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Evaluating a Field-Portable Neutron Resonance Capture Analysis System for Safeguards Applications

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ABSTRACT: Technological strides in the thorium fuel cycle suggest its possible commercialization for nuclear power use within the next ten years. While thorium shares many aspects with the uranium and plutonium fuel cycles, it introduces the requirement for the non-destructive assay (NDA) of ^{232}Th and ^{238}U in varied chemical and physical arrangements. We describe the experimental evaluation of a portable Neutron Resonance Capture Analysis (NRCA) system, sensitive to materials with resonances occurring in the epithermal (1-100 eV) range. NRCA can determine isotopic sensitivity through neutron time-of-flight correlation with unique isotopic resonances, using moderated neutrons from a compact deuterium-tritium neutron generator. The technique, typically performed at beam lines with lengths in excess of 10 meters, is evaluated in a novel miniaturized configuration. This work seeks to determine the feasibility of NRCA for practical in-field isotopic characterization; we will present results from simulation and from experiments on single-element targets. NRCA, when used in concert with a portable Neutron Resonance Transmission Analysis system currently in development has the potential to rapidly and non-destructively quantify and characterize isotopes of interest in support of safeguards material accountancy.

INTRODUCTION:

Neutron Resonance Capture Analysis (NRCA) is a nondestructive sample analysis technique which leverages the unique neutron resonance structure in certain isotopes to infer the composition of an object under test. It requires no sample preparation and has the potential to analyze objects of non-uniform composition and structure. NRCA is widely used for radiative capture cross section data determination. It has also been successful in determining isotopic fractional weight composition of several archeological objects such as religious jewelry [1] and ceremonial swords [2]. The technique has also been proposed by Koizumi, et al. [3] as a method to characterized melted fuel debris from the Fukushima Daiichi reactor. In this methodology, NRCA is used to constrain and inform data from Neutron Resonance Transmission Analysis (NRTA) in determining the content of debris which had been contaminated with boron-10 when the core melted.[1] It is the versatility of this technique and its complementary nature to NRTA that drives this line of investigation.

Our research group has successfully demonstrated the viability of a compact NRTA system for the identification of isotopes with neutron resonances in the epithermal range [4]. This

[1] In Koizumi et al., the dual analysis method is called Neutron Resonance Densitometry (NRD) and relies on the energy of NRCA γ-ray spectra to determine boron-10 content, not time of flight for materials with epithermal resonances, as we discuss here.

compact setup, based on a pulsed deuterium-tritium (DT) generator has shown a 1-eV material identification resolution for resonances up to 10 eV and a 5-eV resolution for the 10-30 eV resonance range. NRTA uses neutron time-of-flight (TOF) transmission spectra to correlate the magnitude and energy of attenuation features with unique resonance patterns in an isotope. Likewise, the neutron resonance structure forms the basis for NRCA, which correlates neutron TOF with γ-rays emitted by the (n, γ) capture reaction (and possibly γ-ray energy) to infer material composition. Schillebeck et al., note that due to methodological differences, NRTA will provide better accuracy than NRCA [5], however the information gained from both techniques maybe used to enhance and constrain one another, particularly in the case of neutron-absorbing contaminants, materials with overlapping resonances, and objects with complex geometries. We are investigating the complementary application of NRTA and NRCA in the use case of thorium safeguards. This paper discusses the initial step, which is to determine the viability of NRCA to identify single element materials in the compact DT-based format.

BACKGROUND:

Specific isotopes experience a local peak in the magnitude of neutron interaction probability, or a resonance, based on the energy of the incident neutron. A resonance, which originates due to the excitation of states in the compound neutron-nucleus system, can be parameterized by the sum of the partial widths for each interaction type possible (e.g. fission, scatter, radiative capture), the energy of the neutron, and other characteristics of the compound system. The relative structures of these energy-dependent resonances are isotopically unique, occurring in the epithermal region (1-100 eV) for many medium and heavy elements. For neutron interactions occurring via the radiative capture partial width, the compound nucleus exists in an excited state with *Q* equal to the neutron separation energy. This energy is emitted via a cascade of γ-rays with energies corresponding to the discretized energy levels of the final nucleus. For the heavier elements with complex level structure there are numerous (hundreds or thousands) of γ-ray energies possible, though some of the more likely transitions manifest as peaks in a pulse height spectrum on a highly sensitive counting system [6].

The increased γ-ray flux detected when neutrons of resonance energies are incident on a target can be used to reconstruct a resonance profile and infer material composition—the foundational concept of NRCA. By using a pulsed white neutron source at a set distance, *L*, from a target and the arrival time, *T,* the neutron energy, E_n, can be calculated in eV by

$$E_n = \left(72.298 \frac{L}{T - T_o}\right)^2, \quad (1)$$

where 72.298 has units of $\sqrt{eV}\mu\mathrm{s/m}$, distance has units of m, and time has units of μs. T_o is the latency time for the 14.1 MeV neutron pulse peak based on the DT generator's zero-time trigger signal. NRCA's key observable is the per unit time capture γ-ray yield, Y_i the timing and magnitude of which can be correlated to $\sigma_{\gamma,i}$, the capture cross section of nuclide *i*, and n_i, the number of atoms per unit area of nuclide *i* (along the direction of travel of the neutron beam). The product of the cross section and atomic areal density is modified by a self-shielding factor, *F*, which accounts for the attenuation of the neutron beam based on the peak cross section of the resonance being examined such that:

$$Y_i = n_i \sigma_{\gamma,i} F. \quad (2)$$

Reference [5] contains a comprehensive discussion of the correlation of the theoretical capture yield, Y_i to the experimental yield via the detector system response and a series of correction factors. For the first phase of this research, we are evaluating the time resolution of the NRCA setup, with the magnitude of the capture yield to be discussed in later work.

FEASIBILITY STUDIES:

Experimental design:

The experiments for this work were conducted in the Vault Laboratory for Nuclear Science at MIT using a P353 Thermo Fisher Scientific DT generator (nominal output 3.5 x 10^8 neutrons per second). The neutron generator is operated at a pulse rate of 5 kHz and 3.1% duty cycle with a nominal pulse width of 3.5 μs. Due to the generator's operational constraints, the actual width of the pulse is $\sigma_{T_0} \approx 0.7\ \mu s$. The initial experimental design consisted of an 10.0 cm HDPE moderator cuff positioned radially around the DT tube and centered on the target plane, as shown in Fig. 1. Subsequent design improvements to the setup for signal optimization are discussed in the next section. The TOF path is varied between 1.70 and 2.50 meters, measured from the front face of the moderator to the NRCA target. MCNP simulations show a neutron moderation distance of 2.0 ± 0.1 cm within the moderator, which is added to the measured TOF path to result in the effective distance which is used for TOF-neutron energy calculation.

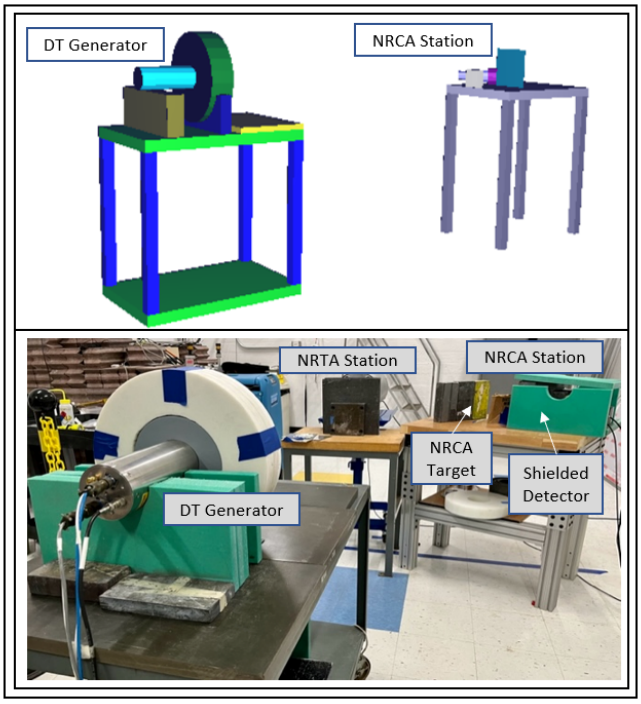

Fig. 1. **Top**: MCNP geometry model of the DT generator and moderator assembly two meters from a NRCA experiment station with detector and large silver target. **Bottom:** A photograph of NRTA and NRCA performed simultaneously on separate targets. Additional lead shielding is in place between the experiments to mitigate the boron neutron capture gammas from the NRTA boron carbide shielding.

The target materials used in the experiments are thin foils: the natural silver target is 0.3mm thick and 10 x 10 cm in area, and the natural tungsten target is 3.5mm and 8 x 8 cm in area. These metals were selected due to their large-magnitude epithermal resonances at 5.2 eV (^{109}Ag) and 18.8 eV (^{186}W). The detector is built in-house, consisting of a photomultiplier tube and a 2 x 2" volume of EJ-315 deuterated benzene scintillation fluid (C_6D_6). EJ-315 was selected after an experimental detector study due to its low neutron capture cross section, high time resolution, and ability to filter neutron from gamma counts via pulse-shape discrimination. The pulse start signal and the detector signal are read into separate channels of a 14-bit, 500 MS/s

CAEN DT5730 digitizer and collected via CAEN's CoMPASS acquisition software. Data analysis is performed in ROOT [7] and SciPy [8].

RESULTS AND DISCUSSION:

Signal optimization:
Initial MCNP and experimental runs underscored the importance of background management, primarily reducing the thermal neutron-induced γ-rays from the moderator, room, and non-target experimental setup. MCNP simulation indicated γ-ray captures from the HDPE moderator contributed 25% of the flux in an unshielded detector, with another ~50% arising from neutron captures in the concrete walls of the vault. In order to eliminate a portion of the gamma-capture background, a 7.5% lithium-polyethylene moderator was designed and manufactured for the setup. The addition of this moderator reduced the off-pulse, off-resonance γ-ray background by 10-15% (experimentally determined), increasing the prominence of the resonance-capture peak for the silver target by 13% (relative to a scattering target without a neutron capture resonance). Thermal neutron backgrounds from a previous pulse, which can capture both in the target and on experimental materials, are reduced by a relatively long (200 μs) DT generator pulse period.

Further background management work has included improving detector γ-ray and neutron shielding, with the best results gained from the detector centered in a lead and HDPE shield. The addition of a lead multiplier cuff inside the Li-HDPE moderator (visible in the lower diagram in Fig. 1) to leverage (n,2n) reactions accessible by 14.1 MeV DT neutrons results in a 40% increase in neutron production and 10% increase in counts in the ^{109}Ag 5.2 eV resonance peak, relative to a scattering fiducial target.

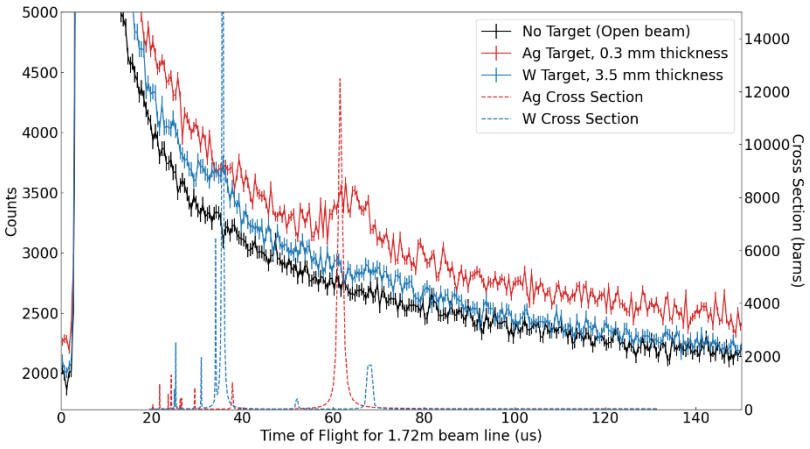

Fig. 2. Time-of-flight spectra collected using a pulsed DT generator and a 2x2" EJ-315 liquid scintillator. Each run was 30 min long with a 3.1% duty cycle, 5 kHz operational frequency, 110 kV accelerator voltage and 40 uA beam current. Cross section data from ENDF/B database.

ToF and Energy Reconstruction:
Using Eqn. 1, neutron energy, and therefore resonance structure, can be inferred from the time at which the γ-ray flux increases above the background, while the magnitude can be correlated with isotopic quantity in a calibrated system (to be discussed in future work). Fig. 3 presents the experimental results of a setup indicated in Fig. 1 (the path length is 1.72 m for these runs), alongside the ENDF/B evaluated cross section data plotted for the fixed-path time of flight for a given neutron energy. Here the TOF γ-ray counts for a 0.3 mm silver target, a 3.5mm tungsten target and an open beam run. Resonance structures for silver are more

pronounced than for tungsten, an effect which MCNP analysis suggests is due to target thickness, however a thinner set of foils is on order for future experimentation.

The time resolution of the current technique is poor, with the 5.2 eV silver resonance peak having a mean of 63.5 ± 6.2 μs. Uncertainty in time resolution in this miniaturized setup come from the finite pulse width of the DT generator, the time resolution of the detector, the moderation time of the 14.1 MeV neutron in the moderator, and the additional moderating effects of experimental components. The scattering effects of a tungsten target in the beam as opposed to an open beam run are evident in the sections without cross section resonances. While these targets are distinguishable in this configuration, more work in background and neutron scatter reduction are require to resolve the more finely spaced resonances of thorium and uranium.

FUTURE WORK:

Background mitigation work will continue, including the use of a cadmium 'black' filter in the target vicinity to decrease thermal neutrons capturing on target materials. Neutron flight path collimators are also being investigated to reduce the effects of room return fast neutrons that re-enter the moderator and arrive at off-times. One of the goals we seek is the reduction of γ-ray background in the detector via shielding while minimizing material on which neutrons may down scatter. Finally, mixed-sample uranium and thorium studies will determine the technique's sensitivity to various ratios of these materials, pursuant to the safeguards use case.

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