Determination of ^{238}U and ^{235}U isotope masses based on passive neutron coincidence measurements using an AWCC

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SUMMARY

- eliminates the need for physical standards via Geant4-based mathematical calibration
- accounts for spontaneous and induced fission, and (α, n) in UO_2
- validated on LEU RBMK/WWR pellets (362–1802 g)
- achieves uncertainties <2 % for ^{238}U and ≈1 g for ^{235}U (>300 g)

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1. INTRODUCTION

Accurate quantification of uranium is of critical importance for the domains of international nuclear safeguards, nuclear material accountancy, and nuclear forensics. Non-destructive assay (NDA) techniques of a classical nature frequently entail the active interrogation of a sample by means of an external neutron source. Alternatively, a set of calibration standards with predefined geometry and matrix composition is required. In practice, this significantly limits the universality of material verification, since real objects differ in shape, dimensions, enrichment, and matrix composition.

Geant4, a Monte Carlo radiation transport toolkit, facilitates the simulation of passive measurements of registered neutron coincidences for arbitrary experimental configurations and samples.

In this study, a single Geant4 simulation cycle is constrained to the tracking of neutrons – encompassing their emission, transportation, secondary neutron production from induced fission, and detection. The sequence commences with a primary event, which is either the emission of neutrons from spontaneous fission or an (α, n) reaction and is concluded once all neutron tracks within the simulated geometry have been processed. It is now established that tracks of other particles produced during neutron transport are discarded immediately upon their creation, since the correlated contribution of delayed neutrons that could originate from these tracks is negligibly small. Concurrently, the $^3He(n, p)^3H$ reaction occurring within the detection region of the Active Well Coincidence Counter (AWCC) is processed, as it serves as the trigger for the detector signal.

The primary parameter of the model is the isotopic composition of the nuclear material, while the properties of the measuring equipment and the matrix are derived from reference data or determined separately through experimentation. In a series of simulated experiments, the resulting neutron multiplicity distribution is optimised with respect to isotopic composition until agreement with the experimental distribution is achieved.

The total neutron multiplicity histogram from several simulated spontaneous fission events of ^{238}U and (α, n) reactions of ^{234}U in Geant4 is utilised as the output of a simulated experiment, corresponding to the mean value for given isotope masses over the measurement duration.

2. EXPERIMENTAL SETUP AND SAMPLES

The AWCC model JCC-51, which is equipped with an array of ^3He proportional counters embedded in a polyethylene moderator, was utilised in this study [1]. The central well is notable for its capacity to provide a reproducible geometry for sample placement. The accumulation of neutron multiplicity histograms recorded by the AWCC was achieved through the utilisation of a Canberra JSR-15 shift register.

The investigation focused on UO_2 fuel pellets from RBMK and VVER reactors with ^{235}U enrichment levels reaching 4.4 %, with a mass range of 362–1802 g. To evaluate the background, "inactive" lead inserts with equivalent mass were used.

Neutron measurements were conducted in the absence of a sample, and a correction was determined for interactions of cosmic rays with uranium. This correction was derived based on the observed dependence of the neutron coincidence rate on lead

mass. Lead samples were positioned in the AWCC chamber at the same location as the investigated UO_2 samples. As illustrated in Figure 1, this dependence is evident in triple coincidences, for which the correction is most significant.

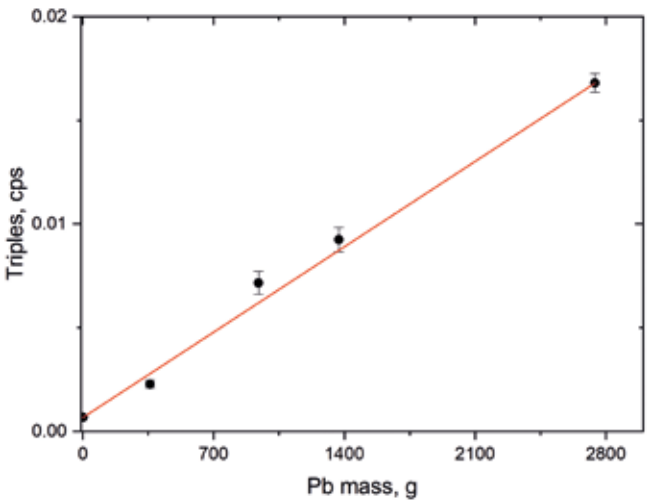


Figure 1: Dependence of the triple neutron coincidence count rate on the lead mass.

3. METHODOLOGY OF PASSIVE NEUTRON COINCIDENCE COUNTING

The neutron multiplicity distribution is described using the factorial moments of the registered neutron multiplicity histogram, where each moment corresponds to the count rate of coincidences of a given order.

The Canberra JSR-15 shift register has been observed to measure two histograms: one for the prompt time window, which is comparable to the neutron die-away time in the detector block and contains both correlated and random neutron events, and one for the delayed window, which accounts for neutron detections separated in time by several orders of magnitude and represents purely random events. In both cases, the trigger signal is the detection of a neutron.

The histogram obtained in the prompt window represents the sum of accidental (random) neutrons and correlated neutrons generated by fission events associated with the trigger neutron. The delayed histogram is comprised of random coincidences. The measurement of both multiplicity histograms is an essential step in the process, as it enables the subtraction of the random component and the determination of the correlated neutron count rates, otherwise known as the multiplicities. The acronyms S, D and T are used to denote singles, doubles and triples respectively [1].

4. MODELLING AND MATHEMATICAL CALIBRATION

The AWCC geometry (Fig. 2) was reproduced in Geant4 v11.2 [2]. The QGSP_BERT_HP physics list was employed to ensure accurate treatment of neutron transport and interactions with matter, including low-energy neutron scattering. The ThermalNeutronScattering model was utilised to describe the phenomenon of elastic scattering in polyethylene and structural materials.

The neutron multiplicity and energy spectrum from the spontaneous fission of ^{238}U were modelled using the parameters provided in [3]. In addition to neutron resulting from the spontaneous fission of ^{238}U and induced fission of ^{235}U and ^{238}U, measurements of enriched uranium also register neutrons originating from (α, n) reactions, produced by α-particles emitted by ^{234}U when interacting with ^{17}O and ^{18}O in UO_2. In order to account for this process, the results of mass spectrometric measurements of the ^{234}U content in low-enriched samples (2 % and 3.6 % ^{235}U) were used [4], while for pellets enriched to 4.4 % the ^{234}U content was determined using gamma spectrometry. The neutron yield from (α, n) reactions in uranium dioxide was derived from [5], and the neutron spectrum was obtained from [6].

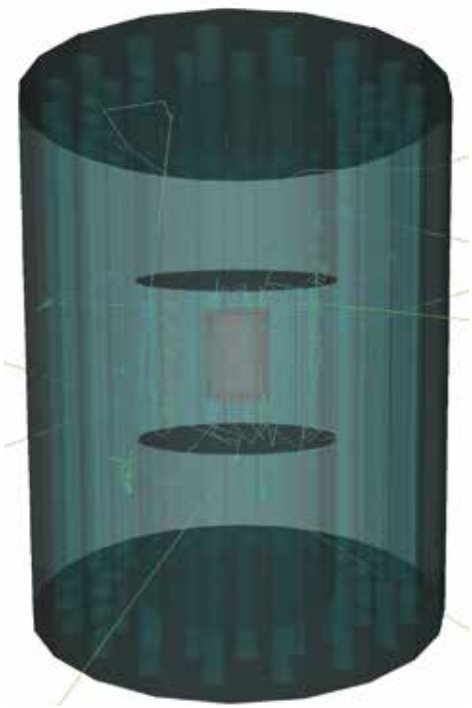


Figure 2. Geant4 model geometry of the AWCC.

The objective of this study is to determine the absolute masses of ^{238}U and ^{235}U. To this end, the $\chi^2(m_{238}, m_{235})$ functional is minimized by comparing calculated and measured S, D, and T values. At each step of the optimisation process, the simulation computes the expected count rates corresponding to a candidate pair of parameters (m_{238}, m_{235}). The ^{234}U content is determined from the $^{234}U/^{238}U$ ratio for low enrichments based on independent measurements.

5. OPTIMISATION

The dependence of the simulated experiment outcome on the isotopic composition is a stochastic function, therefore fitting the simulated S, D, T count rates to the experimental values presents several challenges and requires optimisation methods that are robust against noise in the $\chi^2(m_{238}, m_{235})$ objective function.

However, for the mass range of ^{238}U and ^{235}U investigated in our experiment, and for the corresponding count rates, the classical Nelder–Mead simplex method proved to be adequate, with the only drawback being a relatively long computation time.

It was found that, in the general case, when fitting a larger number of isotopic mass parameters and incorporating higher-order multiplicities, only stochastic optimization methods with adaptive sampling proved suitable, owing to their robustness against noise and local minima.

6. UNCERTAINTY ANALYSIS

In a series of measurements, the mean values of the multiplicity count rates are determined using a weighted average with weights inversely proportional to the variance. To mitigate the influence of outliers (e.g., cosmic-ray bursts), data points deviating from the mean by more than a predefined rejection threshold (commonly 2σ–3σ) are iteratively excluded. The rejection criterion was selected in accordance with established practices for Poisson-distributed counting statistics and verified to avoid introducing bias into the estimated mean values.

The statistical uncertainties of the S, D, and T count rates are evaluated assuming Poisson statistics, considering gate fractions of the multiplicities, under the assumption of a mono-exponential Rossi-α decay profile [1].

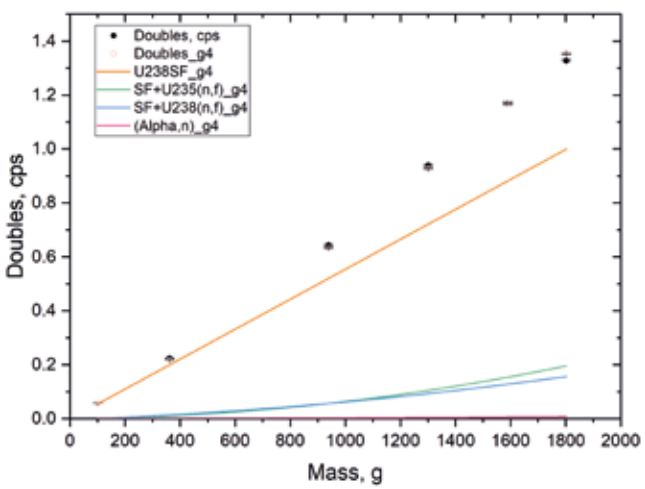
Uncertainties in the isotope masses are derived from the covariance matrix, obtained as the inverse of the Hessian of χ^2 at the minimum. The Hessian is evaluated numerically using a finite-difference approach: each isotope mass is varied independently in the Geant4 model, and the resulting changes in χ^2 are used to approximate the partial derivatives.

The sensitivity analysis also indicates that, under our experimental conditions, doubles (D) and triples (T) provide the dominant contribution to mass sensitivity, while the influence of the (α, n) model is constrained by the independent estimate of the ^{234}U content.

7. RESULTS

Over the entire investigated mass range, an approximately linear increase in singles (S) is observed, dominated by spontaneous fission of ^{238}U. In contrast, doubles (D) and triples (T) exhibit a much steeper, nonlinear increase due to neutron multiplication and induced fission. For the heaviest samples, the contribution of induced fission to T reached approximately 25 %, confirming the importance of accurately modelling induced fission chains.

a)



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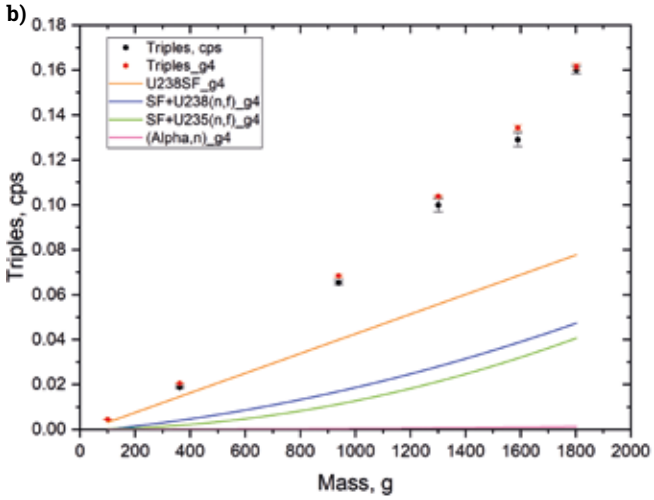
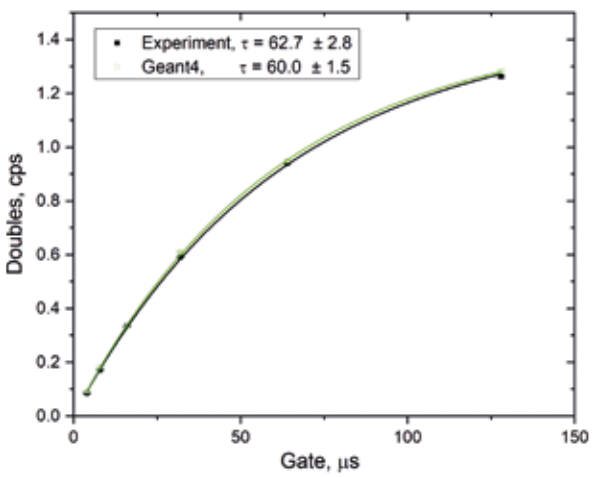


Figure 3. Dependence of double (a) and triple (b) neutron coincidence count rates on the mass of uranium with 2 % enrichment.

The experimental Rossi-α distributions were found to be in good agreement with the simulation results, confirming the correctness of the applied physics models (Fig. 4).

a)



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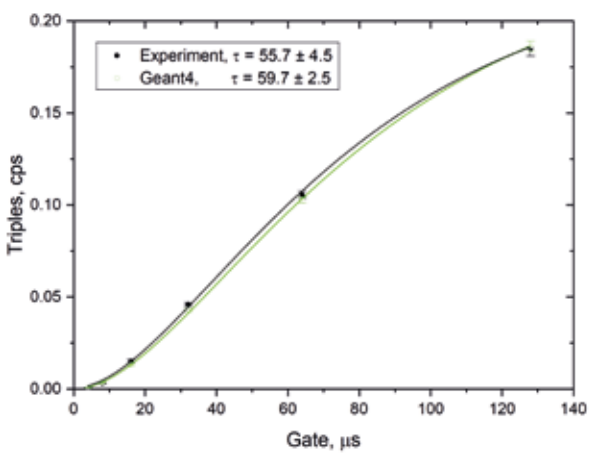

Figure 4. Dependence of the double (a) and triple (b) neutron coincidence count rate on the gate width during measurements of a sample with a mass of 1302 g and enrichment of 2 %.

Comparison with the true masses demonstrates agreement within statistical uncertainties; systematic deviations do not exceed the declared accuracy limits. Data fitting achieved a mass determination accuracy of 1–2 % for ^{238}U(a) and approximately 1 g for ^{235}U(b) for samples with mass greater than 300 g.

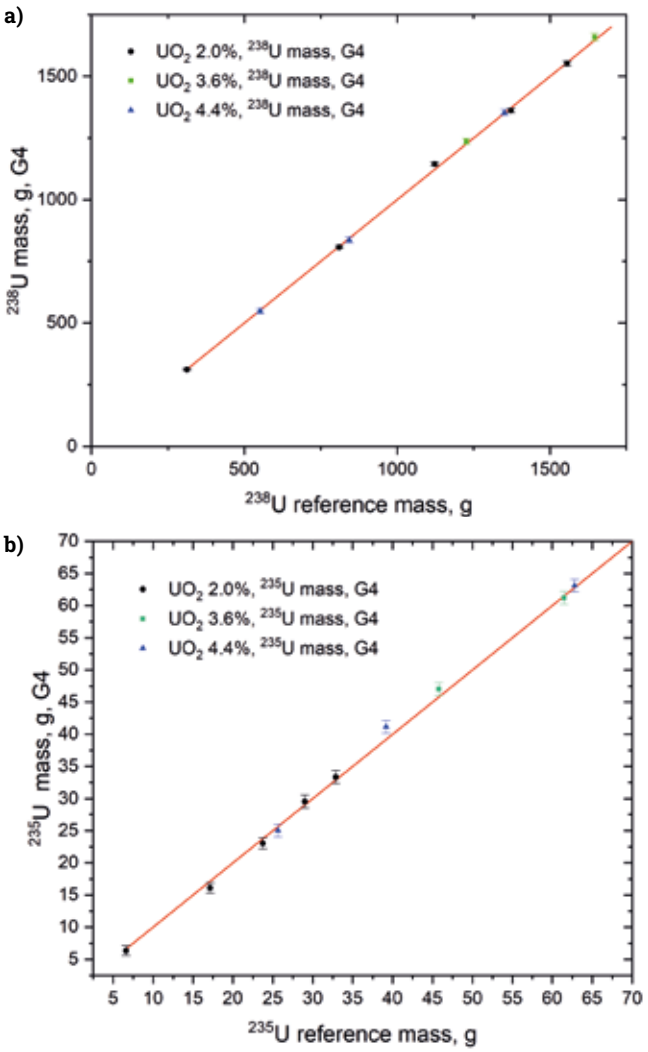

Figure 5. "Derived mass" versus "true mass" for ^{238}U (a) and ^{235}U (b).

The advantages of the proposed approach include independence from physical standards, applicability to various geometries and levels of enrichment, and formal treatment of (α, n) reactions and induced fission.

8. LIMITATIONS AND OUTLOOK

The method is sensitive to the correct description of (α, n) reactions in the UO_2 matrix. For very small sample masses (≤300 g), the information content of the triple coincidences T decreases significantly, which increases the uncertainty of the ^{235}U mass determination.

Future development directions include:

- List-mode analysis without rigid time-gating constraints
- Extension of the model to heterogeneous matrices (e.g. UO_2 with additives or cladding materials)

9. CONCLUSIONS

A passive method for determining the masses of ^{238}U and ^{235}U in an AWCC without physical calibration standards has been proposed and demonstrated.

Mathematical calibration based on Geant4 simulations enables the application of the model to various geometries and enrichment levels.

An accuracy of <2 % for ^{238}U and approximately 1 g for ^{235}U was achieved for samples with masses greater than 300 g.

The approach meets the requirements of non-destructive assay (NDA) for non-proliferation safeguards and nuclear forensic applications and is ready for practical implementation.

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