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Science based stockpile stewardship, uncertainty quantification, and fission fragment beams

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Science Based Stockpile Stewardship, Uncertainty Quantification, and Fission Fragment Beams

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Stewardship of this nation's nuclear weapons is predicated on developing a fundamental scientific understanding of the physics and chemistry required to describe weapon performance without the need to resort to underground nuclear testing and to predict expected future performance as a result of intended or unintended modifications. In order to construct more reliable models, underground nuclear test data is being reanalyzed in novel ways. The extent to which underground experimental data can be matched with simulations is one measure of the credibility of our capability to predict weapon performance. To improve the interpretation of these experiments with quantified uncertainties, improved nuclear data is required.

As an example, the fission yield of a device was often determined by measuring fission products. Conversion of the measured fission products to yield was accomplished through explosion code calculations (models) and a good set of nuclear reaction cross-sections. Because of the unique high-fluence environment of an exploding nuclear weapon, many reactions occurred on radioactive nuclides, for which only theoretically calculated cross-sections are available. Inverse kinematics reactions at CARIBU offer the opportunity to measure cross-sections on unstable neutron-rich fission fragments and thus improve the quality of the nuclear reaction cross-section sets.

One of the fission products measured was ^{95}Zr, the accumulation of all mass 95 fission products of Y, Sr, Rb and Kr (see Fig. 1). Subsequent neutron-induced reactions on these short lived fission products were assumed to cancel out – in other words, the destruction of mass 95 nuclides was more or less equal to the production of mass 95 nuclides. If a ^{95}Sr was destroyed by an (n,2n) reaction it was also produced by (n,2n) reactions on ^{96}Sr, for example. However, since these nuclides all have fairly short half-lives (seconds to minutes or even less), no experimental nuclear reaction cross-sections exist, and only theoretically modeled cross-sections are available. Inverse kinematics reactions at CARIBU offer the opportunity, should the beam intensity be sufficient, to measure cross-sections on a few important nuclides in order to benchmark the theoretical calculations and significantly improve the nuclear data. The nuclides in Fig. 1 are prioritized by importance factor and displayed in stoplight colors, green the highest and red the lowest priority.

A more detailed description of the experimental and theoretical approach for this effort is given in the write-up below:

Reactions on fission fragments: experimental and theoretical approach

Experimental group: C.Y. Wu, L. Bernstein, and M.A. Stoyer (LLNL)
Theory group: I. Thompson, J. Escher, and R. Hoffman (LLNL)
Collaborators: J. Ullmann (LANL) and G. Belier (BIII)

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Accurate neutron-induced cross sections for reaction channels on fission fragments, such as (n,γ), (n,2n) …, are needed for a reliable network calculation of rad-chem diagnostics. These reaction cross sections are not expected to be measured directly because the short-lived fission fragments can not be fabricated into targets for experiments. However, they could be estimated in the framework of Hauser-Feshbach theory of compound nuclear reactions, providing the nuclear data existing for the fundamental quantities such as the level density, radiative strength function etc.. It has been demonstrated that the accuracy of this method for the predicting (n,2n) cross sections on nuclei near stability averages about 7% in comparison with the experimental data and for the (n,γ) cross sections averages about 30% [1].

Extrapolation of these calculations to fission fragments, where a region of nuclei is some distance away from stability, is a major challenge because there is no experimental data available for verification. In addition, calculations may be complicated by the unknown isospin dependence of level density and radiative strength function. Therefore, the confidence level for predicting for the reaction cross sections on fission fragments depends on our understanding of fundamental statistical parameters. Of particular concern is the recent observation by the Ohio University group [2] that the level density could potentially decrease dramatically for nuclei away from stability where fission fragments locate. The implications of this finding can be dramatic since the level density enters into Hauser-Feshbach calculations in an exponential form.

Recent advances in radioactive beam facilities, such as the CARIBU facility at Argonne National Laboratory, HRIBF at Oak Ridge National Laboratory, and TRIUMF, provide a unique opportunity to study the inverse kinematics reactions using nuclear fission fragments as beams. These reactions allow the extraction of the fundamental quantities mentioned above. The experimental priority thus is to develop a program to deduce quantitatively the level density and radiative strength function since these are the most important and yet uncertain fundamental quantities in the (n,2n) and (n,γ) cross-section calculation on fission fragments. These quantities can be derived from the measurements of particle evaporation channels including n, p, and α, together with the detection of deexcited γ rays from the compound nuclei formed in inverse kinematics. This is accomplished by using fission fragment as projectile on a hydrogen or deuterium-loaded target such as CH_2 or CD_2 or other light-to-intermediate mass targets. The sensitivity and reliability of this experimental approach has been demonstrated for stable nuclei in normal kinematics with proton or deuteron as projectile [3-7] and can be benchmarked using a suitable stable nucleus as beam in inverse kinematics.

Our initial plan is to study a case, where the level density is fairly well known, by taking advantage of the most advanced and sensitive instruments in low-energy nuclear physics, FMA [8], Microball [9], and Gammapshere [10]. The level density can be derived from the evaporated p and α spectra measured by Microball with the exit channel identified through the mass measurement by FMA. The radiative strength function, for example, can be derived partially from the γ-ray spectrum resulting from the two-step γ-cascade detected by Gammasphere in the proton capture channel identified by FMA.

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Developing a companion theory program to improve our understanding of these measured quantities, such as the level density, radiative strength function etc., is essential to accomplish the goal of this project; that is, to acquire the new capability to predict the neutron-induced reaction cross sections on fission fragments with a comparable accuracy for nuclei near stability, in particular, the (n,γ) cross section. This program goal has application to other important areas of fundamental nuclear science including both the *s*- and *r*-process of nucleosynthesis pathways, which when taken together are responsible for the formation of nearly all heavy element production beyond iron.

In addition, theorists will help to identify the best case to benchmark experimentally this approach before going to the mass 95 and 147 region of program interest. They also will help in planning the experiments and interpreting the experimental results, and then integrate the new finding into the model calculations for neutron-induced reaction cross sections on fission fragments.

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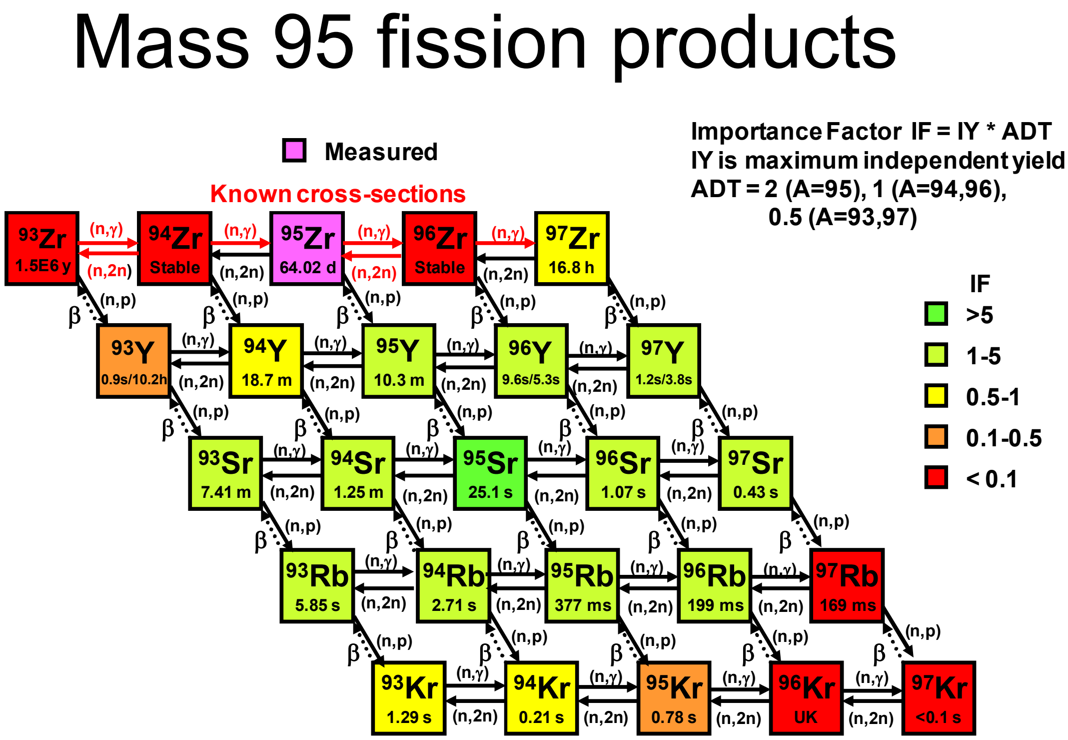

Fig. 1: A portion of the chart of nuclides showing the neutron-rich region near one of the measured fission indicators, ^{95}Zr. Nuclides in this region are interconnected via (n,2n), (n,γ), (n,p) reactions and beta-decay. Colors indicate the importance of measuring cross-sections for short-lived radioactive nuclei. The importance factor is the product of the independent fission yield of the isotope and a mass dependent term (ADT). High importance factors are indicated in bright green.

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