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A Kinematically Complete, Interdisciplinary, and Co-Institutional Measurement of the $^{19}F(\alpha,n)$ Cross-section for Nuclear Safeguards Science

OR13-F(a,n)-PD2LB

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1. INTRODUCTION

The goal of this proposal is to enable neutron detection for precision Non-Destructive Assays (NDAs) of actinide-fluoride samples. Neutrons are continuously generated from a UF_x matrix in a container or sample as a result of the interaction of alpha particles from uranium-decay α particles with fluorine nuclei in the matrix. Neutrons from ^{19}F(α,n)^{22}Na were once considered a poorly characterized background for assays of UF_x samples via ^{238}U spontaneous fission neutron detection [SMI2010B]. However, the yield of decay-α-driven neutrons is critical for ^{234,235}U LEU and HEU assays, as it can used to determine both the total amount of uranium and the enrichment [BER2010]. This approach can be extremely valuable in a variety of safeguard applications, such as cylinder monitoring in underground uranium storage facilities, nuclear criticality safety studies, nuclear materials accounting, and other nonproliferation applications.

The success of neutron-based assays critically depends on an accurate knowledge of the cross section of the (α,n) reaction that generates the neutrons. The 40% uncertainty in the ^{19}F(α,n)^{22}Na cross section currently limits the precision of such assays, and has been identified as a key factor in preventing accurate enrichment determinations [CRO2003]. The need for higher quality cross section data for (α,n) reactions has been a recurring conclusion in reviews of the nuclear data needs to support safeguards.

The overarching goal of this project is to enable neutron detection to be used for *precision* Non-Destructive Assays (NDAs) of actinide-fluoride samples. This will significantly advance safeguards verification at existing declared facilities, nuclear materials accounting, process control, nuclear criticality safety monitoring, and a variety of other nonproliferation applications. To reach this goal, Idaho National Laboratory (INL), in partnership with Oak Ridge National Laboratory (ORNL), Rutgers University (RU), and the University of Notre Dame (UND), will focus on three specific items: (1) making a precision (better than 10 %) determination of the absolute cross section of the ^{19}F(α,n)^{22}Na reaction as a function of energy; (2) determining the spectrum of neutrons and γ-rays emitted from ^{19}F(α,n)^{22}Na over an energy range pertinent to NDA; and (3) performing simulations with this new cross section to extract the neutron yield (neutrons/gram/second) and resulting neutron- and gamma ray-spectra when α particles interact with fluorine nuclei in actinide samples, to aid in the design and reduce uncertainty of future NDA measurements and simulations.

Our plan is to use the state-of-the-art neutron detector array VANDLE (Versatile Array of Neutron Detectors at Low Energy) the measure the reaction cross section with both fluorine and α beams. This approach, using one detector system for complementary measurements of the reaction with two beams, will completely characterize the ^{19}F(α,n)^{22}Na reaction and overcome difficulties with previous measurements which seriously limit the precision of neutron-based assays. The first experiments with an alpha-particle beam began in early 2013 at the University of Notre Dame (UND), and the second campaign with a ^{19}F beam is scheduled to start in early 2014 at the Holifield Radioactive Ion Beam Facility at ORNL. The experiments at Notre Dame cover a wide range of alpha-particle energies from the FN tandem at UND, while the experiments at ORNL will use a helium-gas target and a ^{19}F beam to improve the precision of the measurements over the most crucial 20% of the full range covered in the UND experiments. Therefore, the first campaign provides a high-statistics, relatively wide overview of the cross-section that will be used as a guide to identify regions that could most benefit from the improved precision of the second campaign. Further, by utilizing the same detector system at two different world-class facilities we expect to be able to reduce systematic uncertainties that would otherwise be difficult to identify and subsequently quantify.

First experiment: Alpha-particle beam at Notre Dame

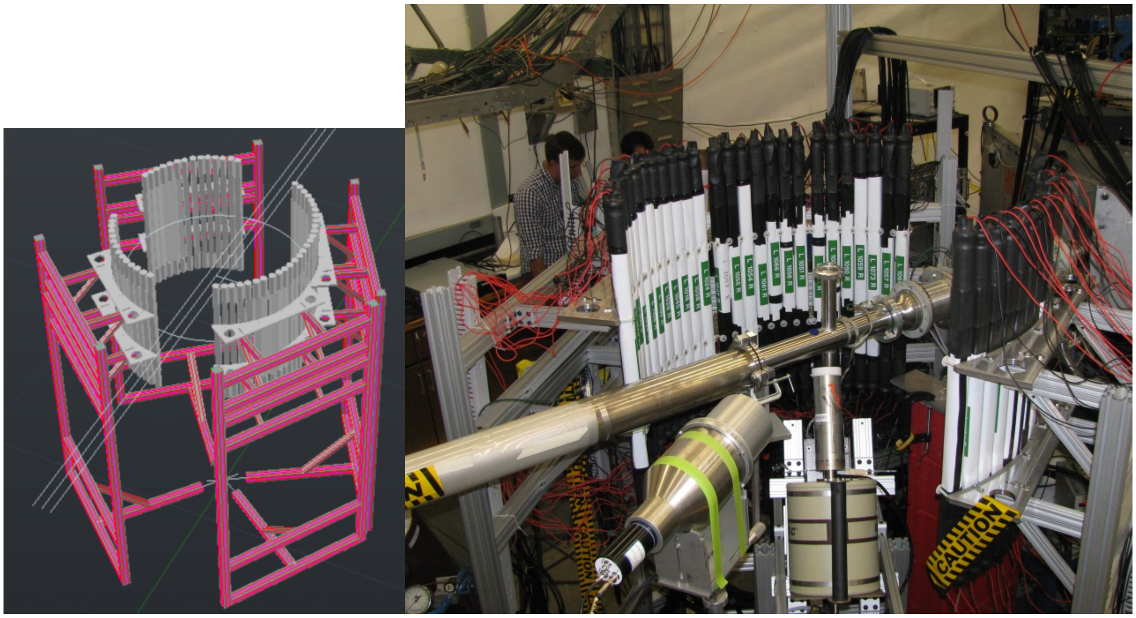

Figure 1. Experimental setup at Notre Dame University.The left panel, drafted by our Rutgers summer student Craig Reingold, shows our original design with a frame sufficient to mount 56 small VANDLE modules surrounding the target. A photograph of the completed setup at Notre Dame is shown to the right with 42 VANDLE modules supplemented with a HPGe gamma-ray detector and a liquid neutron detector from University of Michigan collaborators Michael Febbraro and Prof. Fred Becchetti.

Our first task in this measurement was to choose the target containing fluorine that would be sufficiently thin for good energy resolution, contain enough fluorine for a good yield, and have other materials that would not generate serious neutron backgrounds. In the end, a thin lanthanum-fluoride (LaF_3) film evaporated onto a thin gold foil was chosen. The lanthanum and gold in the targets have such a high proton number that the nuclei repel the alpha-particles in the beam so that they induce no nuclear reactions. We performed numerous measurements during our experiment with a gold-only target to confirm that contributes no significant background. In addition to testing target materials, we also improved the pulsed time structure of the alpha-particle beam from the FN tandem at Notre Dame. Precision beam pulsing is crucial to achieve high energy resolution for the neutrons, since VANDLE determines neutron energies from their time-of-flight between the target (indicated by the timing of the beam pulse) and the detector (indicated by the timing of the detector signal). The timing width of the beam was eventually improved to be less than 3 ns.

42 VANDLE detector modules were mounted in a cylindrical arrangement around the target position (as seen in Figure 1) to enable the placement of a gamma-ray detector made of High-Purity Germanium (HPGe). This portion of the experiment ran over the course of a few weeks in July and August. A detailed measurement of the thickness and composition of the targets was completed following the August run. In addition to executing the experiments at Notre Dame, there was an extensive collaborative effort of the participants beginning in Spring 2013 to plan and coordinate the runs. Over a dozen graduate students from UND, the University of Tennessee, and Rutgers University worked on the experiment, doing a variety of tasks including running the FN tandem accelerator, running the VANDLE data acquisition (DAQ) system, and monitoring the data as it was collected.

2. PRELIMINARY RESULTS: TWO NEW RESONANCES

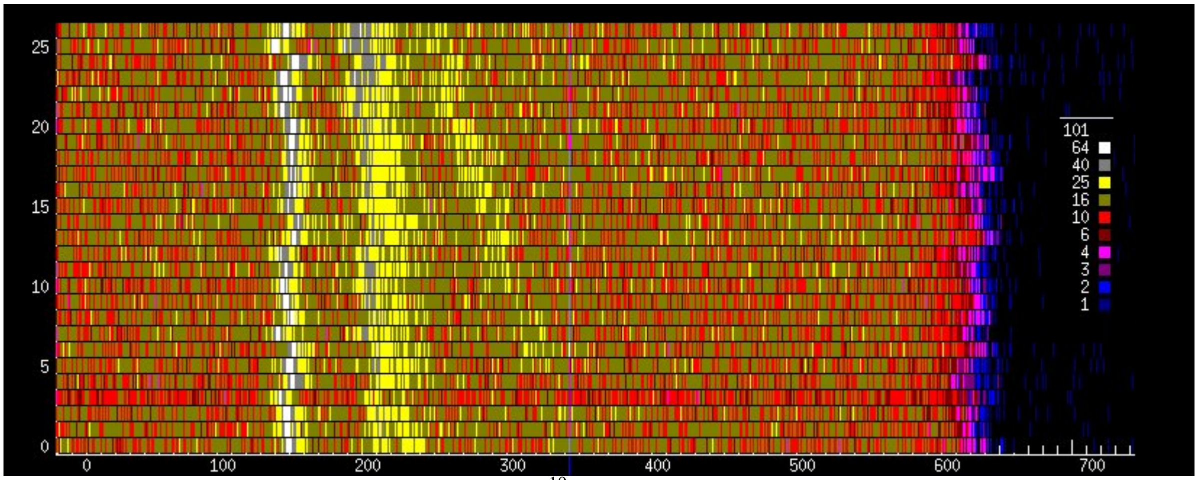

Figure 2. Preliminary time-of-flight $^{19}F(\alpha,n)$ data from VANDLE at Notre Dame.
This figure is a snapshot of the online spectra that displays a vertical stacking of the time-of-flight signals (TOF in 0.5 ns per bin) along the horizontal axis from the first 27 VANDLE detector bars. This data was collected at one of the many alpha beam energies measured in our experiment. The position of the VANDLE modules contributing to this plot roughly correspond to angles from near 160° (module 0) to near 30° relative to the beam (module 26).

A snapshot of the online time-of-flight data for the first 26 VANDLE detector modules is displayed in Figure 2. This plot contains three prominent features. First, the gamma-flash from the alpha-particle beam striking the target is evident near channel 150. At higher channels (near 200), the first (α,n) neutrons to the ground state of ^{22}Na are evident. At even higher channels (225 – 300), (α,n) neutrons populating higher excited states in ^{22}Na are evident. The kinematics of the reaction causes these slower neutrons to experience a more dramatic shift in TOF relative to the faster neutrons that populate the ^{22}Na ground state. The exact spectrum and relative intensities to the different neutron peaks vary from one energy to the next. The orange-red background events are mostly due to beam induced background and random room background of gamma rays. Resolution of the time-of-flight spectra will improve when the gains of all the modules are calibrated. A careful analysis will begin in Fall 2013 to account for all the possible neutron channels, and allow us to determine the cross section as a function of energy as well as the neutron energy spectrum induced at each alpha-particle energy.

The preliminary relative yield vs. alpha energy curve from all the data taken so far is displayed in Figure 3. The general trend of the yield curve follows that of previously published data [WRE2000 & BAI1979] that either did not measure the cross section at α-decay energies or used a thick target that reduced the resolution of their results. Our measurement will cover the full range between 3 and 8 MeV alpha-particles with moderate resolution and will have very fine resolution for the crucial regions. Our data already discern two previously unobserved resonances indicated by the peaks near 5.4 and 7.9 MeV. The yield at energies near these resonances, and at other energy regions of interest, will be repeated with greater precision during the second half of the project when we measure this reaction at ORNL with a ^{19}F beam.

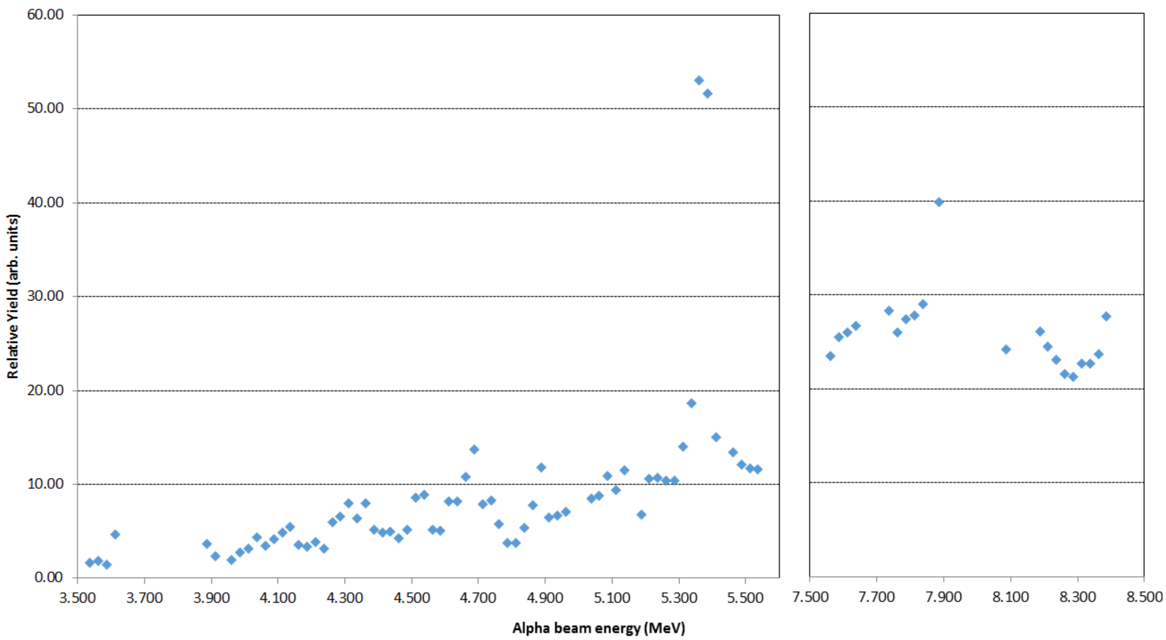

Figure 3. Preliminary relative yield from our measurement of $^{19}F(\alpha,n)$ at Notre Dame University in the summer of 2013. This relative yield data is currently being converted to an absolute cross section determination. During our upcoming fall experimental run at Notre Dame University, we will fill in the noticeable energy gaps in this yield spectrum with additional measurements. This yield curve contains two previously unobserved resonances indicated by the peaks near 5.4 and 7.9 MeV.

3. PATH FORWARD: ANALYSIS AND THE 19F-BEAM EXPERIMENT

Soon after the completion of the August 2013 run, we produced the preliminary relative yield vs. energy curve (Figure 3) illustrating the energy coverage of the Notre Dame experiment. This relative yield curve will be analyzed using simulations that determine the absolute efficiency of the VANDLE setup. These simulations, utilizing the MCNP and GEANT4 codes packages, will include yield determinations resulting from a detailed nuclear reaction calculation. Our measured data has a statistical uncertainty of less than 1% at each energy; the total uncertainty at each energy will be dominated by uncertainties in detector efficiencies, beam current determinations, and other systematic uncertainties.

The final run at Notre Dame is scheduled for the second week of October 2013. During this experiment, we will measure the neutron yield from a ^{13}C target; the well-known $^{13}C(\alpha,n)$ cross section will enable us to reduce our systematic uncertainties and will provide a benchmark for our simulations. Following this run, the VANDLE equipment will be packed up and returned to ORNL for use with our ^{19}F beam measurements. Preparations of the experimental setup for the ORNL measurements are already underway.

4. PRESENTATIONS AND PUBLICATIONS

Preliminary results of the summer experimental campaign were presented in a poster by ORNL summer student Craig Reingold from Rutgers University. He will also present an updated version at the fall meeting of the American Physical Society Division of Nuclear Physics. Project team member Dr. William Peters presented a physics colloquium February 25, 2013 at Ohio University and a physics colloquium at the University of Notre Dame March 20, 2013 - "How to Get a Better Non-Destructive Assay; Enrichment Detection with Neutrons" - covering the motivation and experimental approach for this project. Dr. William Peters also presented a poster on the project at the UITI meeting in Lansing, June 5 and is completing an instrumentation publication on the unique capabilities of VANDLE.

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