2) indicates the calculated absolute full energy peak efficiency values with associated uncertainties for the measured DU samples. The mass of DU contents in the verified NM samples were obtained by substituting the measured count rate and the results of the absolute full energy peak efficiency in equation $\mathbf{C_R = M \cdot S_a . \varepsilon_{ab}}$, where $\mathbf{M}$ is the mass of the assayed isotope in grams, $\mathbf{S_a}$ is the specific activity of the measured gamma - photons with specified energy ($g^{-1}s^{-1}$), and $\boldsymbol{\varepsilon_{ab}}$ is the absolute full energy peak efficiency of the detector at the measured gamma energy. Table (3) indicates the determined and declared values of $^{238}U$ mass content in the measured NM samples.

**Table 2:** The calculated absolute full energy peak efficiency values with associated uncertainties for the measured DU samples.

| Sample ID | **Efficiency (ε)** | ± б $_{\varepsilon}$ |
|---|---|---|
| 1 | 3.195E-5 | 1.26522E-6 |
| 2 | 3.53E-5 | 1.32728E-6 |

| | | |
|---|---|---|
| 3 | 3.52E-5 | 1.32704E-6 |
| 4 | 3.215E-5 | 1.26671E-6 |
| 5 | 3.22E-5 | 1.26868E-6 |

**Table 3:** The determined and declared values of $^{235}U$ mass content in the five NM measured samples.

| Sample ID | Determined $^{238}U$ mass ± $\sigma_M$ | Declared $^{238}U$ mass | Accuracy **%** |
|---|---|---|---|
| 1 | 17.09153±0.00268 | 16.8±0.001 | -1.73532 |
| 2 | 14.78358±0.00231 | 15±0.001 | 1.44283 |
| 3 | 14.73057±0.0023 | | 1.79623 |
| 4 | 17.01379±0.00267 | 16.8±0.001 | -1.27255 |
| 5 | 16.97444±0.00266 | | -1.03833 |

It's clear from table (3) that a good agreement between determined and declared results for $^{235}U$ mass have been obtained with accuracy range -1.74% to 1.80% which indicate to the reliability of the applied method within the study conditions. Comparison between the results of $^{238}U$ mass (kg) obtained from applied absolute method and declared values for the measured DU samples are shown in figure (4).

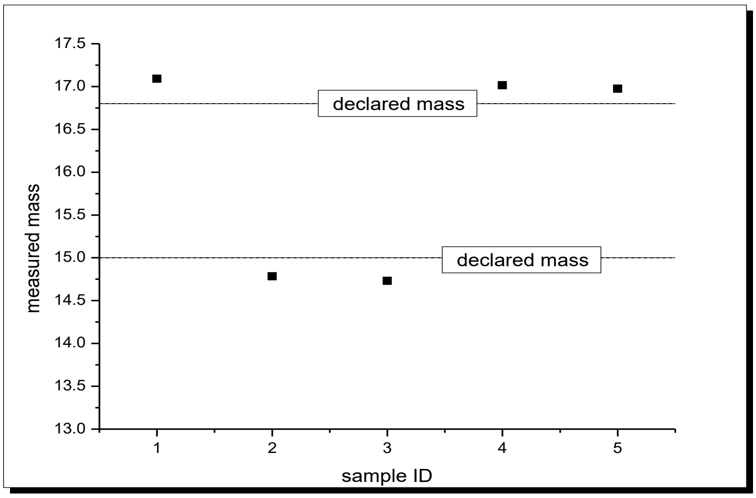


**Figure 4:** comparison between the results of $^{238}U$ mass (kg) obtained from applied absolute method and declared values.

## 4. Conclusion

The applied method is based on companion between experimental measurement of count rates due to nuclear material subject to verification, while the absolute full energy peak efficiency of the detector was estimated using the MCNP code. The samples were assayed without recourse to reference material for calibrating the measuring system. The used technique could minimize labor time for verification of such samples, protects the nuclear safeguards inspector because it minimizes radiation exposure during nuclear safeguards inspections activities. Because of the inspection time is limited, the present study applied on the accounting for and control of NM activities in location outside facility for the verification purposes and saved much time. The present study may be extended to other types of NM and other types of NDA measuring system.

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