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IMPROVING THE NUCLEAR DATA BASE FOR NON-PROLIFERATION AND HOMELAND SECURITY

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ABSTRACT

Many of the technical advances in non-proliferation and homeland security require calculations of transport of neutrons and gamma-rays through materials. The nuclear data base on which these calculations are made must be of high quality in order for the calculated responses to be credible. At the Los Alamos Neutron Science Center, three spallation neutron sources are being used to provide high-quality cross section and structure data with reactions induced by neutrons. Neutron transmission, neutron-induced fission and capture cross sections, neutron emission in fission, and gamma-ray production by neutrons are principal areas of research. Furthermore, these sources are also being used to validate calculations of the characterization and response of new detectors and detection techniques. Current research activities are summarized here.

Key Words: Neutrons, reactions, radiation, detectors, integral experiments

1. INTRODUCTION

The technical issues in Homeland Security and Nuclear Non-Proliferation are obviously broad and challenging, and many approaches involve nuclear techniques. Interrogation of cargo is intended to signal the presence of clandestine materials such as fissile materials, high explosives, and illicit drugs. The exact amount of these materials is not the issue in the first analysis, but only their presence in some quantity above threshold levels that would justify further action to secure the cargo. The materials of the cargo, their geometry, and possible attempts to defeat the interrogation are wide ranging. At the other extreme, for non-proliferation applications, the exact amount of fissile material in a process stream needs to be known to assure that there has been no diversion. In this case, the quantitative measure needs to be very precise and will often require an isotopic analysis. Here the materials and geometries are in principle known, but the environment for making the measurements (high radiation fields) can be very hostile. A diversion of even 0.1% of fissile materials in a reprocessing stream is not acceptable.

To assess approaches to high-sensitivity interrogation or high-precision assay, radiation transport codes such as MCNPX [1] are used. These codes use data derived from evaluated nuclear data libraries, such as ENDF/B-VII [2], and the evaluations often rest on a foundation of experimental data. If the data are incorrect or, more likely, have large uncertainties, then the results of the transport calculation can also be incorrect or have large uncertainties. In cases where there are no experimental data, the evaluator uses nuclear reaction model calculations or systematics to arrive at an estimate of the required nuclear data. In many instances, the types of data needed for non-proliferation and homeland security overlap in many instances with those needed over many years for programs in weapons and reactor development. There is often, however, a different emphasis in the importance of certain materials and certain data and in the accuracy required. Thus new measurements and evaluations of the cross section and other nuclear data are underway at many laboratories to improve the data base for these new applications.

Furthermore, the results from these new simulations need to have some benchmarked data against which to compare, as do the more traditional applications. Integral experiments that mock up, to some degree, the radiation-transport characteristics of present interest test the validity of the simulations. Such integral experiments have been keys to validating and improving the data base for reactor and other applications over many years.

In addition to simulations of radiation transport, the development of improved radiation detectors and sources is another important part of these programs. Radiation detectors, whether based on traditional or new materials, need to be fully characterized and calibrated. Again, simulations of the radiation transport in materials is crucial to understanding the response of detectors, but also the experimental investigations of pulse-height and time response and of fielding issues. Radiation sources for interrogation need to be optimized to detect the materials of interest, and the improvements in radiation detectors are improving the characterization of these sources.

At the Los Alamos Neutron Science Center (LANSCE), our principal activities relevant to non-proliferation and homeland security are a broad range of nuclear data measurements. These data are used by evaluators to improve the data base of interactions between neutrons and nuclei. We use a variety of instruments to make high precision measurements of cross sections for neutron-induced fission, gamma-ray production, and neutron capture and to study fission outputs of neutrons and gamma rays. We also can measure neutron total cross sections with excellent absolute accuracy.

Our neutron sources also can be used to perform integral tests to benchmark neutron transport calculations from thermal to several hundred MeV. The flexible nature of the facility allows this wide range of neutron energy and also a wide range in neutron intensity. We also can characterize some neutron-interrogation schemes as a function of neutron energy and thereby provide information on the optimum probe energy.

In this report, we describe briefly the LANSCE facility and follow with an overview of the nuclear data measurements and integral experiments. Possibilities for new approaches both to nuclear data as well as to integral experiments are discussed.

2. LANSCE FACILITY AND INSTRUMENTS

The LANSCE facility has been described previously [3] and the general characteristics for the neutron-nuclear physics studies are summarized in Table 1. Neutrons are generated at LANSCE by directing the 800-MeV proton beam onto tungsten targets. Three neutron sources are used for nuclear data measurements. At the fast neutron source (WNR), the tungsten target has no moderator, and the useful neutron energy range is from about 0.1 MeV to about 600 MeV. The sample and detector are placed at flight path lengths from 8 to 90 meters so that the incident neutron energy can be determined by time-of-flight. At the moderated neutron source (Lujan Center) with flight paths from 8 to 60 meters, neutrons from sub-thermal to 200 keV are available. For very intense neutron fluxes, a Lead Slowing-Down Spectrometer (LSDS) consists of a 1.2 meter cube of pure lead surrounding the target. Samples and detectors are placed in channels in the lead. An effective path length is 6.3 meters characterizes the time-energy relationship here.

The proton beam current for each of the WNR and Lujan sources is usually tuned to be the maximum consistent with maintaining a clean pulse structure and current limitations of the tungsten target. High current is required for experiments on most of the flight paths. If the neutron flux is too large for an experiment, then it can be reduced by smaller collimation or by placing attenuating material in the flight path for that experiment.

Instruments for cross section measurement include, for fast neutrons at WNR, the FIGARO neutron detector array, the GEANIE array of HPGe detectors for detecting gamma-rays with good energy resolution, parallel plate fission chambers, charged-particle detectors, plastic scintillators and other detectors. At the Lujan source, the DANCE 4-pi array of BaF_2 detectors serves as a calorimeter for neutron-capture measurements. Fission chambers and HPGe detectors are also used. Imaging detectors are also possible at both of the fast and the moderated sources. At the LSDS, fission chambers and detectors for light charged particles are appropriate.

Table 1 – LANSCE neutron sources and instruments used for nuclear data measurements.

Neutron source	En	Flight paths	Instruments	Principal uses
WNR	0.1 – 600 MeV	6 each with lengths of 8 – 90 meters	FISSION, GEANIE, FIGARO, N,Z Others	Fission c/s N,xgamma Fission neutrons and gamma rays Charged particle Production Total c/s, transmission, detector calib, etc.
Lujan Center	Subthermal – 200 keV	2 each with lengths of 8 – 60 meters	DANCE, FISSION	Neutron capture, fission gammas, capture-fission ratio Fission c/s
Lead Slowing-Down Spectrometer	0.1 eV – 100 keV	Several internal channels. Effective 6.2 m flight path	FISSION	Fission c/s on small samples Fuel rod assay

3. DATA BASE MEASUREMENTS

Basic cross section data are provided by measurements at all three sources. They include neutron total cross sections, fission cross section measurements, neutron and gamma-ray output from fission, gamma-ray production by inelastic scattering and other reactions, neutron capture cross sections, charged-particle production, and other reaction data. Some examples are given below. In addition to the direct use of these data for producing evaluated data files, they are also used to refine nuclear reaction model codes, which are used not only in the evaluation of the data for these reactions but also for data on neighboring nuclei.

3.1. Neutron Total Cross Sections

The only neutron cross section that can be measured without referring to other cross sections is the neutron total cross section. It is measured simply by neutron flux attenuation in so-called "good geometry," that is, all scattered or reacted neutrons are completely removed from the flux. The LANSCE beam structure at the WNR source is ideal for these measurements. An extensive program in neutron total cross sections from 5 to 560 MeV was completed several years ago and yielded total cross sections with uncertainties on the order of 1% [4]. These measurements could be extended to lower neutron energies with new techniques employing waveform digitizers to simplify the electronic apparatus. Neutron total cross sections relate very directly to radiographic approaches of interrogating cargo. With these very accurate total cross section data, simulations of what can be imaged inside of a given container will have high credibility.

3.2. Fission Cross Sections

Experimental data on fission neutron cross sections of the major actinides are plentiful but often discrepant. At LANSCE we make precision measurements of these cross sections over 10 orders of magnitude in neutron energy, from sub-thermal to several hundred MeV. In addition to the major actinides, $^{235, 238}$U and ^{239}Pu, several minor actinides are being investigated and include ^{237}Np [5] and $^{240\text{-}242}$ Pu [6]. Parallel-plate fission chambers are used for this work. For neutron energies less than 100 keV, the fission cross section of ^{242m}Am is being measured in a different experimental setup at DANCE. Measurements on heavier actinides are in progress. Further, very precise measurements are planned with a Time-Projection Chamber discussed later in this paper.

3.3. Fission Neutron Spectra

An important characteristic of the fission process is the number and energy distributions of the emitted neutrons, which can serve as an indicator of illicit fissile material. We use the FIGARO array of neutron detectors [7] to measure the number and spectra of these fission neutrons. An example of the data for incident neutron energies between 2 and 3 MeV is shown in Fig. 1 for fission of ^{235}U and ^{239}Pu [8]. We are upgrading this array with a much larger number of neutron detectors and plan to measure not only the fission neutrons in a larger range of energies, 0.1 to 12 MeV, but also the correlation between the emitted neutrons. For most fission events two or more neutrons are emitted, and these neutrons should be correlated both in direction and energy. This type of correlated data is not in ENDF at present because of the nearly complete absence of experimental data.

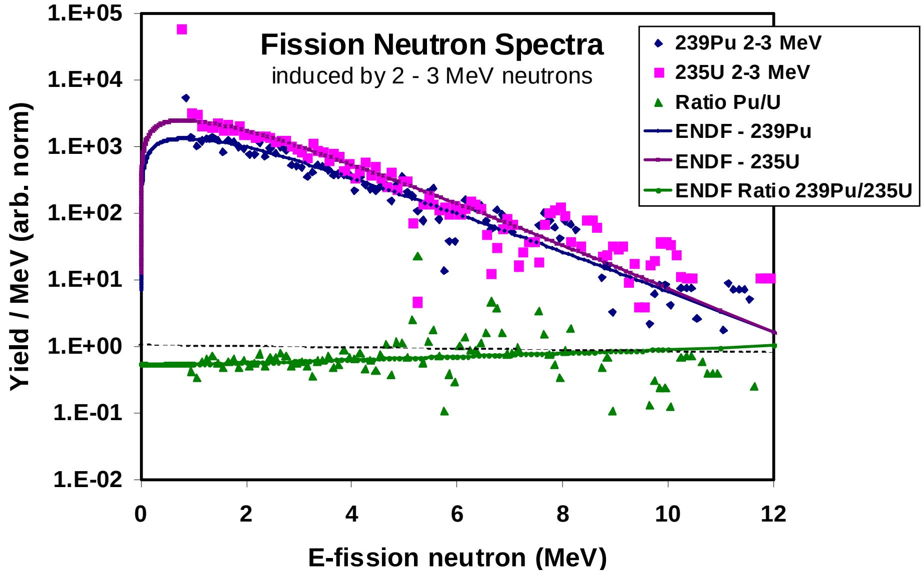

Figure 1. Spectra of neutrons from fission of ^{235}U and ^{239}Pu induced by incident neutrons of 2 to 3 MeV. The FIGARO data are compared in shape with the ENDF/B-VII evaluations. The ratio of the spectra are also presented [8].

3.4. Gamma-ray Production: Gamma-ray production by neutrons gives a unique signal of elements and isotopes. With the GEANIE array of high-resolution, high purity germanium detectors, these production cross sections have been measured at LANSCE now for many years, e.g. [9]. In addition to measurements of the production cross sections, new reference cross sections are being developed so that the ratio of production cross sections to a well-known cross section can be measured. Excitation of the lowest excited state of ^{56}Fe and ^{52}Cr are proposed as possible standards. This work is part of an International Atomic Energy Agency program to improve these reference cross sections [10].

3.5. Neutron Capture: This process depletes the neutron flux. With its resonances in the eV region, it can also be used to identify isotopes or elements in a sample. To measure capture cross sections, we use the Detector for Advanced Neutron Capture Experiments (DANCE) [11], a highly segmented array of 160 BaF_2 crystals arranged as an approximately 4π-calorimeter for neutron capture experiments including measurements of cross sections, gamma-ray multiplicities and capture-to-fission ratios. DANCE is located on a 20-meter beam line at the Lujan Center and uses neutrons from thermal to 200 keV. The high efficiency of the 4π-calorimeter coupled with the intense neutron beam allows measurements with milligram or even sub-milligram samples.

The efficiency of DANCE is approximately 95% for detection of a single gamma ray at 1 MeV. A neutron capture event usually produces several gamma rays, the pulse heights of which are summed in this calorimeter. If all of the capture energy is converted into gamma rays and into scintillation light, then ideally the summed pulse heights correspond to $\Sigma E_\gamma = Q + E_n$, where Q is the Q-value for neutron capture and E_n is the incident neutron energy corrected for the center-of-mass motion. In reality, some of the energy is lost and therefore the peak is broadened toward lower pulse heights. Summing the peak over an energy range down to half the peak still yields an efficiency for detecting capture of more than 70% when we consider also the effect of a ^{6}LiH spherical shell placed between the sample and the scintillators to absorb scattered neutrons Because of the high segmentation of the array, the multiplicity of gamma rays can be well determined from the number of detectors fired. For fission, the multiplicity of gamma rays is generally higher than that for neutron capture. Furthermore, for fission there is a distribution of the total energy emitted as gamma rays from the fission fragments. By placing windows on the energy and the multiplicity of gamma rays, the capture-to-fission ratio can be obtained [12]. An example of an application to nuclear forensics using the capture data is given below.

4. INTEGRAL EXPERIMENTS AND APPLICATIONS

The detailed nuclear data are needed for calculations that assess the effectiveness of proposed technologies to identify and quantify materials that a terrorist could use. To validate the calculations, integral experiments are essential. In this section, we describe some of these experiments and how they might be adapted to interrogation scenarios.

4.1. Interrogation with Neutrons

Neutrons can be used actively or passively to identify clandestine materials. The signature of these materials could be the transmission of neutrons through the cargo, delayed neutrons following fission, or gamma-rays produced by neutron interactions. The opportunity exists at LANSCE to test all of these approaches. Objects can be placed in the beam and the transmitted neutrons detected or even imaged. By moving the object, it can be scanned for gamma-ray production, neutron emission, and delayed neutrons.

Basic neutron transmission experiments with a single detector can validate calculations for transmission. In so-called “good geometry”, the transmission is easy to calculate from the total cross sections of the constituent materials. In “bad geometry” where the detector is close to the object, the calculations are more difficult and require good data on elastic and inelastic scattering. Gamma-rays and fission neutrons produced by a neutron -interrogating probe are additional tools for identifying the composition of an unknown object. The resonance structure of fast-neutron interactions in oxygen, nitrogen and carbon nuclei are particularly attractive for producing signatures of these elements.

If the detector is position sensitive, neutron imaging, either with fast neutrons or moderated neutrons, can be done in real time in the development of interrogation schemes. High specificity to certain materials can be achieved by imaging resonance-energy neutrons.

In the non-proliferation program, the LSDS has been proposed as a method to assess the amount and isotopic content of fissile material in spent fuel rods. The approach is discussed in the next section.

4.2. Assay of Spent Reactor Fuel

Spent reactor fuel rods contain significant quantities of fissile isotopes that must be tracked to insure that nothing is diverted for clandestine purposes. We are investigating methods for assaying these isotopes in spent fuel with a Lead Slowing Down Spectrometer (LSDS). A LSDS has been used at the Karlsruhe Nuclear Research Center [13] for the control of fabrication of fuel pins for light-water reactors. However, its use for complete spent-fuel assemblies has only been studied using Monte-Carlo simulation techniques [14-16]. Since LSDS has the potential to provide detailed information about spent fuel that other technologies are unable to provide [17], we are considering further details to see how well it can be adapted to practical applications, and what kind of accuracy and precision be obtained.

Detailed information on LSDS spectrometers is available in the literature [16-18]. In typical operation, a neutron source provides a pulse of neutrons close to the center a large block of lead. The neutrons scatter and as time progresses, slow down. There is a direct relationship between the time after the pulse t, and the average neutron energy E, given by

$$E = \frac{k}{(t + t_0)^2}$$

where t_0 and k are constants. This relationship is valid between a few microseconds and one millisecond after the pulse, corresponding to neutron energies approximately between 10 keV and 0.1 eV.

The fissile material content of a spent fuel assembly can be determined by embedding it in an LSDS. The assembly will emit fission-spectrum neutrons due to the fission process induced by the incoming neutrons from the slowing down process. As time progresses, the energy of these incoming neutrons decreases, and the number of fission events changes depending on the sum of the fission probabilities of each of the fissile materials in the assembly. Each fission event produces fission spectrum neutrons that can be counted, as a function of the time after the neutron pulse, in threshold fission detectors, detectors that are sensitive to high-energy neutrons only. The structure in the fission neutron function versus time curve, can be utilized to determine the concentration of each fissile isotope in the assembly. A typical fission detector could use pure ^{238}U-coated proportional counters that count the number of fission events induced by the high-energy neutrons in the ^{238}U.

We have simulated the response of the fission detectors, embedded in an LSDS, to a 0.5 MeV neutron pulse, with a starting position 35 cm from the center of the assembly, using the MCNPX [1] code.

In presenting these results, we employ a logarithmically increasing time-bin size throughout these analyses. Figure X shows simulated spectra for four combinations of fissile material (no fission products): ^{235}U only, adding ^{241}Pu, and then adding two different concentrations of ^{239}Pu.

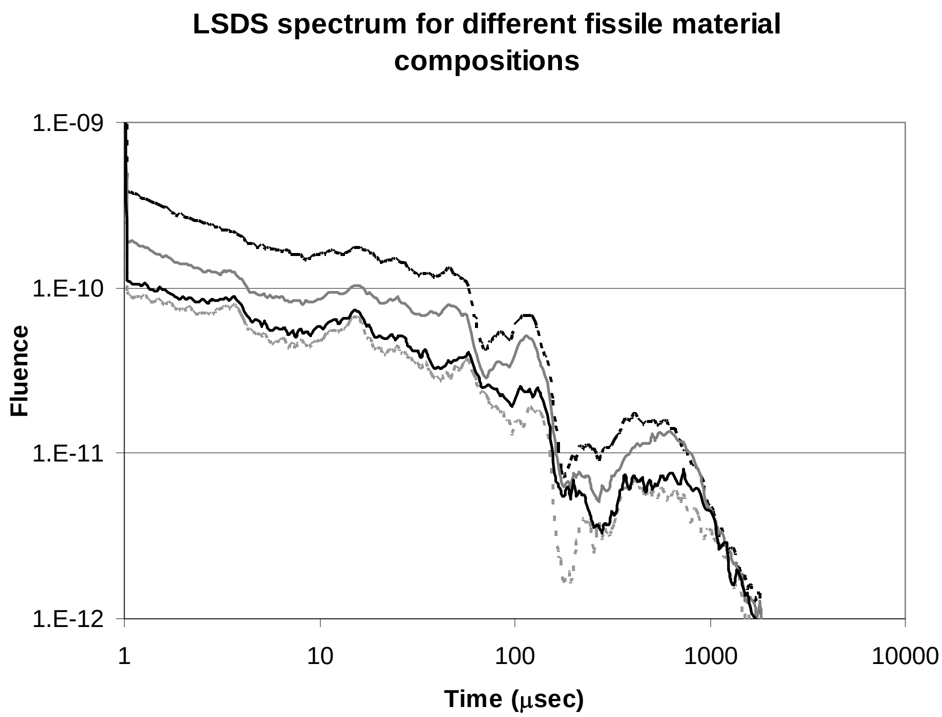

Figure 2: Time spectrum for different fissile isotope mixes in the assembly (fission products are excluded). Bottom (gray dots), – ^{235}U only (0.75%, no Pu), Next up (black line, includes ^{241}Pu 0.98%). Third up (gray line) adds 0.67% ^{239}Pu, and the top (black dots) has 1.47% ^{239}Pu.

The different spectra can be analyzed to determine the content of the major fissile isotopes to a high precision, possibly of the order of 1%. This is important information that can be used to investigate missing pins in the fuel, and other possible diversion scenarios.

4.3. Nuclear Forensics

In the event that an unexpected nuclear explosion occurs and the perpetrators are not known, it will be essential to gather as much forensic evidence as possible
is conducted by terrorists, it will be important to identify quickly the type of explosive, its isotopic content both pre- and post-detonation, and its performance. These characteristics will help point to the source of the nuclear material and to the perpetrators, against whom swift and decisive measures will be taken.

Radioactive debris from the explosion can give very useful information on the materials used in the device. This approach has been used since the beginning of nuclear weapons testing by adding certain elements, not generally found in the device or its emplacement, called “detectors” to the device and then measuring the amount of activation produced. The ratio of radioactive isotopes indicates the energy-dependent fluence. In a terrorist device, there will be no opportunity to add selected elements, of course, but certain isotopes are likely to occur. For example, ^{241}Am is formed by the decay of ^{241}Pu, which occurs in all plutonium material. Neutron capture reactions on ^{241}Am form ^{242}Am, and the ratio of these two isotopes can give information on the neutron fluence. The efficiency of the device is then indicated by the fluence, and that can point to the sophistication, or lack thereof, of the perpetrators. The recent data [11] taken at LANSCE have helped improve the data base for this reaction and this application.

4.4. Detector development, characterization and calibration

To test the response of detectors to neutrons, detectors can be placed directly in one of the neutron beams at LANSCE or in a beam of neutrons scattered from an organic scintillators where the neutron is tagged by the recoil proton. We prefer detectors with reasonably fast response times so that the incident neutron energy can be determined by its time of flight from the source. The full response of a detector to neutrons over a wide range of energies then can be determined in one experiment.

One of the experimental challenges is to perform these calibrations for detectors of greatly differing sensitivities. In the detection of clandestine material, the detector is often chosen to be as efficient as possible. For non-proliferation, the requirements can be the same or markedly different where the detector needs to have high specificity and sensitivity to characteristic radiations but insensitivity to others. Together with users from other LANL divisions and from outside organizations, we are calibrating traditional liquid scintillation neutron detectors, capture-gated scintillators, and inorganic scintillators where pulse-shape discrimination can discriminate neutrons from gamma rays.

5. UNCERTAINTIES AND COVARIANCES

After many decades of developing tools to use the uncertainties and their covariances in calculations, it is now beginning to be possible to calculate an effect in a system together with its uncertainty. The additional required data uncertainties are beginning to be included in data libraries such as ENDF. To advance these uncertainties from their form, which is rather primitive at present, a close collaboration is necessary among experimentalists, theorists and data evaluators. An important new direction for experimentalists at LANSCE is the discussion of the covariances in the uncertainties of our results. This activity involves discussions in depth with data evaluators in order to specify the covariances that are being entered into the evaluated data libraries such as ENDF/B-VII. The uncertainties including those of sample composition, impurities, dead time, neutron fluence normalization all need to go into the final covariance data files. An example of these covariances is given for ^{237}Np(n,f) in ref. [5]. This subject is discussed elsewhere in this conference.

6. FUTURE DEVELOPMENTS

Several upgrades are in process for the LANSCE accelerator and the experimental facilities. The LANSCE refurbishment project, LANSCE-R, will replace RF components which, in their aging state, are limiting the accelerator to 60 macropulses per second, half of the original value. The upgrade will also increase the reliability of beam delivery. There is also the possibility of longer macropulse lengths, which would increase the beam on target.

Another approach to increasing the beam on target for the WNR source is to accumulate the accelerator beam in the existing Proton Storage Ring (PSR) and then switched to the production target. The spacing of the micropulses can be varied according to the experimental requirements, and, in particular, with longer micropulse spacings than that are usually used, measurements can be made with neutrons down to keV or even eV energies. With additional PSR modifications, we expect the neutron production to increase by a factor of at least 5 to 10.

Improvements in detectors promise significantly improved data measurements and utilization of beam time. For fission measurements, a Time Projection Chamber (TPC) is being developed to identify the charge, mass, kinetic energies and angles of the fission fragments. With the knowledge of the angular distribution, corrections can be made to the observed fission cross sections for events near 90-degrees that are not detected because the fission fragments stop in the fissionable sample. This new approach should improve the standard ^{235}U(n,f) fission cross section. The FIGARO detector array is being expanded for a much greater coverage from its present 2% of 4-pi to 10% or even more. Not only will the data collection rate be improved by this factor, but other observables such as neutron-neutron correlations will be accessible for the first time.

7. USER FACILITY

LANSCE is a user facility. Many of the experiments are collaborations with groups from other national laboratories, universities and industry. The neutron sources are scheduled for approximately 7 months each year. Proposals for beam time are accepted once or twice a year and are reviewed by a Program Advisory Committee. Fast Track proposals can be submitted at any time. More information can be found on the LANSCE web page: http://lansce.lanl.gov.

8. CONCLUSION

Many applications for the non-proliferation and homeland security programs can be explored and developed with experiments at LANSCE. The flexibility of the neutron sources and their wide ranges in energy range and intensity, together with well-developed instruments and data acquisition capabilities, provide unique testing opportunities.

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