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## ABSTRACT

Neutron coincidence and multiplicity counting is a standard technique used to measure uranium and plutonium masses in unknown samples for nuclear safeguards purposes, but background sources of radiation can obscure the results. In particular, high energy cosmic rays can produce large coincidence count contributions. Since some of the events occur in the sample itself, it is impossible to measure the background separately. This effect greatly increases the limit of detection of some low level neutron coincidence counting applications.

The cosmic ray capability of MCNP6 was used to calculate the expected coincidence rates from cosmic rays for different sample configurations and experimental measurements were conducted for comparison. Uranium enriched to 66%, lead bricks, and an empty detector were measured in the mini Epithermal Neutron Multiplicity Counter, and MCNP6 simulations were made of the same measurements. The results show that the capability is adequate for predicting the expected background rates.

Additional verification of MCNP6 was given by comparison of particle production rates to other publications, increasing confidence in MCNP6's use as a tool to lower the limit of detection. MCNP6 was then used to find particle and source information that would be difficult to detect experimentally. The coincidence count contribution was broken down by particle type for singles, doubles, and triples rates. The coincidence count contribution was broken down by source, from(a, n), spontaneous fission, and cosmic rays, for each multiplicity.

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## 1. Introduction

Neutron coincidence and multiplicity counting use time correlated neutron detections and knowledge of fission neutron production to find the mass of the sample. Multiple time-correlated neutrons typically result from fissions, which indicate the amount of nuclear material. Background sources of radiation can obscure the signal resulting in higher minimum detectable sample mass and potentially biasing the results. Uranium-238's low spontaneous fission production of 0.014 n s<sup>-1</sup> g<sup>-1</sup> [1] means that sample masses are frequently close to, or below the minimum detectable activity.

High energy cosmic rays can produce events with large neutron multiplicities which give rise to large coincidence count

\* Corresponding author at: University of Florida, 100 Rhines Hall, 549 Gale Lemerand Drive, PO Box 116400, Gainesville, Florida, 32611, United States. *E-mail address*: rweinmann@ufl.edu (R. Weinmann-Smith). contributions, as illustrated in Fig. 1. This example shows multiple neutron particle tracks being created by cosmic ray spallation in a lead brick sample. This effect greatly increases the limit of detection of some low level neutron coincidence counting applications.

The Monte Carlo N-Particle transport code (MCNP6) has a cosmic ray capability [2] which was used to calculate the expected detector response to coincidence counts from cosmic rays for an empty detector, lead bricks, and 66% enriched uranium. Experimental measurements were conducted for comparison to the calculations. Comparisons of certain results calculated with MCNP6 were made to other publications. MCNP6 was then used to find particle and source information that would be difficult to determine experimentally.

#### 2. Neutron multiplicity measurement method

Neutron multiplicity is used to assay uranium and plutonium samples for nuclear material accountancy and nuclear safeguards. Spontaneous fissions have a distinct neutron multiplicity distribution, where the probability of releasing each discrete number



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**Fig. 1.** MCNP6 example of a high energy neutron spallation in a  $20 \times 20 \times 30$  cm<sup>3</sup> lead brick plotted in MORITZ [3].

of neutrons is known. Time-correlated neutrons are recorded in a neutron multiplicity distribution, which can be used to calculate total and coincidence count rates and quantify the sample.

The neutron counter used here is a mini Epithermal Neutron Multiplicity Counter (miniENMC) [4]. It uses a moderator to slow neutrons to thermal energy, and then helium-3 tubes to capture the neutrons. Every neutron pulse opens a gate for a certain amount of time where coincidence neutrons are recorded. To account for accidental coincidences, a second gate opens after a relatively long period of time, allowing any correlated neutrons to die away. Subtracting the two gives the real coincidences. Accidental coincidences come from uncorrelated overlaps from any source of neutrons, including separate spontaneous fissions, ( $\alpha$ , n) reactions, background neutron sources, and cosmic rays [5]. The time correlated cosmic ray neutrons are detected as real coincidences.

The factorial moments of the measured multiplicity distribution are frequently used in multiplicity counting. The first three moments of the multiplicity distribution are used to calculate the singles, doubles, and triples. The singles rate is the total number of counts per time. The doubles rate is the total counts in the "real" multiplicity distribution bin multiplied by the number of neutrons captured in the gate per time. The triples rate is calculated using Eq. (1) per unit time, which gives the number of times three pulses are recorded. In the equation, *r* is the reals, *a* is the accidentals, and *n* is the multiplicity. The singles, doubles, and triples, can be used to estimate the mass of a sample [1].

$$\sum_{n=0}^{\max} \left( r_n \frac{n(n-1)}{2} \right) - \frac{\sum_{n=0}^{\max} (a_n n) \sum_{n=0}^{\max} (r_n n)}{\sum_{n=0}^{\max} a_n}$$
(1)

#### 3. Cosmic rays, origin and effects

Galactic cosmic rays are generated from a combination of galactic phenomenon such as stellar flares, supernova explosions,

pulsars, and the explosion of galactic nuclei. They are considered to be isotropic due to various magnetic field interactions and 200 million year mean half-life in the galaxy. The galactic particles consist of approximately 87% protons, 12% alpha particles, and 1% heavier nuclei, with energies above 1 GeV required to reach the surface of the Earth. They produce secondary particles in the atmosphere and these compose the majority of particles that reach the surface. These particles are mostly pions, muons, nucleons, electrons, and photons [6–8].

Solar particles are about 98% protons without enough energy to penetrate the earth's atmosphere, so they are ignored here. However, the sun does influence the strength of the magnetic field around the Earth. At peak solar activity the Earth's magnetic field is amplified, causing cosmic rays to lose energy and reducing the number of cosmic rays that reach the surface of the earth by a maximum of 30%. The sun's cycle lasts 11 years and, as of May 2014, is in the last few years of its peak [9].

Longitude, latitude, and altitude also affect the cosmic ray intensity. At high latitudes the magnetic field is weaker allowing cosmic rays to penetrate further. Altitude greatly affects secondary cosmic rays, as air is their primary attenuator. To account for changing atmospheric density, the altitude is measured in units of g cm<sup>-2</sup> of atmosphere above a height. Los Alamos, New Mexico averaged about 810 g cm<sup>-2</sup> during this study. The cosmic ray variation with time and location has to be considered to account for their contribution to neutron multiplicity counting.

### 4. Measurements

Measurements were taken with a Mini-Epithermal Neutron Multiplicity Counter [4]. It is a neutron multiplicity detector with a split sample cavity, which can be seen in Fig. 2. It has 104 helium-3 tubes at a pressure of 10 standard atmospheres in four concentric rings. The detector efficiency is 61.8% and the die-away time is 19.1 µs, which makes it ideal for precise multiplicity counting. The measured lead was in the form of two standard bricks and a total of 22.6 kg. The uranium was 1 kg of  $U_3O_8$ , 66% enriched in <sup>235</sup>U, and contained in tinned-steel [10]. Lead was chosen as a surrogate for fissionable materials because of its high Z number which should produce similar cosmic ray reactions to uranium and plutonium, without the spontaneous fission and  $(\alpha, n)$  reactions. The measurements were taken 15 months after the selected date for the MCNP6 simulations. The simulations had already been performed and could not be repeated for the measurement date due to the intense computer time required. Variation due to this is expected to be minimal as the solar cycle plateaued during this time. While this is sufficient for demonstrating the principles of the calculation, real applications would require carrying out the simulation at the correct date and location.

#### 5. MCNP6 calculation method

The detector counting rate calculation was split into several stages to reduce the amount of computing time required. The cosmic ray source calculation began with atmospheric transport of the galactic cosmic ray spectrum incident on the Earth at an altitude of 65 km. where it was tallied at an altitude of 2 m above the ground at Los Alamos. Then, a calculation was done to find the ratio of particles that would reach the top and the sides of a box that enclosed the detector. This ratio was used to construct a source definition for the top and sides of the box to model the fully detailed detector and sample configurations.

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