Project No. 09-802

Improved Fission Neutron Data Base for Active Interrogation of Actinides

Integrated University Program

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In collaboration with:
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Los Alamos National Laboratory
University of Kentucky
Texas A&M University

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NEUP Nuclear Energy University Programs
U.S. Department of Energy

Prepared by: Andreas Enqvist, Sara A. Pozzi, Brian Wieger
University: University of Michigan

Final report period

This period focused on getting ready for the planned measurement campaign at Los Alamos Neutron Science Center (LANSCE) Oct. 1 - Oct. 17 2012. Leading up to the measurement the data acquisition system was developed and tested to expand to using up to 32 channels of simultaneous data, such as detectors and fission chambers. A structure made specifically for the measurement campaign was also designed and commissioned to be built. Further the detectors for the measurement, EJ-309 liquid scintillators, Sodium-iodide (NaI) detectors and Li-glass detectors based on enriched Li-6 were tested and characterized.

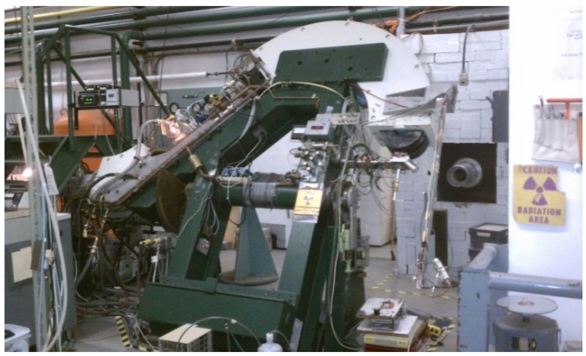

Fig. 1 Detector characterization at Ohio University's Edwards Accelerator Facility. The picture shows the beam-swinger connected to a tandem Van De Graaff generator.

Neutron light response functions and detector resolution functions were measured at Ohio University's tandem Van de Graaff generator for EJ-309 liquid scintillators, having dimensions 7.6-by-7.6, and 7.6-by-5.1 cm. A ~7.44 MeV deuteron beam was used on a ^{27}Al target generating a continuous spectrum over the energy range from a few hundred keV to over 10 MeV. EJ-309 is a liquid scintillator substance which is very suitable for radiation detection with good pulse shape discrimination and nonhazardous properties.

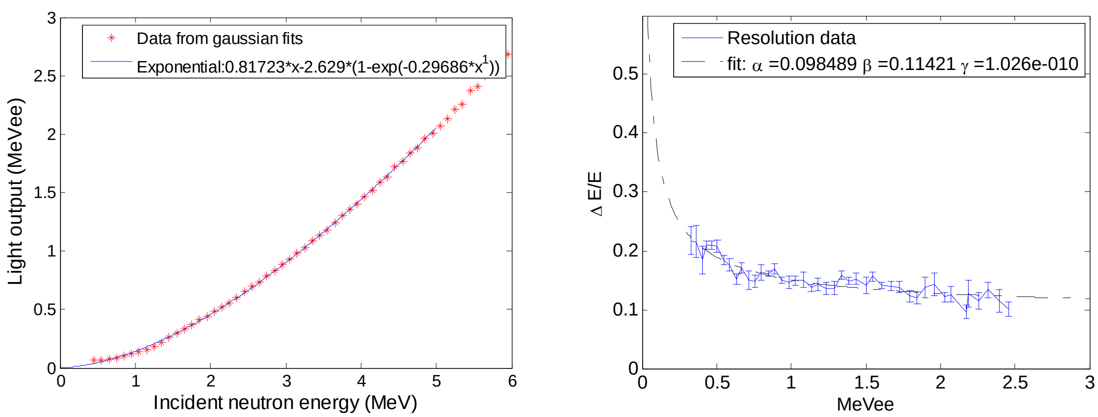

Fig. 2 Neutron light output response function and detector energy resolution as a function of pulse height.

The setup can be seen in Fig 1. The resulting light output response function is fitted to an exponential form. Furthermore, the long flight path and good timing characteristics of the pulsed neutron source enables unfolding of the detector energy resolution.

The new measurement campaign has been successfully completed and final data validation and analysis are underway. In Fig. 3 the setup is shown together with a computer model used for the Monte Carlo simulations that also models the whole detector setup and the purposely build detector holder.

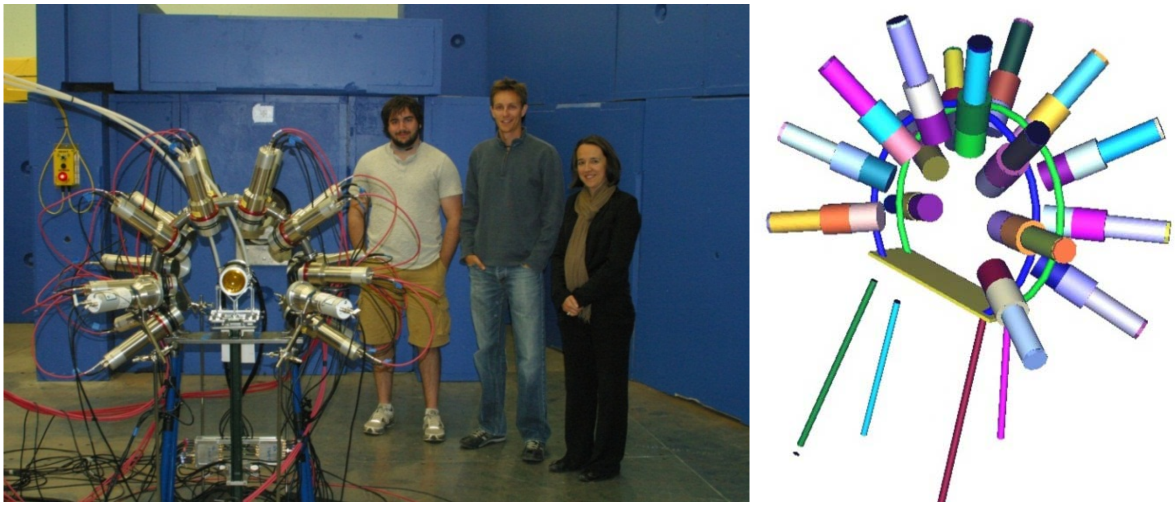

Fig. 3 Setup at LANSCE (left) and a visualization (right) of the same detector setup used in Monte Carlo simulations.

Early detector characterization focused on smaller cells of EJ-309 liquid scintillators with which the new campaign could go further down in detectable neutron energy. However, Li-6 glass detectors were also incorporated into the measurement as shown in Fig. 4.

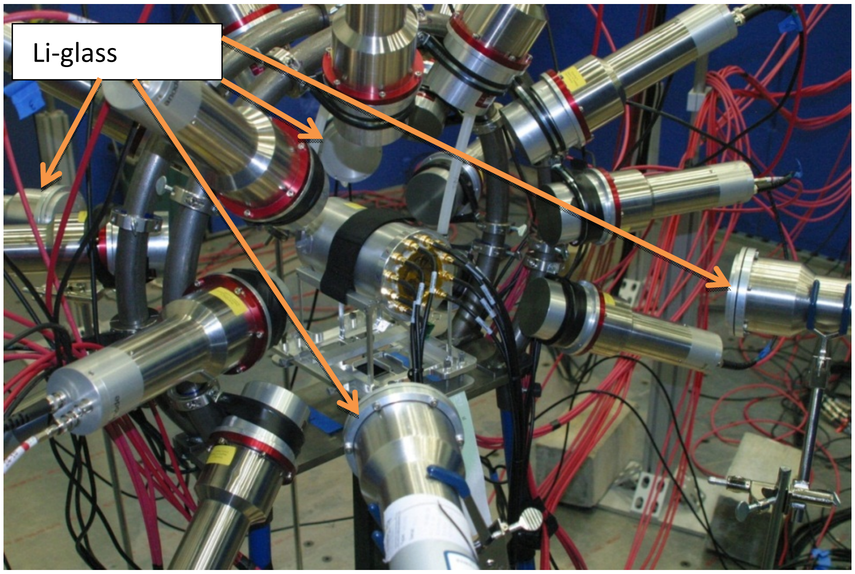

Fig. 4 Experimental setup used at LANSCE. The main holder for EJ-309 detectors can be seen encompassing the central fission chamber with U-235 foils.

The data analysis is using both LANSCE acquired data and additional characterization measurements performed at UM after the LANSCE measurement campaign to reduce the different time and position uncertainties which otherwise leads to large propagated uncertainties in the time-of-flight (TOF) spectrum.

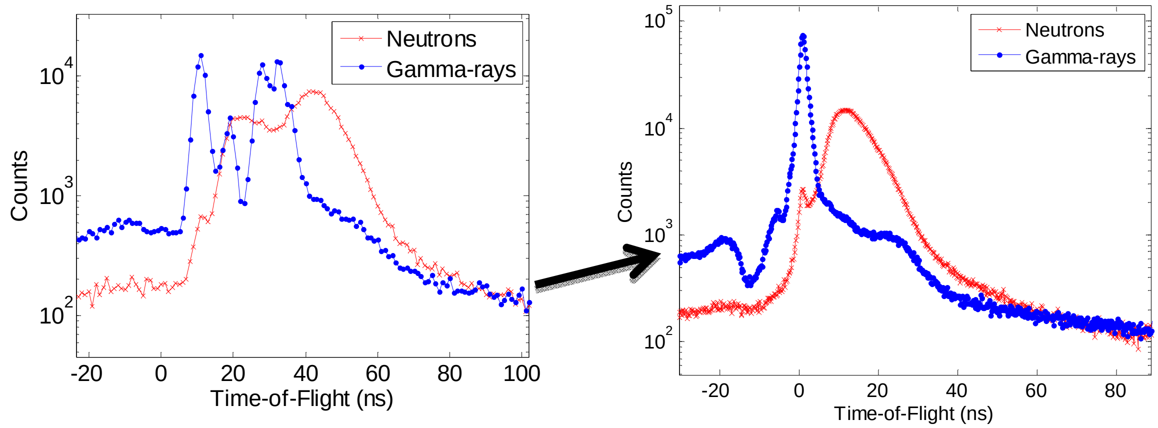

Fig. 5 Here the improvement in timing of the data achieved by applying setup- and detector-dependent time corrections. The left is the uncorrected TOF spectrum, while the right is corrected TOF spectrum.

The analysis for energy-angle correlations is ongoing with early results showing the neutron-neutron cross-correlation dependence as a function of the angle between detectors. Higher order correlations are also being investigated such as bi-correlations visualized from the LANSCE data in Fig 6.

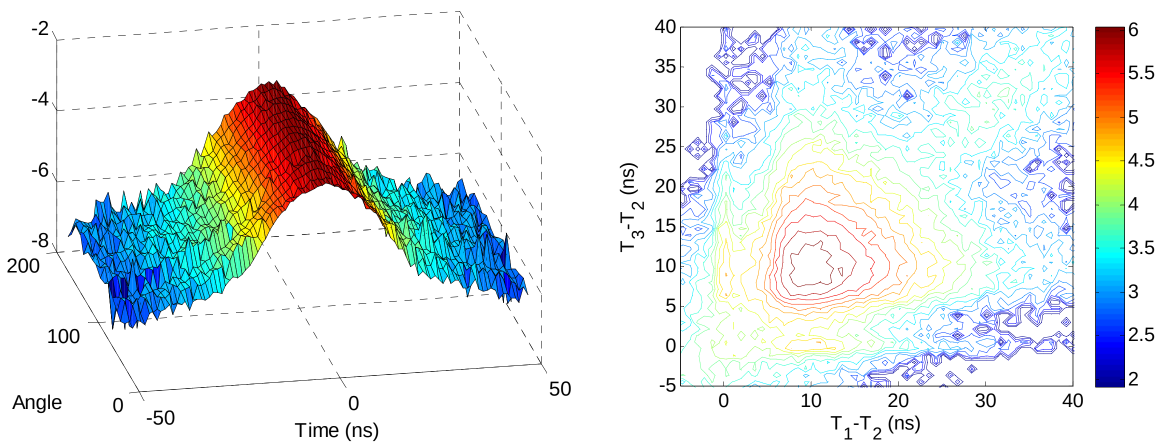

Fig. 6 Setup at LANSCE (left) and a visualization (right) of the same detector setup used in Monte Carlo simulations.

More specifically the extension into lower energy neutron pulses as compared to previous campaigns is continuously being investigated. The analysis for the EJ-309 emission spectrum is near completion, while the Li-glass detector data needs some further complementing simulations which will be used to characterize the detection efficiency needed to unfold the incoming flux.

Patents/Publications/Presentations

The work on detector resolution was presented as a poster at The Symposium on Radiation Measurements and Applications (SORMA-west), Oakland, CA in May.

Journal papers:

A. Enqvist, B. M. Wieger, L. Huang, M. Flaska, and S.A. Pozzi, R. C. Haight, H. Young Lee and C. Yen Wu, "Neutron-Induced 235U Fission Spectrum Measurements Using Liquid Organic Scintillation Detectors" PHYSICAL REVIEW C **86**, 064605 (2012).

S. A. Pozzi, S. D. Clarke, W. Walsh, E. C. Miller, J.L. Dolan, M. Flaska, B. M. Wieger, A. Enqvist, E. Padovani, J. K. Mattingly, D. Chichester, and P. Peerani, "MCNPX-PoliMi for Nuclear Nonproliferation Applications," NIM-A, vol. 694, pp. 119-125, Sept. 2012.

Conference contributions:

"Multiplicity Distributions and Energy-Angle Correlations in Spontaneous Fission Neutron Emissions", B. Wieger, A. Enqvist, S. A. Pozzi, Institute of Nuclear Materials Management Annual Meeting, July 15 – 19 (2012) Orlando, Florida.

"Neutron light output functions measured for EJ309 liquid scintillation detectors", A. Enqvist, C. C. Lawrence, T. N. Massey, S. A. Pozzi, Proceedings of INMM Annual meeting, Orlando, FL, July 15-19 (2012).

S. A. Pozzi, S. D. Clarke, W. Walsh, E. C. Miller, J. Dolan, M. Flaska, B. M. Wieger, A. Enqvist, E. Padovani, J. K. Mattingly, D. Chichester, and P. Peerani, "Validation of MCNPX-PoliMi Fission Models," To be presented at the IEEE Nuclear Science Symposium Conference Record on CD-ROM, Anaheim, CA USA. 27 Oct. – 3 Nov, 2012. (poster)

A. Poitrasson-Rivère, M. Flaska, M. C. Hamel, K. Ide, J. K. Polack, S. D. Clarke, and S. A. Pozzi, "Digital Data Acquisition and Processing for a Neutron-Gamma-Ray Imaging System," To be presented at the IEEE Nuclear Science Symposium Conference Record on CD-ROM, Anaheim, CA USA. 27 Oct. – 3 Nov, 2012. (talk)

Students

Lu Huang, Chinese, Nuclear Engineering.
Brian Wieger, American, Nuclear Engineering.

Prepared by: Michael A. Kovash
University: Kentucky

Final report period

Task A: Simulations have begun which will help to determine the accuracy we might achieve in measuring the cross section for neutron-proton scattering in the range from 100 keV to 2 MeV. These Monte Carlo results were used to make improvements to the design of our neutron beam line.
Task B: A draft publication has been prepared reporting our measured cross sections for neutron-proton scattering in the energy range from 150 to 500 keV. The data have been analyzed in the effective range theory.
Task C: A 9-layer prototype detector bar will be assembled and tested with neutron and gamma-ray sources at our home accelerator laboratory.

Task D: Our analysis of the data collected earlier in 2011 has been completed, and final cross sections for neutron-proton scattering have been determined in the energy range from 150 to 450 keV. The data show excellent agreement with a prediction based on effective range theory.

Task E: Work continued on our analysis of the scintillator response data collected on BC418 at both LANSCE/WNR, and UKy. The results between 100 keV and 4 MeV are now completed.

The UKy students listed below have been involved in Tasks A - C, above, and continue to participate fully in prototype testing and performing measurements using the layered bar detector.

Patents / Publications / Presentations

Kovash presented an invited paper at the NEUP Advanced Nuclear Data Development Meeting, Fort Worth, October, 2011.

Kovash presented an invited paper at the LANSCE Topical Workshop, Los Alamos, January, 2012.

Kovash presented a paper at the NNSA SSAA meeting in Washington, D.C. in February, 2012.

Students

1. Khayrullo Shoniyozov - PhD candidate in physics citizenship: Uzbekistan
2. Zachariah Miller - PhD candidate in physics; citizenship: USA
3. Hongwei Yang - PhD candidate in physics; citizenship: China

Prepared by: Lawrence Rees
University: Brigham Young University

Students involved: Stephen Black (chemical engineering), Alex Corey (physics), and KaeCee Terry (physics). All are undergraduates and US citizens.

Final report period

Growing and Testing Ammonium Halide Crystals

The difficulties we have encountered in growing ammonium halide crystals have taken us off from our originally proposed schedule. Our efforts in the last repot period have been along four separate lines of research.

1) Speleo Resources has continued their efforts to grow ammonium bromide crystals.

Crystals of up to about 1 cm^3 are now being grown regularly in temperature controlled baths. It has proven to be very important to keep the temperature very constant and to include urea in solution for optimum crystal growth. Clear, high-quality crystals take about 3 to 4 weeks to grow.

Attempts to incorporate $Eu^{(2+)}$ into the system, however, have yet to be successful. The europium oxidizes to $Eu^{(3+)}$ as evidenced by yellowish or brownish discoloration of the solution. Measurements confirm the expectation that the $Eu^{(3+)}$ is not incorporated into the crystal lattice. The $Eu^{(2+)}$ ion will remain in water solution for a few hours or perhaps even a few days, but high-quality crystals will not grow that fast.

To combat the oxidation problem, various reducing agents have been added to the solution. Since $Eu^{(2+)}$ is a rather strong reducing agent already, and the added reducing agents either failed to keep the europium reduced, or they reacted with the solution (reducing the water to hydrogen gas).

Additionally, solvents other than water have been tried as well. The solvents used have been alcohols, acetone, ethylene diammine, dimethyl sulfoxide, and several eutectic salt mixtures. The alcohols and acetone had very low solubilities, and produced only dendritic crystals. The ethylene diammine reacted vigorously and boiled out ammonia gas. DMSO appeared to work, but the solubility was low, and it appeared to react slowly at the higher temperatures needed for crystal growth. The best alternate solvent appeared to be a deep eutectic salt mixture, consisting of urea + acetamide + NH_4Br + a trace of water (perhaps 3 to 5%). This appeared to be chemically stable but even without any water, this mixture is a viscous liquid at room temperature. Several nice-looking NH_4Br crystals were grown from a eutectic melt liquid, but the solubility is lower and the crystals don't seem to grow any faster or better than could be done already from a water solution.

At this point, we will try growing some ammonium iodide crystals, but the oxidation problem will still remain. We will also try growing some low-quality ammonium bromide crystals, which can be grown quickly. We will see if we can get europium into the crystals before it oxidizes or escapes the growing crystal lattice.

2) We have been working on growing NH_4I crystals at BYU and have had some success. It is easier to grow good quality iodide crystals than bromide crystals. But we have not tried incorporating Eu into the crystals yet. We are currently trying to add some Sr into two batches of NH_4I crystals that we are growing.

The ammonium iodide crystals that we have grown are quite clear. There is some discoloration near the centers of most crystals, but this discoloration disappears after a while. Crystals have measured up to about 2 cm in largest dimension. The largest crystals shown in Fig. 1 are about 4-5 mm across.

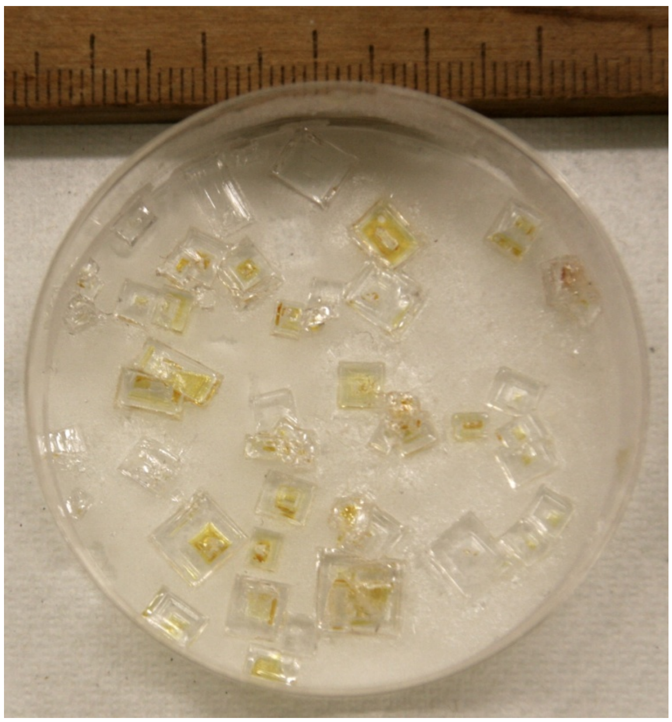

Fig. 1 Ammonium iodide crystals grown at BYU.

3) We have been analyzing data taken at Ohio University to experimentally measure the neutron detection efficiency of two plastic-scintillators, cadmium capture-gated detectors as a function of neutron energy. The first of these two detectors was a 6”×6”×5” dia., one-phototube detector with wedge-shaped plastic scintillator pieces separated by cadmium sheets (the wedge-detector). The second was a 10”×10”×6” four-phototube detector with 1-cm-thick plastic scintillator slabs separated by cadmium foils (the four-fold detector). The purpose of these measurements is to establish a baseline for the performance of plastic scintillator in cadmium capture-gated detectors.

We have seen some unexpected structure in the neutron spectrum, but this is consistent with University of Michigan measurements taken at Ohio.

We have finished much of the preliminary analysis of the data. We are working to see neutron signals at levels of about twice the noise. This can be done because of the large capture pulse that is uncorrelated in height with the initial neutron energy. Since we are using signal digitizers, we can trigger on the capture pulse and look at earlier times for neutron signals (proton recoil signals) that are too small to be used for the initial hardware trigger.

4) In the absence of scintillating ammonium halide crystals for direct measurements, we have undertaken some Monte Carlo (MCNP-PoliMi) calculations to compare plastic scintillator (EJ-200) response with the response of ammonium halides in a hypothetical cadmium capture-gated detector. The detector we modeled in what follows is a 6”×6”×5” dia. detector with 1-cm-thick plastic scintillator slabs separated by 0.1 mm-thick cadmium foil. The source is unshielded ^{252}Cf.
The version of MCNP-PoliMi we used was modified by the University of Michigan to include a gamma cascade from neutron capture in ^{113}Cd. Although details of this cascade are not precise, they do provide a guideline for comparison.

Table 1 summarizes the number of cadmium capture events where the energy deposited by Cd gammas in the scintillator is above a given threshold. The data are given as a ratio of captures with ammonium halide to captures with plastic scintillator as a function of the threshold energy for gamma capture, E_γ .

Table 1. Comparison of Cd gamma energy absorbed in ammonium halides compared to plastic scintillator.

Cd gamma threshold	NH_4Br/plastic	NH_4I/plastic
E_γ > 1 keV	0.909511	0.758275
E_γ >1 MeV	1.263189	1.100149
E_γ > 3 MeV	1.838923	1.838923

The histogram in Fig. 2 shows the number of capture events with the total energy deposition from Cd capture gammas in a range up to 9 MeV. Although the total number of events leaving energy in plastic scintillator is larger than for ammonium halides, the latter are more likely to produce large cadmium-capture pulses. The ammonium halides are about twice as likely as plastic to absorb over 3 MeV energy from a cadmium-capture event. The value of 3 MeV is a little greater than the gammas typically found in background and hence is very useful for distinguishing neutrons from false doubles.

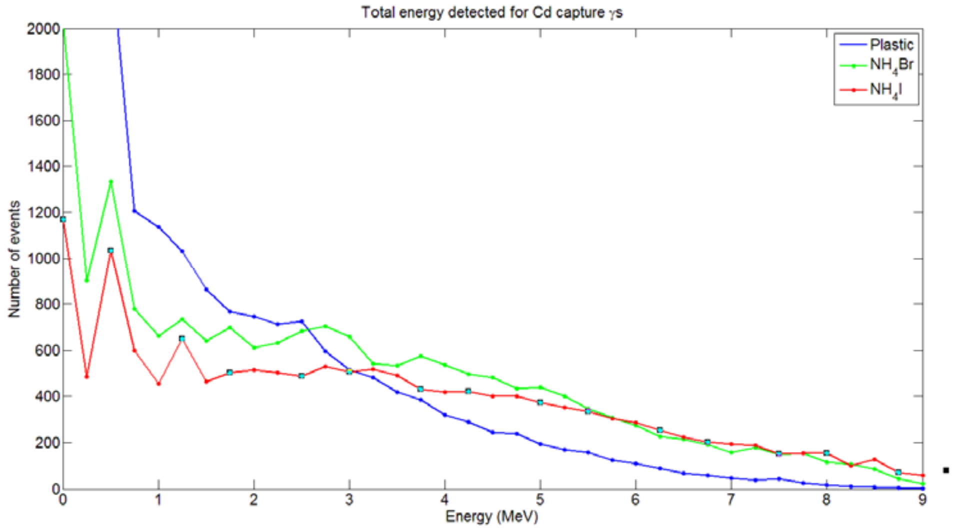

Fig. 2. Cd gamma energy absorbed in ammonium halides and plastic scintillator.

One important characteristic of ammonium halide crystals that is not addressed in these calculations is that the light output from proton recoil events (instigated by incident neutrons) should be proportional to the proton energy. With plastic scintillators, the relationship between proton energy and light output is notoriously nonlinear. This means that an ammonium-halide-based scintillator should function much better as a spectrometer. For either organic or inorganic scintillators, we know that if there is a cadmium capture event, all of the neutron's original kinetic energy is lost in the detector. However, the total proton recoil pulse should be more closely related to the incident neutron energy when inorganic scintillators are employed.

Building and Testing a Four-PMT Cd Capture-Gated Detector

Our mandate is not only to develop and test inorganic scintillators, but also to compare them to detectors using organic scintillators. Before last quarter, we had studied a single 5' diameter x 6" deep cadmium capture-gated detector for comparison. However, we realized that this detector would be too small to have good efficiency for detecting the gamma-rays emitted when neutrons capture in cadmium. For this reason, we built a 10" x 10" x 6" plastic detector with four photomultiplier tubes to improve capture efficiency. We took this detector in November to the John E. Edwards Accelerator Laboratory at Ohio University to measure its efficiency as a function of incident neutron energy. We are still in the process of analyzing the data.

One of the important characteristics of proposed inorganic scintillators is the high Z of the detecting material. Unlike most neutron detectors, cadmium capture-gated detectors need scintillating materials that are more effective at stopping gammas. Using ammonium halides will provide more efficiency with the same size detector. We can estimate detector efficiency of larger arrays with Monte Carlo calculations;

however, we need to use the Ohio data as a baseline to see how well the calculations predict the measured values.

The analysis of the Ohio data has been compared to the MCNP predictions. With those results as a guideline, additional series of MCNP and MCNP-PoliMi simulations were performed to characterize ammonium halide-based detectors with similar geometries.

Fig. 3. Four-PMT Cd Capture-Gated Detector using plastic scintillator.

Patents / Publications / Presentations

Nothing for this time.

Students

Stephen Black, USA, chemical engineering

Prepared by: Robert C. Haight
Laboratory: Los Alamos National Laboratories

Final report period

- The construction of the new flight-path has been completed. The new experimental area features an extended neutron pit to reduce background and enable a longer time-window for detection of correlated neutrons. The new building is pictured from the outside in Fig. 1 and the new flight path, before installation of the fission chamber and the neutron detectors, is shown in Fig. 2.
- The fission chamber was installed in the experimental area after beam characteristics had been taken.
- The characterization of the beam involved measuring the beam profile, general neutron flux and background.
- Beam filters have been installed in the form of lead and polyethylene to reduce the pulsing overlap as well as to reduce some of the photon flux
- Energy dependence of the neutron multiplicities is being considered in planning, location, and configuration of current and future detector arrays
- An investigation of the new measurement conditions impact on improved detection of high order multiplicities is ongoing.

Fig 1. New experimental building where neutrons emitted in neutron-induced fission will be studied. Spallation neutrons from 0.1 to about 200 MeV are produced by the pulsed 800 MeV proton beam from the LANSCE accelerator and are collimated in flight paths for time-of-flight measurements.

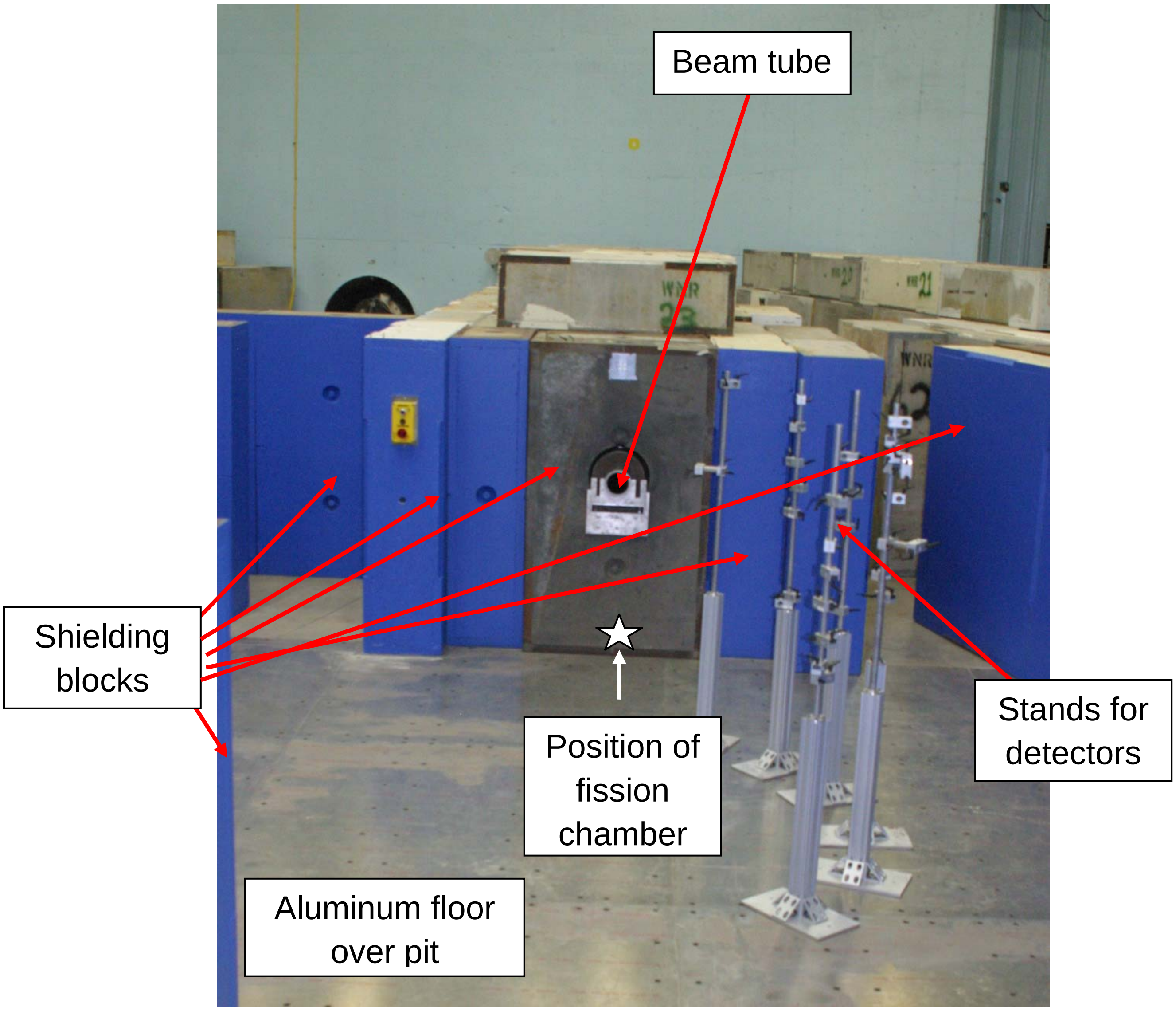

Fig 2. The new flight path and experimental area where neutron multiplicity measurements will be made. The view is looking toward the neutron source, which is 22 meters in front of the position of the fission chamber. Stands are shown before neutron detectors are mounted in them. The aluminum floor covers a pit 18 x 18 x 7 $feet^3$ which serves as a "get lost" area for scattered neutrons.

Currently testing is underway with additional new fission chamber targets, specifically based on plutonium isotopes. Early prototypes with partial loading have been successfully tested. Future run cycles of the accelerator starting next year will then be possible to utilize the new isotopic targets for measurements not just internally but also for visiting groups.

The 2012 measurement campaign with University of Michigan was successfully completed in October 2012 (Fig. 3), and that data is currently being used to complement previous measurements, and as a benchmark for future improvements.

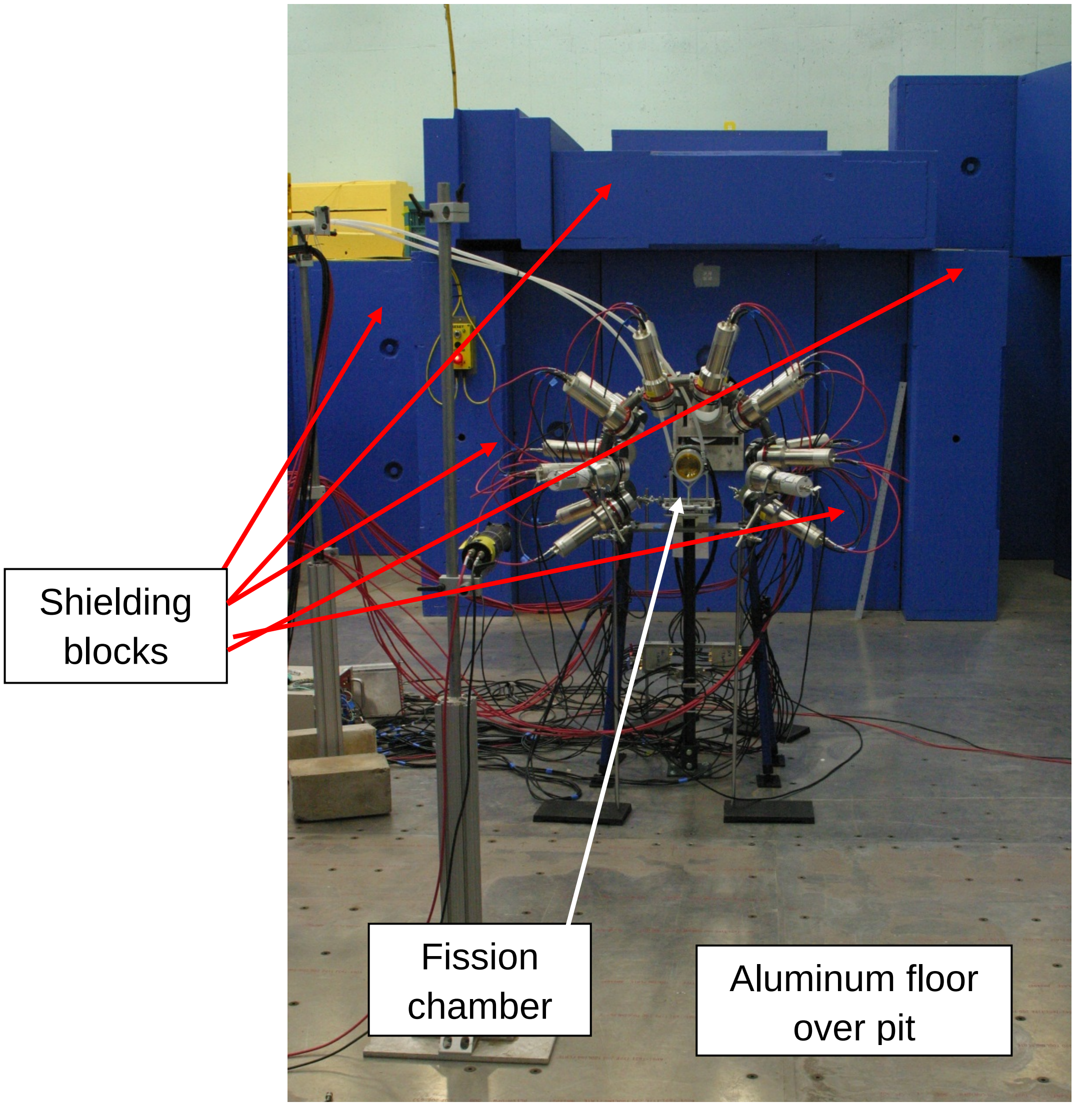

Fig 3. The new flight path and experimental area where neutron multiplicity measurements are made. The view is looking toward the neutron source, somewhat obscured by the newly installed parallel plate avalanche chamber, as well as the University of Michigan detector setup surrounding it. The aluminum floor covers a pit 18 x 18 x 7 feet^3 which serves as a "get lost" area for scattered neutrons.

Patents / Publications / Presentations

Requests for beam time were submitted to LANSCE/WNR Nuclear Physics Advisory Committee. Presentations were made. The Committee rated the proposals highly and the requests were approved for beam time.

Prepared by: Dr. Pavel V. Tsvetkov
University: Texas A&M University

Final report period

The Texas A&M Team's efforts provide computational support to the overall project to develop improved neutron detection system for active interrogation measurements of actinides contributing to basic physics data. Within the 3-year work scope, the team will contribute to selection and construction of detectors by quantifying performance needs and uncertainties, and by development of a fission neutron emission spectra database for actinides relevant to the AFC program. Following experimental schedules, once experimental data are obtained from the project partners (LANSCE, UM, UK, BYU), experimental data will be analyzed and compared to the corresponding high fidelity simulation results. The uncertainties will be quantified and correlated to experiments and simulations. The integrated consistent basic fission neutron spectral datasets will be created taking advantage of experimental data augmented by simulated basic parameters as appropriate. The effort progresses towards developing an ability to augment actual physical data in nuclear data sets with surrogate and contemporary experimental data (not yet in evaluated data files) to assess needed fidelity in fission spectrum evaluations as well as to quantify impact of improvements in measured spectra on fuel cycle and reactor simulations.

Brief summary of work completed to date and key accomplishments:

The past quarter shifted the work focus on documenting the project results and moving towards the project final report and deliverables. The integrated framework for consistent studies of fission spectrum effects on system calculations is being used to produce reference and corresponding modified cases with artificially adjusted spectra data. Developed model for simulations of experiments conducted by team members as they refine the actual experimental configurations is being integrated into the system model framework. The capability and data sets for sensitivity/uncertainty evaluations using surrogate and experimental data in modified nuclear data sets is being finalized. All these activities assume use of Monte Carlo methods, MCNP/MCNPX and MCNP-PoliMi to avoid additional constraints and model assumptions of deterministic methods. These tasks have been progressing on schedule and as planned. While developing an integrated framework, it has been realized that evaluations have to carry studies accounting for configurations with both thermal and fast neutron spectra and with broader range of actinide and structural material descriptions – these studies are currently in progress. In thermal spectrum systems, the fidelity of the fission spectrum potentially influences the formation of neutron distributions in the resonance energy range while in a fast spectrum system, the entire neutron distribution depends on the accuracy of fission spectrum data, especially if high fidelity simulations are thought of. These observations were made while proceeding with developments of the automated framework and performing reference simulations of power units with LWRs (AP1000) and VHTRs and the corresponding fuel cycle estimates (transmutation efficiency). The experimental setup details have been incorporated within the detailed model developed for simulations with MCNP-PoliMi. The model of the LANCE cave includes beam port data and detector arrays. During the reported quarter the model has been made fully functional and prepared for further simulations focusing on comparative studies and evaluations with respect to obtained experimental data. It is understood and observed in preliminary simulations that the cave configuration itself is likely to be important as it may potentially affect experimental results.

Subcontract establishment at TAMU: January 6, 2010, interim funding – December 15, 2009

Task 1.2. Sensitivity studies to determine and quantify dependence of simulations on fission neutron data.

Personnel involved: Dr. Tsvetkov's research group

Task Status: completed.

- Issues/Concerns: None

Task 1.4. Assessment of evaluated nuclear data files for selected actinides and fidelity of the data vs. basic physics data needs

Personnel involved: Dr. Tsvetkov's research group

Task Status: completed

Issues/Concerns: None

Task 1.5. Quantification of the existing uncertainties in the available fission neutron emission spectral data including their impact on DOE NE program needs and specific experiments needed per nuclide and per energy range

Personnel involved: Dr. Tsvetkov's research group

Task Status: completed

Issues/Concerns: None

Task 1.6. Identification of ideal detector characteristics for active interrogation options for actinide basic physics characteristics

Personnel involved: Dr. Tsvetkov's research group

Task Status: This task is in progress

Issues/Concerns: None

Task 1.7. Mockup modeling of experiments following Year 1 experiment schedules

Personnel involved: Dr. Tsvetkov's research group

Task Status: completed

Issues/Concerns: None

Patents / Publications / Presentations

Reports:

TRU MANAGEMENT AND ^{235}U CONSUMPTION MINIMIZATION IN FUEL CYCLE SCENARIOS WITH AP1000 AND VHTRS, Marie-Hermine M. Cuvelier and Pavel V. Tsvetkov

FISSION SPECTRUM UNCERTAINTY EFFECT ON FUEL CYCLES WITH AP1000 AND VHTR, Marie-Hermine M. Cuvelier and Pavel V. Tsvetkov

Students

Marie Cuvelier, France. - Focuses on the system framework development and LANCE facility modeling.
Chris Chapman, US. – Sensitivity/uncertainty analysis and framework development efforts for surrogate data integration.
Kristina Yancey, U.S. – Focuses on the computational system framework and fuel cycle modeling.