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^{236}U resonance parameters at 5.467 eV from neutron transmission measurements using thin liquid samples

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ABSTRACT

The ^{236}U isotope is an important buildup product that is generated in the ^{235}U fuel cycle and influences reactor neutronic calculations. The aim of the present work is to improve upon the existing neutron total cross section data for the strong ^{236}U resonance at 5.467 eV.

High accuracy neutron transmission measurements were performed using the time-of-flight technique at the Rensselaer Polytechnic Institute linear accelerator. An approach was used to fabricate thin ^{236}U samples using liquid, allowing for non-saturated resonances. The transmission measurements were made at the 15 m flight station with a ^{6}Li glass scintillation detector. Methods to characterize the background and experimental resolution function were developed. The ^{236}U resonance parameters and their uncertainties at 5.467 eV were determined by fitting the transmission data using a Monte Carlo approach with the SAMMY multi-level R-matrix Bayesian code.

The resonance parameters determined in this work are: energy, E, equal to 5.467 ± 0.005 eV; neutron width, Γ_n, equal to 2.13 ± 0.04 meV; and radiation width, Γ_γ, equal to 27 ± 1 meV. The fission width, Γ_f, was not fitted and was fixed to the ENDF-7.1 value of 0.290 meV. These parameters gave a neutron capture resonance integral of 330 ± 5 b that is lower than all of the selected evaluations: Mughabghab by 4.7%, ENDF-7.1 by 3.7%, JEFF-3.1 by 4.8%, and JENDL-4.0 by 7.2%.

1. Introduction

^{236}U is a significant buildup product that is produced from neutron capture in the ^{235}U fuel used in a majority of nuclear power reactors. When a ^{235}U nucleus absorbs a neutron, it forms the ^{236}U* compound nucleus. This compound nucleus may undergo a fission reaction or, alternatively, may decay to a ^{236}U nucleus through the capture (n,γ) reaction. The ^{235}U capture-to-fission cross section ratio at thermal energy (0.0253 eV) is approximately 17 captures per 100 fissions, or a 17% yield of ^{236}U. This is greater than the yield given for the most prevalent fission products (Baum et al., 2010). The buildup of ^{236}U influences the neutron population in a reactor core and fuel. Therefore, knowledge of the ^{236}U cross section is important to nuclear reactor design and criticality safety.

Despite its importance, very few total cross section measurements exist for the ^{236}U resonance at 5.467 eV. The extremely large peak cross section of this resonance (>13,000 b) results in zero transmission through thick samples. A transmission resonance that goes to zero is called “saturated” and much information about the peak total cross section is lost. Thin samples are difficult to fabricate to the tight tolerances needed for precision measurements. It is difficult to fabricate thin oxide powder samples as they may suffer from voids due to uneven settling and/or clumping. Other types of measurement, such as capture, self-indication, and activation have employed thin ^{236}U metal. Such thin metal samples are hard to manufacture with a uniform thickness. An approach is required which enables the use of an extremely thin ^{236}U sample to prevent the transmission from becoming saturated. Historically, such samples were fabricated using U_3O_8 oxide powder. The ^{236}U transmission measurements presented in this paper were done with the use of homogeneous liquid samples. Liquid samples have the benefit of being uniform and are much less susceptible to voids. Neutron capture measurements were not performed because they would not provide any appreciable information for this predominately capture resonance (Barry, 2003).

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A literature review was performed to find published values for the ^{236}U 5.467 eV resonance parameters using the Computer Index of Nuclear Reaction Data (CINDA, 2014). The CINDA results show the scarcity of total cross section (i.e., transmission) data for the ^{236}U resonance at 5.467 eV. Only the work of McCallum (1958) and Harvey and Hughes (1958) published resonance parameters (Table 6) resulting directly from a transmission current nuclear data evaluations because both experiments were performed prior to 1960 with poor energy resolution fast choppers and oxide powder samples. Due to the poor energy resolution, area analysis techniques were used to extract resonance parameters from the data. These experimental conditions and area analysis methods are suboptimal by modern standards. In contrast, the measurements presented in this paper have better energy resolution and do not use oxide powder samples. In addition, modern resonance shape fitting analysis techniques are used to extract the resonance parameters.

Other results from CINDA reporting ^{236}U 5.467 eV resonance parameters (Table 6) include neutron capture and self-indication measurements by Carlson et al. (1970) and the activation measurements of Baumann et al. (1968).

The ^{236}U 5.467 eV resonance parameters found in the ENDF-7.1 (Chadwick et al., 2011), JENDL-4.0 (Shibata et al., 2011), and JEFF-3.2 (Koning et al., 2006) nuclear data evaluations (Table 6) rely heavily on the evaluation of Mughabghab (1984). Because of the deficiency in ^{236}U total cross section measurements at 5.467 eV, the resonance parameters of Mughabghab are highly influenced by the work of Carlson et al. It should be noted that the evaluations do not incorporate the most recent values given by Mughabghab (2006).

Due to the lack of available total cross section data for the ^{236}U resonance at 5.467 eV and the importance of this isotope in the ^{235}U fuel cycle, we performed a new measurement at the linear accelerator (LINAC) located at Rensselaer Polytechnic Institute (RPI). This paper uses an approach to fabricate thin ^{236}U samples using liquids, allowing us to accurately determine the transmission through the 5.467 eV resonance for the first time since 1958.

2. Material and methods

As incident neutrons pass through a uniform thickness of material they may be transmitted through the sample and be detected or may undergo an interaction with the sample, such as elastic scattering or capture, preventing it from being detected. By measuring the ratio of counting rates in the detector with and without the sample in the beam the transmission can be calculated.

The transmission is related to the total cross section by the following equation:

$$T(E) = e^{-n\sigma_t(E)} \quad (1)$$

where n is the number density of the target material (units of atoms/barn) and $\sigma_t(E)$ is the total cross section at energy E in units of barns (10^{-24} cm^2).

A measurement of the ^{236}U resonance at 5.467 eV presents a significant challenge since this resonance has a very large peak cross section that can easily cause the transmission to be zero if the sample thickness (i.e., number density) is too large, rendering Equation (1) unusable when solving for the total cross section.

2.1. Liquid samples

For convenience in discussion and comparison, the experiment and results are described as “RPI-2014”. The RPI-2014 transmission experiment used three liquid samples containing different concentrations (i.e., thickness) of ^{236}U. The thickness for each sample was chosen to produce a non-saturated transmission resonance at 5.467 eV. The thickest sample provided a minimum transmission of approximately 0.1. The liquid samples were prepared by Oak Ridge National Laboratory (ORNL, 2013) by dissolving U_3O_8 oxide powder (89% enriched in ^{236}U) into deuterated nitric acid (DNO_3) to form a nitrate solution. The DNO_3 was chosen due to its low and flat neutron total cross section in the energy region of interest. The nitrate solution was then diluted with heavy water (D_2O) until the desired concentration of ^{236}U was achieved for each sample.

Each liquid sample was contained inside of a spectroscopic quartz cell (Starna, 2014). Quartz was chosen over Pyrex or glass as it contains no boron. Additionally, quartz is inert to most chemicals. The cell characteristics are shown in Table 1. The cells were slightly modified by replacing the original Teflon stoppers with glass stoppers to prevent leakage. Additionally, the stem was covered with paraffin wax to seal the glass stopper in place, thus reducing the chance for leakage. Finally, a rubber finger cut was placed over the paraffin and stem as a tertiary seal.

A 10 mm length of D_2O has a neutron transmission of approximately 0.67 that does not excessively attenuate the incident neutron beam. The diameter of the quartz cell was a third larger than the diameter of collimated neutron beam, eliminating any concern for the neutron beam impinging upon the inner diameter walls of the cell.

Mass spectrometry was performed at ORNL to determine the sample compositions. The atom percent of each isotope is shown in Table 2. The ^{236}U concentration was quoted with a 1% uncertainty in the mass spectroscopy analysis.

The number density for each isotope in the liquid samples is presented for the thick (Table 3), medium (Table 4), and thin (Table 5) liquid samples.

2.2. Experimental setup

The RPI LINAC produces a high energy pulsed neutron source (up to 60 MeV) by accelerating electrons and colliding them with a tantalum neutron production target, generating bremsstrahlung radiation and photoneutrons. The time-of-flight (TOF) technique was used to measure the neutron counting rate as a function of neutron flight time.

The neutron producing target used in this experiment is known as the Enhanced Thermal Target (ETT) (Danon, 1990). This target was selected to optimize the number of neutrons interacting in the ^{236}U resonance at 5.467 eV, thereby reducing the run time required to minimize statistical uncertainty on the collected data and improve the signal-to-background ratio.

Evacuated drift tubes were employed for a majority of the neutron flight path to the detector in order to reduce air scattering. The neutron beam was collimated to a 3.49 cm (1.375 inch) diameter beam at the sample location.

The neutron transmission experiment was performed at the 15 m flight station. The neutron detector is a 7.62 cm (3 in.) diameter, 0.3 cm thick NE-905 loaded scintillator glass (6.6% lithium, enriched in ^{6}Li to 95%) and optically coupled to a photomultiplier tube. This detector was placed directly in the beam 14.973 ± 0.005 m from the neutron production target. This detector was housed inside of a 10.16 cm (4.0 inch) thick lead shield to

Table 1
Quartz cell specifications. The cell length and inner diameter were quoted (Starna, 2014) to have an uncertainty of 0.01 mm and 0.02 mm, respectively.

Cat. No.	Length	Inner diameter	Volume
35-Q-10	10 ± 0.01 mm	47 ± 0.02 mm	17 ml