Project No. 13-5485

Improved Delayed-Neutron Spectroscopy Using Trapped Ions

Fuel Cycle Research and Development

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The neutrons emitted following the β decay of fission fragments (known as delayed neutrons because they are emitted after fission on a timescale of the β-decay half-lives) play a crucial role in reactor performance and control. Reviews of delayed-neutron properties highlight the need for high-quality data for a wide variety of delayed-neutron emitters to better understand the time-dependence and energy spectrum of the neutrons as these properties are essential for a detailed understanding of reactor kinetics needed for reactor safety and to understand the behavior of these reactors under various accident and component-failure scenarios. For fast breeder reactors, criticality calculations require accurate delayed-neutron energy spectra and approximations that are acceptable for light-water reactors such as assuming the delayed-neutron and fission-neutron energy spectra are identical are not acceptable and improved β-delayed neutron data is needed for safety and accident analyses for these reactors. With improved nuclear data, the delayed-neutrons flux and energy spectrum could be calculated from the contributions from individual isotopes and therefore could be accurately modeled for any fuel-cycle concept, actinide mix, or irradiation history. High-quality β-delayed neutron measurements are also critical to constrain modern nuclear-structure calculations and empirical models that predict the decay properties for nuclei for which no data exists and improve the accuracy and flexibility of the existing empirical descriptions of delayed neutrons from fission such as the six-group representation.

Trapped radioactive ions suspended in vacuum allow an innovative new way to measure delayed-neutron properties by inferring the neutron energy from the large momentum kick it imparts. When a radioactive ion decays in the trap, the recoil-daughter nucleus emerges from the ~1-mm^3 trap volume without scattering and the recoil energy can be measured. The energy of the emitted neutron can be easily and precisely reconstructed from this recoil by conservation of energy and momentum. This ability to measure the nuclear recoil unperturbed enables a novel way to perform delayed-neutron spectroscopy with high efficiency, few backgrounds, and improved energy resolution – avoiding all the well-known challenges associated with detecting neutrons.

The research performed under this project focused on collecting high-quality data for a variety of different fission fragments and demonstrating an advanced measurement technique that can be applied to improve the quality of delayed-neutron data for all the fission products. With the beams now available from the Californium Rare Ion Breeder Upgrade (CARIBU) facility at Argonne National Laboratory, the nation’s premier fission-fragment facility, and an optimized ion trap with a highly-efficient detector array, high-quality measurements of the delayed-neutron branching ratios and energy spectra for these isotopes were performed. By using fast plastic scintillator detectors for β detection and MCP detectors for recoil-ion detection, the recoil energy was determined simply from the time-of-flight of and the distance travelled by the daughter nucleus. The high β-ion rates yielded the statistics for a 1% neutron-emission branching ratio and allowed detailed studies of systematic effects and the neutron energy spectrum.

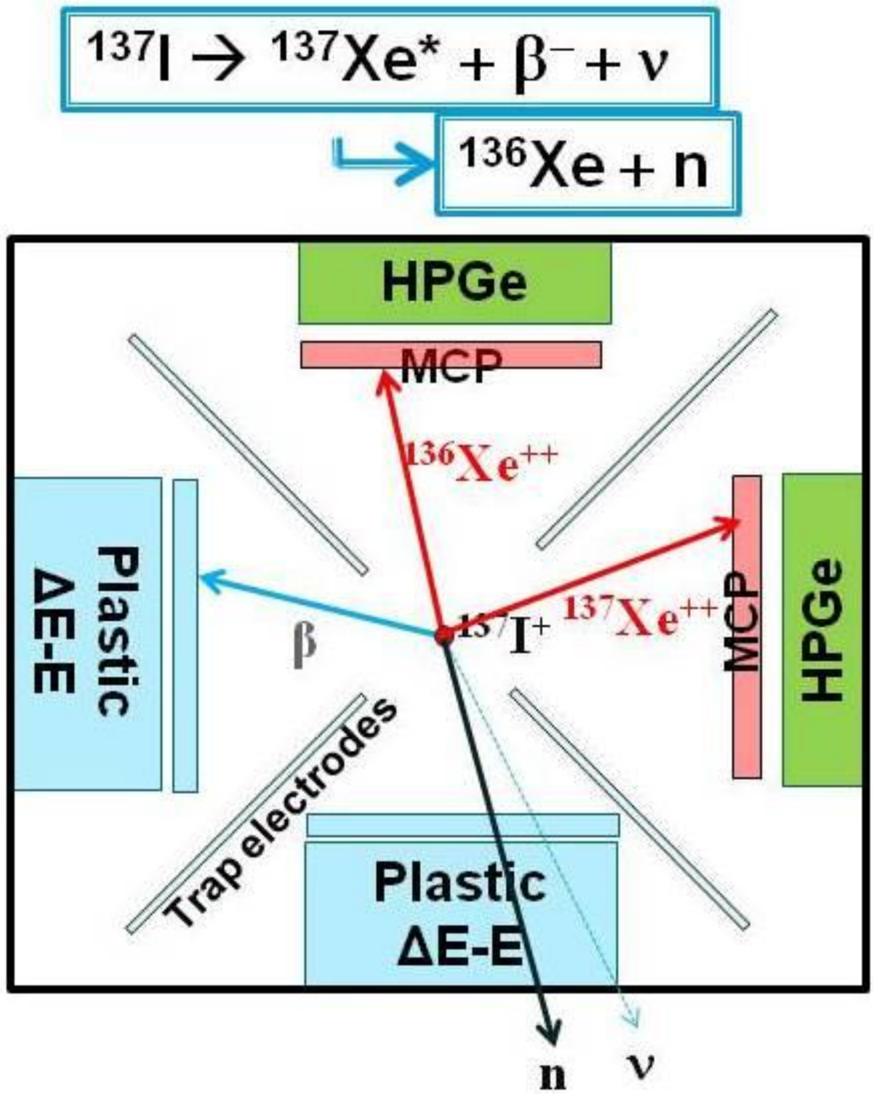

Figure 1. Experimental setup for beta-delayed neutron studies using trapped ions.

The basic concept of our experiments is illustrated in Figure 1. Neutron-rich isotopes are loaded into a radiofrequency linear quadrupole trap (the β-decay Paul Trap, surrounded by two plastic scintillators in a ΔE-E configuration for detecting β particles, two microchannel plate (MCP) detectors for detecting recoiling ions, and two high-purity germanium (HPGe) detectors for γ-ray spectroscopy. The time elapsed between the β particle detection and the recoil-ion detection determines the time-of-flight (TOF) of the ion, and hence its recoil energy. The neutron energy is obtained from the nuclear recoil using conservation of momentum. In the case of β decay without neutron emission, the ion's recoil energy is smaller, and hence its time-of-flight is longer, allowing separation from the higher energy recoils characteristic of neutron emission.

We conducted two experimental campaigns using a low-energy beamline at the CAlifornium Rare Isotope Breeder Upgrade (CARIBU) facility at Argonne National Laboratory. Beta-delayed neutron data were collected from $^{137\text{-}140}$I, ^{144,145}Cs, and $^{134\text{-}136}$Sb, with statistics considerably higher than that which was achieved in our earlier work. As part of the β-delayed neutron emission measurements, the β decays of ^{134,134m}Sb were also investigated to calibrate the intrinsic efficiency of the micro channel plate detectors to low-energy (5-20 keV) recoil ions. This is an essential part of the β-delayed neutron analysis as it sets the overall normalization for the branching ratios determined using the recoil-ion technique. We also conducted beta spectral shape measurements from the decays of ^{92}Rb, ^{96}Y, and ^{136}Sb. The results of these measurements will improve the interpretation of the β-delayed neutron emission data for the ^{137,138,140}I, ^{144,145}Cs, and ^{135,136}Sb data collected at CARIBU. The results of these experiments formed the basis of the PhD thesis of Aga Czeszumskza, a graduate student from UC Berkeley. We have published a number of papers describing our measurements. Two papers describing the final results for the beta delayed neutron emission probabilities and energy spectra are nearing completion and will be submitted for publication in the near future.

We developed three techniques to extract the branching ratios for beta delayed neutron emission from our data. As mentioned above, the beta decays in which a neutron is emitted produce the fastest recoil ions. The number of these events appears in the numerator of the equation shown in Figure 2. The total number of beta decays (which appears in the denominator of the equation) can be determined in three different ways. We can simply count the total number of all betas detected, or we can use associated beta-delayed gamma rays, or we can count the number of slower recoil ions. Each of these methods has its advantages and disadvantages as described in the figure. The results of our measurements and analysis for the beta-delayed neutron branching ratios for ^{137,138}I and ^{144,145}Cs are shown in Figure 3. Our results for the beta delayed neutron energy spectrum from ^{137}I are shown in Figure 4 and those for ^{138}I and ^{144,145}Cs are shown in Figure 5. Our results for both the branching ratios and energy spectra are in reasonably good agreement with previous measurements.

In this project we demonstrated that the technique of measuring nuclear recoils from the decays of trapped ions can be used to extract both beta delayed neutron branching ratios and energy spectra from a wide range of fission fragments. As higher intensities of fission fragment beams are made available at CARIBU and as improvements are made in detection capabilities, measurements of this type can be extended to nuclides even further from stability. Such data is important for designs of advanced nuclear reactors and for understanding the synthesis of heavy elements during the astrophysical r-process.

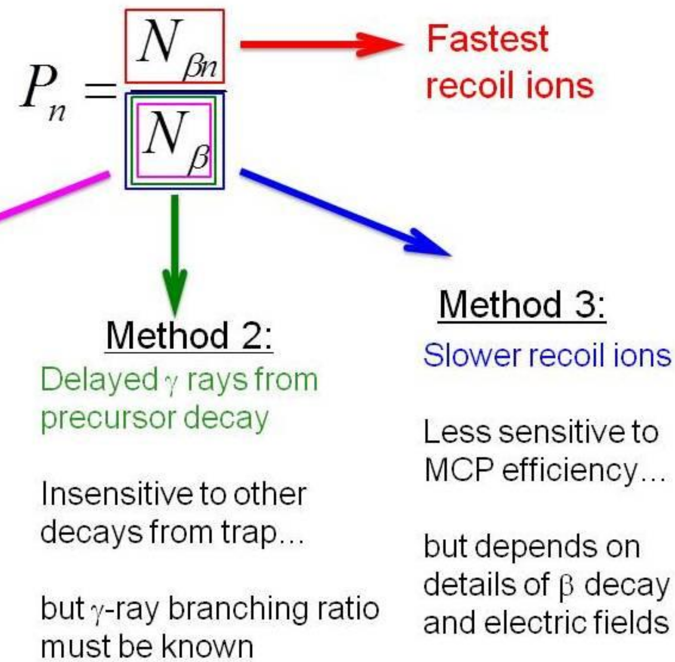

Figure 2. Methods we have used to determine beta-delayed neutron branching ratios.

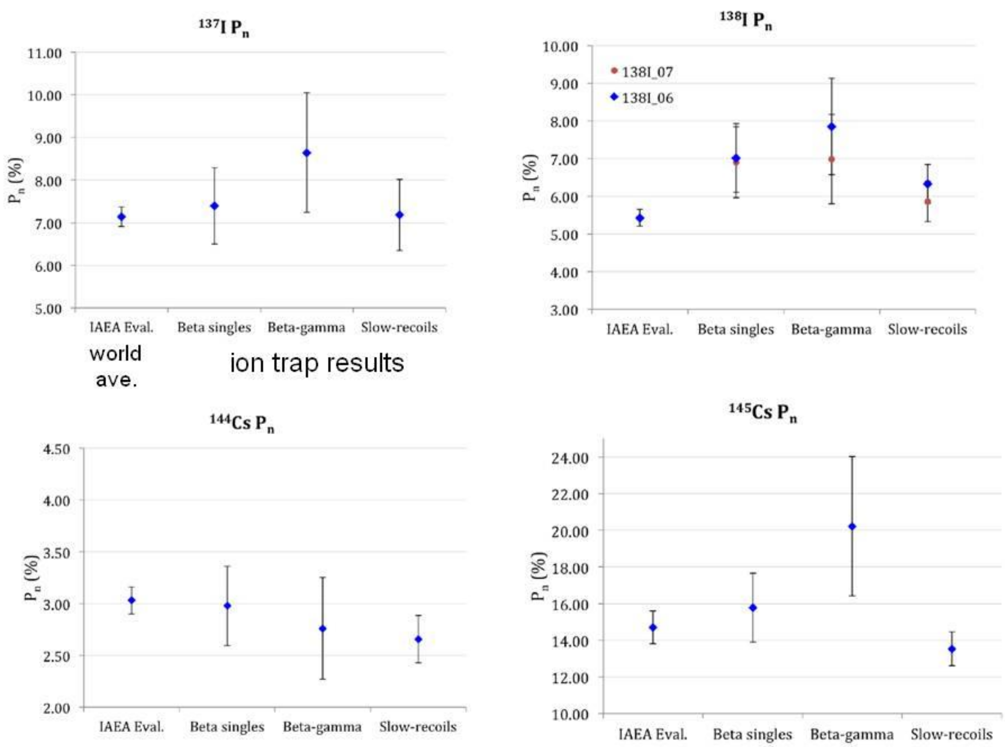

Figure 3. Our results for the beta delayed neutron branching ratios for ^{137,138}I and ^{144,145}Cs compared to IAEA evaluations.

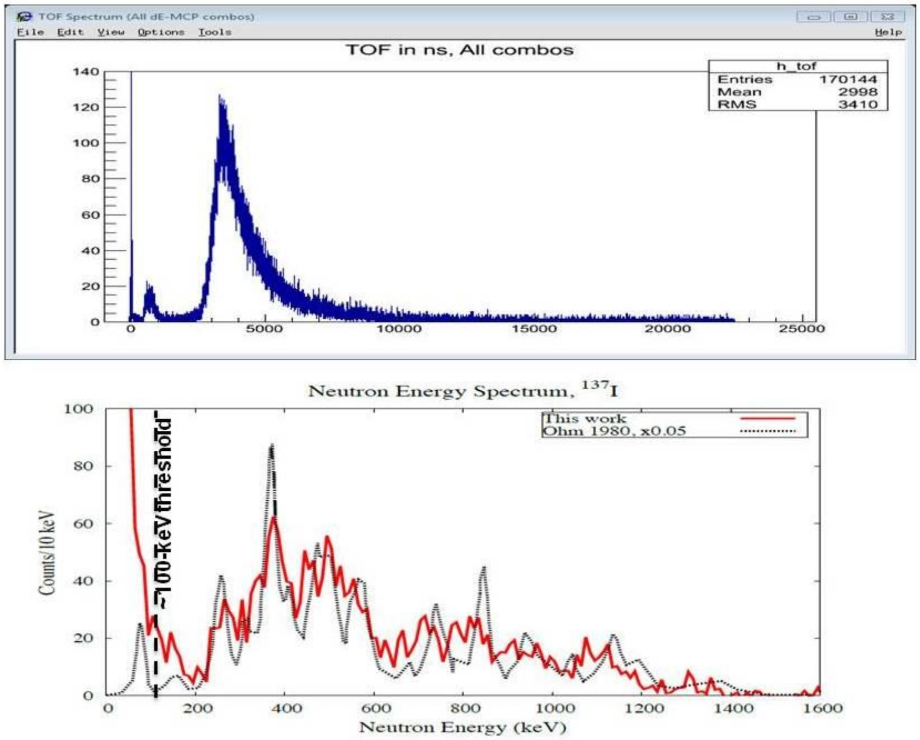

Figure 4. Top panel shows the recoil ion time-of-flight spectrum observed form the decay of ^{137}I. The large peak at long times is produced by beta decays in which no neutrons are emitted. The smaller peak at short times is attributed to beta delayed neutron emission. The lower panel shows the beta delayed neutron energy spectrum inferred from the fast recoil ions using conservation of energy and momentum.

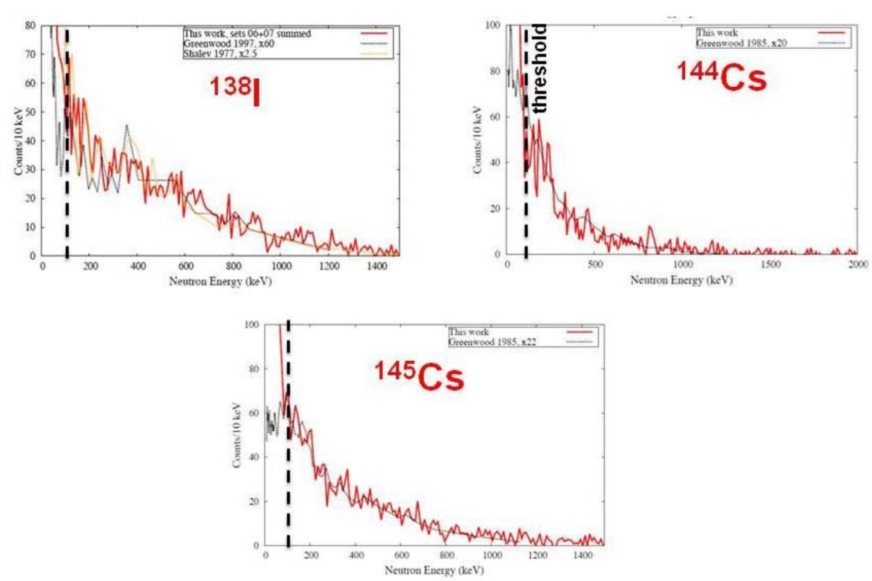

Figure 5. Beta delayed neutron energy spectra determined from decays of ^{138}I, and $^{144,145}Cs$.

Personnel

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