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Neutron Capture Measurements on Unstable Nuclei at LANSCE

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Although neutron capture by stable isotopes has been extensively measured, there are very few measurements on unstable isotopes. The intense neutron flux at the Manual Lujan Jr. Neutron Scattering Center at LANSCE enables us to measure capture on targets with masses of about 1 mg over the energy range from 1 eV to 100 keV. These measurements are important not only for understanding the basic physics, but also for calculations of stellar nucleosynthesis and Science-Based Stockpile Stewardship. Preliminary measurements on ^{169}Tm and ^{171}Tm have been made with deuterated benzene detectors. A new detector array at the Lujan center and a new radioactive isotope separator will combine to give Los Alamos a unique capability for making these measurements.

INTRODUCTION

Neutron capture is one of the first reactions studied in nuclear physics, and measurements have been made on virtually all stable targets. Most cross sections on unstable targets have been inferred from theoretical calculations. This is a difficult reaction to calculate, however, because details of the nuclear structure of the compound nucleus near the neutron binding energy are important. Evidence for this is the order of magnitude differences in thermal neutron capture cross sections for adjacent and near-adjacent nuclei.

In addition to fundamental scientific curiosity, several applied programs have a need for neutron capture cross sections in the keV region. Several facilities at Los Alamos combine to provide a unique capability to make these measurements. I will describe a new detector array being designed and built at the Los Alamos Neutron Science Center (LANSCE) to measure capture on small quantities of unstable and stable nuclei., on the order of a milligram, and present some preliminary measurements of capture on ^{171}Tm.

NEUTRON CAPTURE REVISITED

Defense Applications

One of the main drivers for this renewed interest in capture reactions is in understanding nuclear device performance. The extensive data associated with 50 years of nuclear testing is being systematically reviewed using modern analysis and computational techniques, with a goal of understanding nuclear weapons physics. One of the principal methods of diagnosing device performance has been through the use of radiochemical tracer isotopes. The high neutron density during an explosion can result in multiple reactions on a single nucleus, driving it far from stability. In order to calculate the observed isotopic yields, accurate cross sections are required for the nuclei involved, many of which are unstable.

s-Process Nucleosynthesis

A second area of interest is in studying the stellar synthesis of elements heavier than iron. This is believed to occur primarily through neutron capture and β decay. These reactions take place in a "r" (rapid) process that occurs in the explosive environment of supernovae, and in a "s" (slow) process of sequential capture along the valley of β stability that occurs in the "asymptotic giant branch" stage of evolution of low to medium mass stars, or in red giants. The overall mechanism of the s process appears to be understood, and solar and stellar abundances are in general well reproduced (1,2).

The flow of the s-process is illustrated in Fig. 1, for masses near A = 151. Of particular interest is the branching of the flow at unstable nuclei, where the competition between neutron capture and β decay can shed information on the stellar environment where the s process occurred. If the β decay rate in the hot stellar environment is the same as in our laboratory, the neutron density at the s-process site can be inferred from observed abundances and the neutron capture cross section. The most accurate abundances to use are those of nuclei that are formed only by the s-process.

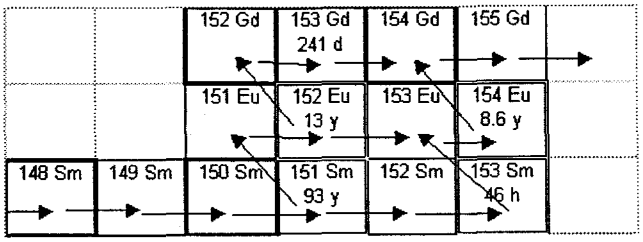

FIGURE 1. S-process flow near mass 151. The branching at ^{151}Sm is of particular interest. Heavy outlines indicate "s-only" isotopes, and double outlines indicate unstable isotopes.

Neutron densities and stellar temperatures extracted from branch-point analysis are given in Ref. 1. They appear quite consistent with a ^{22}Ne(α,n) neutron source at 30 keV and a neutron density of 4 x 10^8 cm^{-3}. However, this consistency is based on calculated values for neutron capture cross sections on unstable nuclei. A comparison of calculated (3) and measured (4) capture cross sections for the stable Sm isotopes shows that the even-even isotopes are calculated to 10% while the even-odd isotopes show poorer agreement. A minimum 25% uncertainty is often assumed for the calculated values of the capture cross section, and clearly more accurate measurements are needed.

Interestingly, although the measured parameters point to a 30 keV ^{22}Ne(α,n) neutron source for the s-process, detailed stellar models favor the ^{13}C(α,n) reaction, with an average energy of 10 keV, as the main neutron producing reaction driving the s process (1). Models predict that the s process occurs during a 20-year pulse of ^{13}C burning followed by a 1-year pulse of ^{22}Ne burning, which alters the observed abundances. Thus while cross sections must be measured over the entire 10 keV Maxwell velocity distribution characteristic of ^{13}C burning, the effect of this episode on the observed stellar abundances must be determined through detailed stellar models.

Transmutation

A third area of growing interest is field of accelerator-driven transmutation technology, in which neutrons produced by an accelerator-driven spallation source are used to transmute long-lived radioactive waste from reactors into shorter-lived, easier-to handle isotopes. A number of capture cross sections need to be accurately known, including capture on fission fragments such as ^{99}Tc and ^{135}Cs, and on lesser actinides such as ^{234}U and ^{242}Pu. Although some measurements do exist, it is likely that more experiments will be required.

LOS ALAMOS CAPABILITIES

The ability to make these measurements requires a confluence of capabilities that exists only at Los Alamos. The first of these is the intense neutron spallation source of the Manual Lujan Jr. Neutron Scattering Center (5). The high neutron fluence available in the critical keV region allows us to make direct measurements on samples with mass 1 milligram or less. Next is the ability to safely handle and produce radioactive targets. Los Alamos National Laboratory supplies many of the radiopharmaceutical isotopes distributed by the U.S. Department of Energy, and most of these are produced by at LANSCE by proton spallation on heavy targets. The Nuclear and Radiochemistry group at Los Alamos (CST-11) maintains hot cells for handling the production targets and chemically separating isotopes of interest.

In addition to chemical separation, a magnetic isotope separator, the Radioactive Species Isotope Separator (RSIS) has been specifically designed and built to separate highly radioactive isotopes. The RSIS will be housed entirely in a hot cell. Test operation of the separator has shown separation factors between adjacent isotopes of about 10^4. As an example, it will take about 1 to 3 weeks to produce 1 mg of ^{170}Tm, depending on the efficiency of the ion source, which ranges from about 10 to 30% for Tm.

Finally, a new detector array is being constructed. This will be discussed further below.

PRELIMINARY MEASUREMENTS

A preliminary experiment to measure neutron capture cross sections on a radioactive target used the isotope ^{171}Tm, which has a half-life of 1.9 yr. This isotope was chosen for several reasons. First, milligram quantities of isotopically pure ^{171}Tm were produced by reactor irradiation of ^{170}Er and chemical separation by high-performance liquid chromatography, without the use of an isotope separator. Second, the low-energy of the decays (97 keV β, 66 keV γ) presented the lowest radiological hazard of the possible targets. The target was elecroplated on a 12.7 μm Be foil which had a 70 nm layer of titanium vapor-deposited on the surface. This was covered by a second 12.7 μm Be foil.

The experiment was done on Flight Path 4 of the Lujan Center at LANSCE, which views the "high intensity" water moderator. The detectors were two C_6D_6 scintillators, each 5 inches diameter by 3 inches thick, mounted on an RCA 4522 phototube. The detectors were mounted inside a 30cm x 30 cm x 10 cm thick Pb shield inside a larger polyethylene house. The flight path was 8.08 m. The results of the measurement are shown in Fig. 2. The results are compared to calculations(6) made with the GNASH code, which underpredicts the data by a factor of 2 to 5. A target of stable ^{169}Tm was also measured, and the results agreed very well with the

GNASH calculations, which used parameters determined from an earlier measurement of capture on ^{169}Tm by Macklin (7).

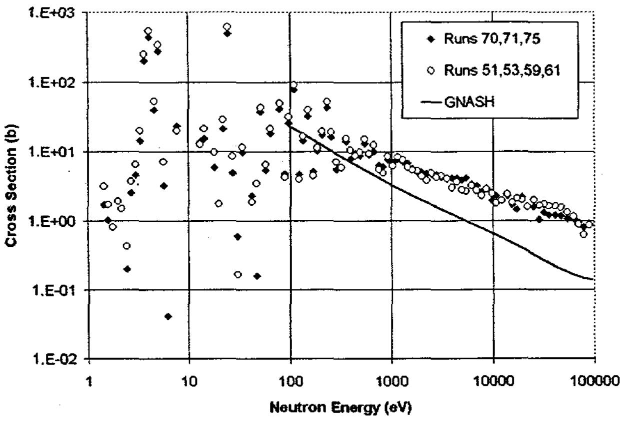

FIGURE 2. Cross section measured for the radioactive target ^{171}Tm, compared to the GNASH theoretical calculation (6).

In this configuration, the signal from the ^{171}Tm was only about 10% above the background from a blank Be foil assembly. Comparison of the background to the count rate with no foil indicates that neutron scattering was responsible for part of the gamma background. The background will be reduced in future measurements by decreasing the mass near the detector array and improving the shielding against neutrons from adjacent flight paths.

DANCE

The results of the preliminary experiment were encouraging and led to a proposal for a new detector array and flight path at the Lujan Center. The new detector will be located at 20 m on Flight Path 14, which views the newly installed "upper tier" water moderator. This moderator will have about half the flux of the high-intensity water moderator on Flight Path 4. However, planned improvements to the accelerator ion source and beam delivery are expected to eventually put up to 200 μA on the spallation target, about a 3X increase over the current proton current. By using different collimation which will view the entire moderator surface, the neutron flux at 20 m will actually be greater than on Flight Path 4 at 8 m.

In addition, a new detector array, the Device for Advanced Neutron Capture Experiments (DANCE) is being designed, and prototype detectors and collimation will be tested at LANSCE in fall, '98. The array will be, by necessity, fast and highly segmented. A 2 Ci target, which was the activity of the ^{171}Tm target reported above, has 7.4 x 10^{10} decays/second, or 740 decays in a 10 ns bin. To get one count per detector in a 10 ns period, assuming 20% efficiency, would require 148 detectors. This count rate will present interesting challenges. A capture event produces about 3 to 10 gammas. The segmentation of the detector array will enable multiplicity to be used to help discriminate against decay gammas.

The leading candidate for detector material is BaF_2, which has a 0.6 ns component that peaks in the ultraviolet at 220 nm. It also has a slow component that produces five times as much light, so filters or phototubes with special photocathodes will be required. We note that scintillators containing Cs or I have a large neutron capture cross section even at 10 keV, and would therefore produce an unacceptable background. Neutron absorbers, used successfully by others, do not appear to be effective above 1 keV (8).

DISCUSSION

We are designing and building a new detector array for measuring neutron capture on small quantities of unstable isotopes. These measurements will provide valuable cross sections for applied programs and nuclear astrophysics. A partial array is expected to be operational in 1999, and we are very excited about the prospects for physics with this detector.

ACKNOWLEDGEMENTS

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