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Countering distortion scenarios outcome from shaping and the chemical mixing in U₃O₈-Al compact using reliable model

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ABSTRACT
Overcoming the distortion appears in the production of fuel plate in Material Testing Reactor type (MTR) type fuel is considered an important challenge in safeguard measurements. A methodology is presented using a mathematical model and MCNP code to minimize defects that result from the chemical mixing in U₃O₈-Al Compact. The mathematical model relates between distance (sample – detector) and the count rate of 185.7 keV gamma energy line that characterize U-235 isotope. The tested samples are compact of uranium oxide and aluminum alloy. The mixing ratio is 48 volume % of U₃O₈ and 52 volume % of Al-metal powder. The sample volume is 35.48 cm³ and its density is 4.8 g/cm³. The manufacturer data concerning U-235 mass is 0.7785 and 21.3 g. The compact has a parallelepiped shape that has dimensions of 69, 8.5 and 60.5 mm. The used instrument is a High-Purity Germanium detector (HPGe) and the distances between the detector-sample are in the range from 9.5 to 11 cm. Results coming from the model and MCNP-5 complement each other and validated by comparing with declared values. The method could play an important role in safeguards and processes of Quality Control (QC) in the Fuel Manufacturing Pilot Plant (FMPP).

KEYWORDS
U₃O₈-Al; ²³⁵U concentration; distance; count Rate; mcnp-5

1. Introduction
FMPP is a facility for fuel assemblies manufacturing that needed for operating the Second Research Reactor of Egypt, ETRR-2. The manufacturer line utilizes uranium hexafluoride (UF₆; 19.7 ± 0.2% U-235). UF₆ is processed using other raw materials such as pure powder of aluminum, and nuclear grade 6061 aluminum alloy in sheets, bars, and rods through an array of manufacturing stages, inspection, and QC to produce the required fuel assemblies. The MTR type composed of fuel plates arranged in a fuel element. A distributed fuel plate in MTR is dispersed within a powder of aluminum matrix and the produced alloy is framed with aluminum clad. There is a collection of various types of powdered fuel such as an alloy of U-Al, uranium oxide, and uranium silicide (Zidan, 2000; Mansour & Elseaidy, 2016; Elseaidy & Ghoneim, 2010).

1.1. Fuel manufacturing procedure
The procedure of fuel manufacturing U₃O₈-Al Compact which considered as the initial step for producing fuel plates. It includes sequential steps that consist of wet and dry treatment, mixing and compacting finally, framing and welding.

1.1.1. Wet handling
At room temperature, UF₆ exists in the solid form when heated to high degrees of temperature its vapor pressure increases. UF₆ is added to a certain quantity of distilled water hence a uranyl fluoride solution is produced. The solution of uranyl fluoride is then precipitated by ammonia solution, 25-wt %. Ethanol and 1 wt % ammonia solution are added for washing and filtration purposes to get clean and dry Ammonium Diurinate (ADU).

1.1.2. Dry process
The ADU produced in the previous step is calcinated at 800°C to obtain a powder of U₃O₈ which is milled and sieved. Thermal treatment is applied at 1400°C to get a high-density powder which is milled and sieved again to get the required particle size.

1.1.3. Mixture and compact production
A mixture of 48 volume % U₃O₈ and 52-volume % Al-metal powders is homogenized through rotation at 16 rpm for about 3.5 h. After mixing, the application of appropriate pressure is used to obtain the fuel compact with the desired characteristics.

1.1.4. The process of framing and welding
The produced compact is placed in a frame of aluminum alloy of 6061type then two faces of the same
material sandwich the frame and the compact inside. Welding occurs using the TIG method from all sides, except a small distance from the two sides in the rolling direction, to permit gas evaporation in the rolling step (Zidan, 2000; Mansour & Elseaidy, 2016; Elseaidy & Ghoneim, 2010).

1.2. Gamma calculations

Gamma-ray spectrometers are systems that have wide uses in various experimental applications. These detectors are utilized for the assay purposes of radionuclides that emit gamma rays and in measuring radioactivity at nuclear facilities (El-Gammal, El-Nagdy, Rizk, Shawky, & Samei, 2005; Vidmar et al., 2008). Amongst gamma-ray detectors is a HPGe detector that has many applications in the radioactive and nuclear field. The wide uses of this detector are because of its good energy resolution (Ewa, Bodizs, Czifrus, & Molnar, 2001). Uranium is an important element in the nuclear fuel cycle. It is a target of nuclear safeguard inspectors during inspections, characterization, and verification of nuclear materials within nuclear facilities. Uranium is a gamma emitter which has three isotopes with the mass numbers (234, 235, and 238). HPGe detector can be used to measure uranium for nuclear safeguards applications during this measurement Monte Carlo methods are used to complete calculations. The Monte Carlo (MC) method is used to determine the full-energy peak efficiency of high-purity germanium (HPGe) (El-Gammal, 2007; Rodenas, Gallardo, Ballester, Primault, & Ortiz, 2007). From the literature review, it is clear that the efficiency of the detector is one of the most important parameters for MC calculations. In the present work, a model is optimized to predict the count rate of a compact measured at different distance values. The model is of great importance for safeguard inspectors, operators, and QC responsible.

2. Methodology

The equation utilized to calculate the mass of U-235 in the U₃O₈-Al Compact is presented in Equation (1). The measured count rates of gamma energy line 185.7 keV, the calculated absolute full-energy peak efficiency by using MCNP-5 modeling at the same energy and the specific activities of the measured gamma energy line were substituted in Equation (1).

\[ M_{\text{U-235}} = \frac{CR}{(S_a \cdot \varepsilon_{ab})} \]  

where, 
- \( M_{\text{U-235}} \) is the U-235 mass (g), 
- \( S_a \) is the specific activity at single gamma line (g⁻¹s⁻¹), 
- \( \varepsilon_{ab} \) is the detector absolute full-energy peak efficiency at 185.7 keV (Shaban, Hazzaa, & El-Tayebany, 2019).

3. Experimental setup and techniques

3.1. Specifications of U₃O₈-Al compact and instrumentation

Al-powder is experimentally considered the most essential material that enters in manufacturing the compact powder. Its purity should not be less than 99.5%. The impurities that exist in higher quantities in Al-powder are cadmium, boron, cobalt, lithium, and oxygen. Aluminum powder particle shape must be spherical with a smooth surface, appropriate size, the particles with larger size should not be more than 80 wt. %. The density of the aluminum powder is 2.7 g/cm³. Al-6061 alloy is a magnesium-based alloy used in the structural components manufacturing for the fuel element produced, in different geometrical shapes, like a sheet, plate, rod, and rectangular bar. The advantage of the Al-based magnesium alloy 6061 is their increased corrosion resistance. The uranium oxide powder as U₃O₈ must fulfill the requirements of the fuel element produced for our reactor (Elseaidy & Ghoneim, 2010; Mansour & Elseaidy, 2016).

The fuel compact has the following dimensions and weight:

- Width, 60.5 ± 0.3 mm, Length 69.0 ± 0.3 mm, thickness 8.5 ± 0.2 mm and weight 171.6 ± 1.5 g. The compact density is about 4.8 g/cm³, equivalent to 3.1 g U/cm³. The content of the isotope U-235 must be 21.3 ± 0.2 and 0.7785 ± 0.009 g for enriched and natural compact, respectively. The surface of compact must be free of any faults such as pores, cavities, fissures, scaling, cracks, and/or any other type of material discontinuity.

The specification of the used detector is High-Purity Germanium detector (HPGe), model Canberra GL0515R with an active area of 540 mm². The active volume height is 1.5 cm and FWHM at 122 keV is 540 eV. Multi-channel analyzer (inspector, Model IN2K), to collect the input pulses, the operating voltage for the detector is (~2500V) (Canberra, 1996; Canberra Industries, 1995; Gunnink & Miller et al., 1994). The sample used here is a compact of uranium oxide and aluminum alloy. The sample volume is 35.48 cm³ and its density is 4.8 g/cm³. The manufacturer data about the mass of U-235 is 0.7785 and 21.3 g. The compact sample has a parallelepiped shape. It has dimensions of 69, 8.5 and 60.5 mm.

3.2. Monte Carlo modeling for measuring system

Monte Carlo simulation based on a sequence of random trials that happens during the simulation. The simulation will get results within some ‘statistical error’ (David & Kurt, 2009). The General Monte Carlo Code (MCNP-5) was utilized to compute the absolute efficiency of the detector (El-Gammal et al., 2005). The characteristics and specifications of the planar HPGe detector and U₃O₈-Al Compact have modeled as
many details as possible to simulate the experimental setup. The executed input files for this calculation were 28 input files. The histories that used in the created input files were $10^8$ histories (number of photons) with run time 20 min. The specifications of the used laptop are 2.5 GHz Intel Core i7 processor since a tally F8 is used to determine the pulse height of the detector (MCNP- A general Monte Carlo N Particle Transport Code, 2003). The absolute efficiency of the detector at 185.7 keV energy line was calculated through that tally. The setup for natural and enriched Compact simulated by MCNP code is shown as in Figure 1.

In this work, the data outcomes from the experimental work in ref. (Abdalsamaha, 2009) for three distances 9.5, 10.8 and 11 cm was developed to predict the count rate, absolute efficiency and mass of U-235 at different distances within the experimental range from 9.5 to 11 cm.

4. Results and discussion

A Mathematical model for experimental results is an essential part to get the relation between the count rate and distance to predict another count rate values at different distances within the experimental range, MCNP calculations for proposed distances to get the absolute efficiency and U-235 mass calculations for the previous results by substituting in Equation (1). Finally, it presented different distortion scenarios for U-235 mass.

4.1. Mathematical model

Mathematical model is developed using correlation concluded from the relation between count rate and distance (sample–detector). The model also depends on an equation that results from the relation between count rate and absolute efficiency for an alloy of uranium oxide that is mixed and compacted with aluminum. The mathematical model has resulted from the polynomial fitting equation of the count rate – distance curve for three experimental values at distances 9.5, 10.8 and 11 cm as shown in Figure 2(a) and count rate – absolute efficiency curve as shown in Figure 2(b). From the fitting curves, another count rate values and absolute efficiencies can be achieved at different distances.

4.2. Model validation

In this section, the model was validated by substituting into the polynomial fitting equation to get the count rate and absolute efficiency and the use of Equation (1) to obtain U-235 mass. Tables 1a, 1b presented the relation between the source–detector distance, count rate at energy line 185.7 keV and the absolute efficiency of the detector. The Table described Predicted Gamma parameters at distances from 9.5 to 11 cm. Tables showed that the predicted values of count rates, absolute efficiencies, and estimated U-235 masses.

4.3. MCNP calculations

4.3.1. Results of U-235 mass calculations for a natural compact sample (0.72%)

Figures (3, 4 and 5) show that MCNP calculations for proposed distances within the experimental range to get the absolute efficiency, U-235 mass calculations in natural uranium (0.72% of U-235) for the previous results by substituting in Equation (1). These Figures interpreted that the count rate and absolute efficiency are calculated with associated uncertainties, the masses for distance values (9.5, 9.6, 9.7, 9.8, 9.9, 10, 10.1, 10.2, 10.3, 10.4, 10.5, 10.6, 10.7, 10.8, 10.9, and 11 cm) were estimated to be (0.76731, 0.77641, 0.78381, 0.79012, 0.78666, 0.78956, 0.80276, 0.80443, 0.80464, 0.80288, 0.80000, 0.79652, 0.79058, 0.78229, 0.76642, and 0.76691 g) respectively. The count rate uncertainty $\sigma_{CR}$ is produced from statistical uncertainties in the net area under 185.7 Peak which was ($\leq$ 1.1%). All estimated uncertainties due to Monte Carlo calculations for absolute efficiency values were less than 0.15%. The uncertainties of $^{235}$U masses in MCNP-5 calculations were found to be in the range from 0.95% to 1.18%, the difference between the masses estimated using the MCNP-5 and declared (operator measurements) were 1.4 and $-2.62\%$.

The results show that the optimum distance at which the U-235 mass that predicted is matched with the declared was at 10.9 cm.
### 4.3.2. Results of U-235 mass calculations for the compact sample with enrichment (19.75%)

Table 2 shows predicted count rates and MCNP calculations from 9.6 to 10.9 cm distances to get the absolute efficiency.
efficiency, U-235 mass calculations in enriched uranium (19.75% of U-235). This section shows that the MCNP code is validated for measuring U-235 mass at enrichment 19.75%. The uranium-235 mass that depends on MCNP calculations is in agreement with the declared operator values (21.3 g).

### 4.4. Distortion scenarios

The following data discuss various safeguard scenarios outcome from the change in chemical mixing of the compacted material (19.75 of uranium enriched) in which the uranium mass decreased by 8.450% as shown in Table (3). Using Equation (1), the mass of uranium-235 is obtained. As a result of the sample distortion, the uranium mass became 19.5 g.

Table (4) describes the distortion in enrichment ratio to be 6.103%. The uranium-235 mass is changed to be 20 g.

Table (5) presented significant data where a small difference in enrichment values (from 6.103 to 6.103286), means a difference in small fraction upon three digits from the previous values, the uranium mass is changed with difference 0.6 g. which represents significant data in nuclear safeguards measurements.

### 5. Conclusion

The process of manufacturing a compact of mixed uranium oxide with aluminum is an essential stage in the production of fuel plates in MTR type fuel. This Stage should be supervised by the operator in quality control (QC) as small distortions in manufacturing compact leads to change in the uranium-235 mass. The compact in storage or that in operation should be verified by safeguards inspectors. The values of U-235 mass represent significant data in nuclear safeguards measurements. The mathematical model for experimental results was executed to predict another count rate values for different distances within the experimental range, MCNP calculations for proposed distances to get the absolute efficiency and U-235 mass calculations. Finally, different distortion scenarios were presented for U-235 mass. This method is considered more useful for the inspector to be able to predict the Uranium-235 mass in case if any

### Table 2. Predicted count rates and U-235 mass calculations at distances from 9.6 to 10.9 cm.

| Distance(cm) | Count rate | Absolute photo peak efficiency | 235U M (g) ±σM |
|--------------|------------|--------------------------------|----------------|
| 9.6          | 1.97096E+02 ± 0.52802 | 2.00577E-04 ± 0.96276E-8 | 21.30000 ± 0.05706 |
| 9.7          | 1.93886E+02 ± 0.51942 | 1.97310E-04 ± 0.94708E-8 | 21.30000 ± 0.05706 |
| 9.9          | 1.87817E+02 ± 0.51771 | 1.91134E-04 ± 0.91744E-8 | 21.30000 ± 0.05871 |
| 10.9         | 1.60520E+02 ± 0.47578 | 1.63355E-04 ± 0.08984E-8 | 21.30000 ± 0.06313 |

### Table 3. Distortion in chemical mixing leads to a decrease by 8.450% of uranium mass.

| Distance(cm) | Count rate | Absolute photo peak efficiency | 235U M (g) ±σM |
|--------------|------------|--------------------------------|----------------|
| 9.6          | 1.940629E+02 ± 0.49777 | 2.15720E04 ± 0.97074E-8 | 19.50000 ± 0.05001 |
| 9.7          | 1.909125E+02 ± 0.48969 | 2.12218E04 ± 0.97620E-8 | 19.50000 ± 0.05001 |
| 9.9          | 1.848950E+02 ± 0.474252 | 2.05529E04 ± 0.94543E-8 | 19.50000 ± 0.05001 |
| 10.9         | 1.581273E+02 ± 0.450662 | 1.75774E04 ± 0.96675E-8 | 19.50000 ± 0.05557 |

### Table 4. Distortion in chemical mixing leads to decrease by 6.103% of uranium mass.

| Distance(cm) | Count rate | Absolute photo peak efficiency | 235U M (g) ±σM |
|--------------|------------|--------------------------------|----------------|
| 9.6          | 1.94955E+02 ± 0.51117 | 2.11294E04 ± 0.97221E-8 | 20.00000 ± 0.05244 |
| 9.7          | 1.91839E+02 ± 0.50300 | 2.07917E04 ± 0.95642E-8 | 20.00000 ± 0.05244 |
| 9.9          | 1.85801E+02 ± 0.49776 | 2.01373E04 ± 0.94652E-8 | 20.00000 ± 0.05358 |
| 10.9         | 1.58841E+02 ± 0.46175 | 1.72153E04 ± 0.87846E-8 | 20.00000 ± 0.05810 |

### Table 5. Distortion in chemical mixing leads to a decrease by 6.103286% of uranium mass.

| Distance(cm) | Count rate | Absolute photo peak efficiency | 235U M (g) ±σM |
|--------------|------------|--------------------------------|----------------|
| 9.6          | 1.960055E+02 ± 0.513392 | 2.06245E04 ± 0.94984E-8 | 20.60000 ± 0.05401 |
| 9.7          | 1.928674E+02 ± 0.51669 | 2.029430E04 ± 0.93418E-8 | 20.60000 ± 0.05318 |
| 9.9          | 1.868165E+02 ± 0.50048 | 1.96576E04 ± 0.92451E-8 | 20.60000 ± 0.05316 |
| 10.9         | 1.596906E+02 ± 0.46422 | 1.68033E04 ± 0.85772E-8 | 20.60000 ± 0.05988 |
distortion occurred in the manufacturing of the compact that is used in the production of fuel plates in MTR-type fuel.

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**Disclosure statement**

No potential conflict of interest was reported by the authors.

**Compliance with Ethics Requirements**

This article does not contain any studies with human or animal subjects.

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