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Estimation and Validation of Enrichment Percentage of U3O8 Samples using Innovative Mathematical Formula and Python Code

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Abstract

Depending on the enrichment levels, enriched uranium can be used for many purposes, such as the fabrication of fresh fuels for power and research reactors. Furthermore, under the 1968 Treaty on the Non-Proliferation (NPT), the states, specifically non-nuclear-weapon states of comprehensive safeguards agreements, should be inspected and verified to prove "peaceful use" commitments. Consequently, verifying the enrichment process and the percentage is considered an essential element for investigation purposes. The primary motivation of this paper is to estimate and validate U_3O_8 samples enrichment percentage using innovative formula. The enrichment calculations were conducted using certified safeguarded nuclear materials with varying low enriched uranium (LEU) ratios, both experimentally with a HpGe detector and computationally with Python 3.10 code. The results prove precise and acceptable values compared to the certified declared enrichment percentage (from depleted 0.32% - 4.5%) with differences in the range of 1~ 3.8%. The new formula used in this work applies to national, regional, and international safeguards inspection targets.

Keywords: Nuclear safeguards, Enrichment formula, Nuclear Materials, Python code.

Introduction

Determining the fractional abundance of a specific uranium isotope by radiation measurement is a critical goal. This method is often used in uranium samples to obtain the fraction of fissile ^{235}U, commonly called the uranium enrichment. The term "enrichment" is used because the fraction of the ^{235}U in the sample is usually higher than that in naturally occurring uranium [1]. As a consequence, enriched uranium is a type of uranium in which the isotope separation of uranium-235 (written ^{235}U) has been increased. Naturally occurring uranium is composed of three major isotopes with their isotopic atom abundances as follows: uranium-234 (^{234}U, 0.0050–0.0059%), uranium-235 (^{235}U, 0.7198–0.7202%), and uranium-238 (^{238}U, 99.2739–99.2752%) [2,3]. These isotopes (U-234, U-235, and U-238) are described as follows: The first is a highly radioactive trace component found in natural uranium, but it is not useful in any applications. The second isotope is the only fissile material that occurs in nature in significant quantities. The third is the most abundant isotope (99.284% of the weight of a sample of natural Uranium is U-238), but it is not fissile. U-238 can be split by high-energy neutrons, releasing large amounts of energy, and is often used to enhance the explosive power of thermonuclear or hydrogen weapons [4]. Enriched uranium is a critical component for civil nuclear power generation and military nuclear applications. The nuclear fuel of CANDU reactors is natural uranium, while in materials testing reactors (MTR) is highly enriched uranium (20%-90%). As the percentage of ^{235}U increases, the reactor size for a given power level can decrease, and the power density in the core of the reactor increases. Ship propulsion reactors use enrichments of at least 10% to keep the size of the power plant relatively small. Nuclear submarines, satellites, and many small research reactors use fuel enriched to 90% or more. Because of the high-power densities achievable with such enrichments, research reactors generally require only a few kilograms of fuel to achieve criticality (i.e., a self-sustained nuclear reaction). The International Atomic Energy Agency (IAEA) attempts to monitor and control enriched uranium supplies and processes to ensure nuclear power generation safety and curb nuclear weapons proliferation [3-6]. Determining uranium enrichment in samples is essential for product control in enrichment and fuel fabrication plants, in addition to international safeguards inspections to ensure the used uranium stock for peaceful purposes (Hastings, 1991). Recently, the uranium enrichment of the environmental sample was analyzed to verify the declared information for nuclear safeguards. High-resolution gamma spectrometry (HRGS) and monochromatic micro-X-ray fluorescence (MMXRF) can analyze the sample with a short detection time and high reproducibility [7]. The types of uranium used to feed a uranium enrichment cascade include natural, recycled, depleted, natural-equivalent, and enriched uranium. Recycled uranium, typically derived from reprocessed spent fuel, also includes the isotopes ^{232}U, ^{233}U, and ^{236}U [8-11]. The enrichment is determined by applying the "enrichment meter principle" based on the full-energy peak produced by 186 keV gamma-ray. The "peak ratio" technique can be applied to analyze high-resolution spectra in codes such as FRAM or MGA [12-18].

Computational Process by Python Code

The choice of Python reflects the combined goals of implementing a Python-style interface for ease of use while maintaining the computational performance of the lower-level FORTRAN language [19,20]. Higher-level languages, such as Python (for simple interfaces or interpretive execution) or C++ (for codes with a complex, modular structure), have significant advantages, especially on faster and modern computers. However, in the case of MagBoltz, the computations for gas mixtures of interest remain intensive, sometimes requiring several hours to scan the parameter points of interest on one CPU.

Python library enables users to rapidly create, manipulate, display, debug, read, and write Geometry Description Markup Language (GDML)-based geometry used in MCRT simulations. First, a Python application programming interface is created to describe and manipulate Geant4 and FLUKA geometries, with full support for directly reading and writing their respective geometry description file formats. Then, triangular meshes are formed to represent geometric objects to visualize the geometry and enable the use of advanced mesh-based geometric algorithms [21]. It allows for a quick and simple generation of predefined problems for non-experienced users and highly customized problems for experienced users. Furthermore, it easily integrates using an arbitrary optimization method [22]. The user code can be expressed as a flow chart to show the basic elements for creating python code, as revealed in Fig. 1.

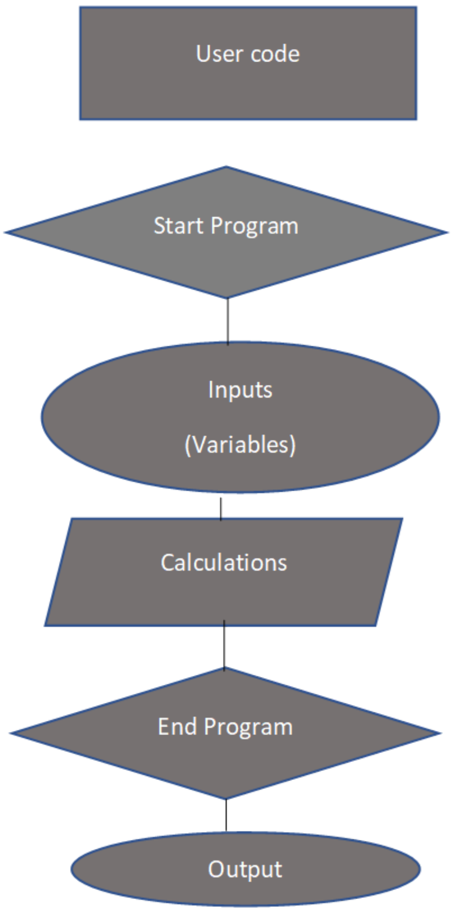

Fig. 1 Basic structure of user code in Python program

Experimental Work and Methodology

Nuclear safeguards refer to all measures established to enable the timely detection and prevent the diversion of nuclear material from peaceful uses [23]. Signing the Non-Proliferation Treaty (NPT) and establishing an international safeguards system led many to believe that this problem could be controlled. In addition, it was recognized that many features made new enrichment techniques like the gas centrifuge and the jet nozzle processes commercially attractive. However, concern about the problem was mitigated by optimism that adequate safeguards arrangements could be devised [24].

The presented work is based on experimental measurements of certified nuclear materials with the chemical form U3O8 by determining the count rates at photo peaks of 185.7 and 1001.2 keV for U-235 and U-238, respectively. Gamma-ray measurements with HPGe detectors at high count rates are required. These can be found mainly in safeguards and neutron activation analysis. HPGe detector with Canberra GL0515R model was used for the experimental work [25-27]. To conduct this work, we used certified declared safeguarded nuclear materials with cylindrical shapes containing compact powder of uranium oxide, with a total mass of 200.1 g and a total uranium mass of 169.681 g (Table 1). At the time of measurement, these materials were in possession of the Egyptian Nuclear and Radiological Regulatory Authority (ENRRA). The calculation of enrichment values, expressed as the abundance of U^{235}/U, is based on an innovative formula, as shown in Eq. (1).

Table 1. Data of the certified nuclear material samples (NBS-SRM-969) [28].

Sample ID	U_3O_8 Weight (g)	^{235}U/U Abundances	^{238}U /U Abundances
SRM969-031	200.1±0.2	0.3206±0.0002	99.6627±0.0004
SRM969-071	200.1±0.2	0.7209±0.0005	99.2738±0.0004
SRM969-194	200.1±0.2	1.9664±0.0014	98.0159±0.0018
SRM969-295	200.1±0.2	2.9857±0.0021	96.9826±0.0029
SRM969-446	200.1±0.2	4.5168±0.0032	95.4398±0.0032

$$\%E = \left\{\left(\frac{e\pi A_5}{12.32\lambda_5}\right) \Big/ \left[A_5\left(T_{\frac{1}{2}}\right)_5 + A_8\left(T_{\frac{1}{2}}\right)_8\right]\right\} \times 100 \quad (1)$$

Where λ is the decay constant, T1/2(5) and T1/2(8) represent the half-life time of 235U and 238U, respectively. Sayed's enrichment formula gives a correlation of eπ term with %E and 235U, 238U activities (A5 m A8), the half-life time, and the decay constant of 235U.

$$A = N\lambda = \frac{Ne\pi}{12.32T_{\frac{1}{2}}} \quad , \quad A = \frac{Cr}{\zeta\gamma} \quad (2)$$

The %E was calculated based on the innovative correlation between eπ (where e equals 2.718 and π is 3.141), A, and T1/2 as given by Sayed's theorem and its application [29-31].

Results

This study focused on uranium enrichment evaluation using gamma-ray emission counts of 235U isotope as detected by HPGe spectrometer. The count rate of signature peaks of U-235 and U-238 was measured for each certified nuclear material. The results obtained are shown in Table 2.

Table 2. The obtained results of enrichment using set of certified nuclear materials (U3O8).

Sample ID	Enrichment (ζ)±6			Accuracy%	
	Certified (a)	Experimental results (b)	Calculated by python (c)	[(a-b)/b]*100	[(a-c)/c]*100
SRM969-03	0.3206±0.0002	0.3088±0.0034	0.3100±0.0021	3.821	3.419

1					
SRM969-071	0.7209±0.0005	0.7034±0.0041	0.7085±0.0023	2.487	1.750
SRM969-194	1.9664±0.0014	1.9468±0.0036	1.9322±0.0028	1.007	1.770
SRM969-295	2.9857±0.0021	2.9441±0.0054	2.9401±0.0032	1.413	2.895
SRM969-446	4.5168±0.0032	4.4530±0.0035	4.4305±0.0029	1.432	1.2621

The similarity of the calculated enrichment percentage (%E) with the declared certified values can be observed in this table. The highest difference, ~3.8%, mainly belongs to the depleted uranium samples. This may be explained by low U235 activity and short counting time. Fig. 2 compares enrichment values, certified data, and experimental and calculated results.

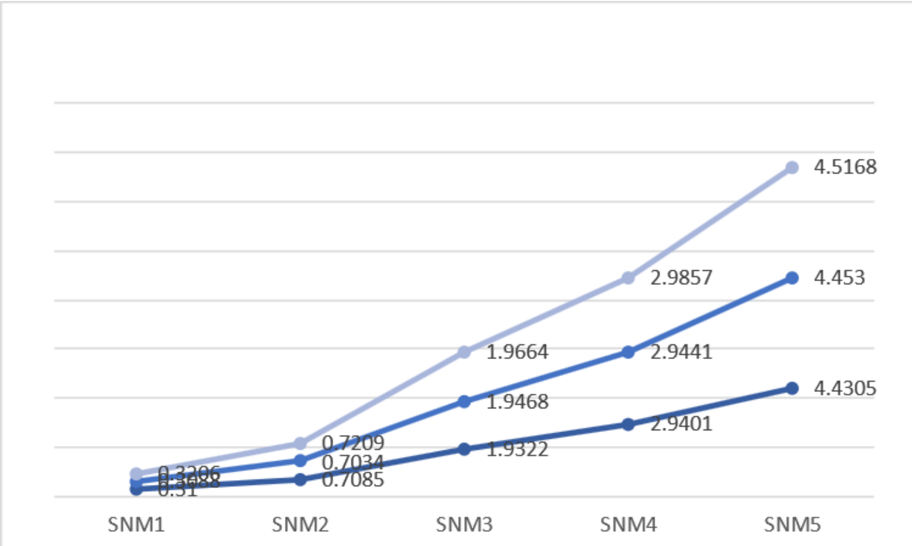

Fig. 2 Uranium enrichment percentage (certified, experimental and calculated) values

Conclusion

The enrichment percentage calculations were performed using the deduced Sayed's enrichment formula. Two approaches estimated the enrichment values (%E). The first one depends on the experimental results using the count rates of specified photo peaks of U-235 and U-238. The second depends on the computational method using python code 3.10. The experimental and calculated results were compared, and the accuracy ranged from 1% to 3.8%.

The slightly high difference belongs to the low activity of the depleted uranium sample. The eπ formula denoted excellent uranium enrichment calculations for different enrichment ranges (0.32% – 4.5%).

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