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First principle active neutron coincidence counting measurements of uranium oxide

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ABSTRACT

Uranium is present in most nuclear fuel cycle facilities ranging from uranium mines, enrichment plants, fuel fabrication facilities, nuclear reactors, and reprocessing plants. The isotopic, chemical, and geometric composition of uranium can vary significantly between these facilities, depending on the application and type of facility. Examples of this variation are: enrichments varying from depleted ($\sim$0.2 wt% ^{235}U) to high enriched ($>$20 wt% ^{235}U); compositions consisting of U_3O_8, UO_2, UF_6, metallic, and ceramic forms; geometries ranging from plates, cans, and rods; and masses which can range from a 500 kg fuel assembly down to a few grams fuel pellet. Since ^{235}U is a fissile material, it is routinely safeguarded in these facilities. Current techniques for quantifying the ^{235}U mass in a sample include neutron coincidence counting. One of the main disadvantages of this technique is that it requires a known standard of representative geometry and composition for calibration, which opens up a pathway for potential erroneous declarations by the State and reduces the effectiveness of safeguards. In order to address this weakness, the authors have developed a neutron coincidence counting technique which uses the first principle point-model developed by Boehnel instead of the "known standard" method. This technique was primarily tested through simulations of 1000 g U_3O_8 samples using the Monte Carlo N-Particle eXtended (MCNPX) code. The results of these simulations showed good agreement between the simulated and exact ^{235}U sample masses.

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1. Introduction and theory

1.1. Neutron coincidence counting theory

Neutron coincidence counting is a technique used in safeguards and nuclear waste management to quantify the mass of spontaneous fission or fissile nuclear materials in a container, such as plutonium or uranium. Passive neutron measurements of plutonium utilize the spontaneous fission rate of even plutonium isotopes (^{238}Pu, ^{240}Pu, and ^{242}Pu) by passively determining the fission rate in the sample, and thus its effective plutonium mass. This is achieved through the use of the first principle point-model equations developed by Boehnel in 1984 [1]. Due to the low spontaneous fission rate of uranium, neutron measurements of uranium require a neutron interrogation source to induce fission in the ^{235}U atoms. Measurements of this type are commonly referred to as "active measurements". The most common interrogation source is americium–lithium (AmLi) [2]. This source produces neutrons singularly through an (α,n) reaction, thus minimally affecting the coincidence neutron count rate. Neutrons from an AmLi source have an average neutron energy of about 0.3 MeV [3] which is high enough to penetrate most uranium samples with minimal attenuation, thus creating a homogeneous fission rate. However, ^{3}He-based neutron coincidence counters use High-Density Polyethylene (HDPE) to slow down neutrons to thermal energies in order to increase the probability of neutron capture. During the slowing down process, neutrons with epithermal energies can leave the HDPE and enter the uranium sample. The probability of inducing fission in ^{235}U is significantly larger for epithermal neutrons than fast neutrons due to resonance fissions. This difference can be seen in the fission cross section of ^{235}U, shown in Fig. 1 [4]. This large fission cross section creates a fission rate in the sample predominately on the surface [5]. This heterogeneous fission rate prevents the point-model from being applied to assay measurements of ^{235}U because it violates the point-model assumption that the source of all neutrons can be approximated as a point. The traditional approach to active neutron measurements of uranium samples is to use a "representative sample" or "known standard" approach for material quantification.

1.2. Neutron coincidence counting uranium assay methods

The "known standard" measurement method is a common technique used for ^{235}U assay in which known uranium standards

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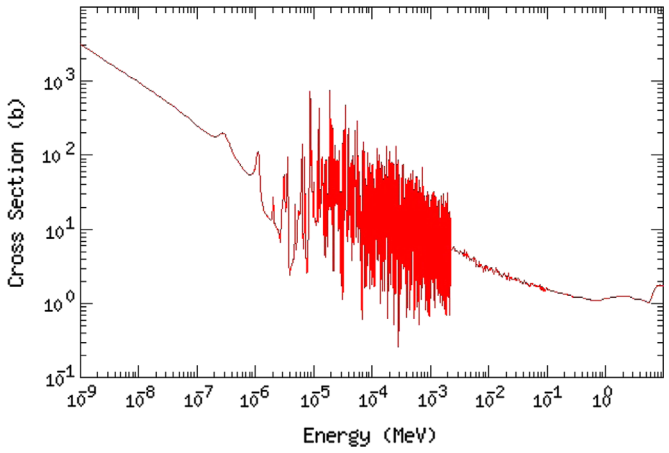

Fig. 1. Neutron induced fission cross section of ^{235}U [4].

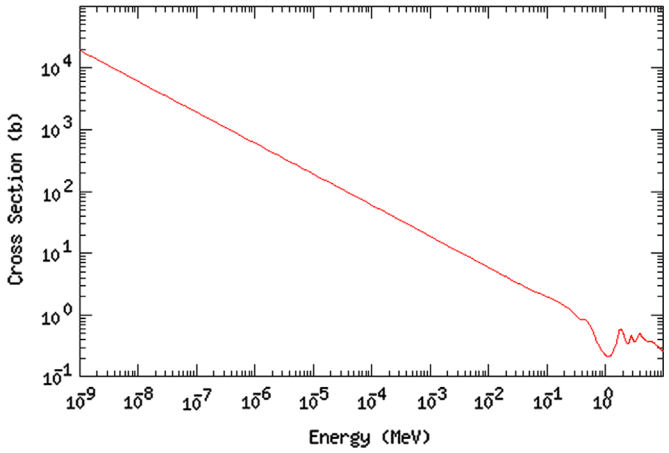

Fig. 2. Absorption cross section of ^{10}B [4].

with representative composition are used to create a calibration curve based on the measured doubles count rate (D). The main disadvantage of this method is that only uranium samples which are representative of known standards can be measured. Other measurement methods have tried to address this shortcoming, such as the coupling method [2,6], but even these methods still require measurements of known standards. The method which we have developed uses a boron-carbide (B_4C) filter to surround the sample. This filter prevents the majority of neutrons with epithermal energies from reaching the uranium sample. This is achieved by the large ^{10}B absorption cross section at thermal ($\sim 4\times 10^3$ b) and epithermal ($\sim 10^2$ b) neutron energies, which can be seen in Fig. 2 [4]. Since only fast neutrons can enter the uranium sample, the fission rate in the sample will be nearly homogeneous. This homogeneous fission rate allows for a modified version of the point-model equation to be applied to uranium assay. It should be noted that method currently assumes a minimal amount of moderation within the sample.

1.3. First principle uranium assay method

For active measurements based on first principles methods, an active set of point-model equations can be derived. These equations are similar to their passive versions with two main differences. The first difference is that the α emission rate of uranium is small enough that the α variable in the point-model equations can be approximated as zero.

The second main difference is the factorial moments, which are defined as the average number of neutrons (ν_1) and neutron pairs (ν_2) emitted from a fission event. In the passive point-model equations, there are two factorial moments ν_s (spontaneous fission) and ν_i (induced fission). Since the spontaneous fission rate of uranium is small, all fission events can be considered to be from induced fission. There are two main sources of neutrons in this detector system: neutrons with fission energies from induced fission events (from the sample being measured) and neutrons with (α,n) energies from the interrogation sources (typically located at the top and bottom of a neutron coincidence counter). Depending on the (α,n) source used, these two factorial moments can have noticeably different values. Due to the approximate induced fission threshold for ^{238}U at 1 MeV, induced fissions in ^{238}U from neutrons with an AmLi energy spectrum (average of 0.3 MeV) are considered negligible. Neutrons with fission energies can cause fission in ^{238}U, but this is accounted for in the in the samples self-multiplication. The active doubles equation for an AmLi (α,n) source is [2]

$$D=\frac{F_o m_{U235}\varepsilon^2 f_d M_L^2 \nu_{Li2}}{2}\left[1+\frac{(M_L-1)\nu_{Li1}\nu_{Fis2}}{(\nu_{Fis1}-1)\nu_{Li2}}\right] \tag{1}$$

where D is the doubles count rate from the AmLi measurement, F_o is the specific fission rate within the uranium sample, m_{U235} is the mass of ^{235}U in the sample, ε is the neutron detector efficiency, f_d is the doubles gate fraction, M_L is the leakage self-multiplication, ν_{Li1} is the first moment of induced fission for neutrons with an AmLi energy spectrum, ν_{Li2} is the second moment of induced fission for neutrons with an AmLi energy spectrum, ν_{Fis1} is the first moment of induced fission for neutrons with a fission energy spectrum, and ν_{Fis2} is the second moment of induced fission for neutrons with a fission energy spectrum.

Because F_o, the specific fission rate, is based on the induced fission rate and not the spontaneous fission rate, it cannot be treated as a constant. F_o can instead be calculated from

$$F_o=\frac{\phi_{Li}\sigma_{f\_Li}N_A}{M_{molar}} \tag{2}$$

where ϕ_{Li} is the neutron flux within the sample during an AmLi measurement, σ_{f_Li} is the average ^{235}U fission cross section for neutrons with an AmLi energy spectrum, N_A is Avogadro's Number, and M_{molar} is the molar mass of ^{235}U. The neutron flux, ϕ_{Li}, and the average fission cross section for AmLi neutrons, σ_{f_Li}, can be considered independent of the sample being measured, for most measurements. Situations in which this may not be true include samples with a large amount of moderation or neutron absorbing materials. For measurements of low moderation and low neutron absorption, most samples which are typically measured, ϕ_{Li} and σ_{f_Li} can be determined through computer simulations of a generic measurement sample.

Like the point-model equations, Eq. (1) cannot be used directly for the calculation of ^{235}U mass. By combining Eqs. (1) and (2) and rearranging terms we acquire

$$m_{U235}=\frac{2DM_{molar}}{\phi_{Li}\sigma_{f\_Li}N_A\varepsilon^2 f_d M_L^2\nu_{Li2}[(1+((M_L-1)\nu_{Li1}\nu_{Fis2})/((\nu_{Fis1}-1)\nu_{Li2}))]} \tag{3}$$

All variables in Eq. (3) are known through nuclear data, detector calibration, sample independent computer simulations, or measured data, with the exception of m_{U235} and M_L. Since m_{U235} is what we are trying to determine, M_L must be approximated using knowledge about the sample, such as its expected density and geometry. For the results shown in this paper, M_L was estimated by total self-multiplication (M_T) values from detailed Monte Carlo N-Particle eXtended (MCNPX) simulations.