Detection of Highly Enriched Uranium Through Active Interrogation

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Abstract

We describe how Highly Enriched Uranium (HEU) can be detected through use of active interrogation probes, such as incident neutrons, or bremsstrahlung photons, followed by the detection of delayed neutrons or gamma rays. These delayed particles are a signature for the presence of special nuclear materials. We describe the development of radiation transport simulation tools (the MCNP code) together with underlying databases that are needed to simulate these processes, and to support experimental detector development work at Los Alamos. We describe the underlying physics advances in modeling and representing the fission process, to allow us to accurately simulate these processes. These capabilities allow the determination of which isotopes are present, and can also tell us about the amount of material present.

1. Introduction

Detection and characterization of special nuclear material (SNM) is central to several programs related to homeland security, nuclear material safeguards, nuclear criticality safety, and emergency response. The initial objective is often to determine the presence or absence of SNM in an unknown object using non-invasive means. If SNM is determined to be present, then there is often a need to characterize the material by inferring the type of SNM, the mass, and the configuration (geometry). Many such applications are complicated by shielding and the competing needs for speed and accuracy.

One method used to determine the presence of SNM is to detect radiation emitted from the material. Passive detection techniques rely on observing intrinsic radiation emitted by the SNM, such as neutrons from spontaneous fission or (α,n) reactions, or characteristic photon lines.

Passive detection is more challenging if the SNM is heavily shielded. In addition, the effectiveness of passive detection can depend on the type of SNM. For example, the spontaneous fission source strength from weapons-grade Pu is ~ 65 n/s/g, whereas highly enriched uranium (HEU) has a spontaneous fission source strength of only ~ 1.e-03 n/s/g. Furthermore, passive detection of photons from HEU suffers from the fact that most HEU photons are low enough in energy that they are easily shielded. Thus, detecting shielded HEU is often portrayed as the “Grand Challenge” of this field.

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An alternative to passive detection is to employ active interrogation. Active detection relies on external radiation sources that are used to interrogate the unknown object and induce a response of some sort. This response is then detected and analyzed to characterize the SNM (if any) contained in the object.

In the next section of the paper we will list various active detection techniques and go into some detail on two active detection methods being pursued at Los Alamos National Laboratory (LANL). We will follow with a discussion of simulations needed to support the development of these techniques, with a focus on required nuclear data enhancements. We will then describe benchmark experiments that have been performed using active interrogation of HEU and compare with preliminary simulation results. Finally we will summarize and indicate future needs and directions.

2. Active Interrogation

Many options have been proposed for active interrogation and detection. In particular, various combinations of neutrons and photons as source radiation and detected radiation are being studied.

One example, nuclear resonance fluorescence [Reference 1] relies on the absorption of source photons in well-defined narrow resonances specific to individual isotopes to result in “holes” in transmitted photon spectra corresponding to the characteristic resonance energies of materials that are present. Another method [Reference 2] is to detect β-delayed high-energy gamma rays emitted by fission products following pulsed neutron interrogation.

Rather than describe all possible active interrogation techniques, this paper will focus on two such techniques being pursued at Los Alamos. The pulsed source radiation is either 14-MeV neutrons or bremsstrahlung photons. In either case, delayed neutrons from fission create an artificial intrinsic steady-state source of neutrons distributed throughout the fissile material. This neutron source is then used to “re-interrogate” the material. These delayed neutrons and prompt neutrons from fission events induced by the delayed neutrons are detected between pulses. Noise-analysis techniques that have been previously employed for passive plutonium systems are then used to analyze the response.

2.1 Photon Interrogation

A Linatron 2000™ linear electron accelerator has been used at Los Alamos as the active source of interrogating photons. The accelerator can be operated at 6, 8, or 10 MeV with a pulse rate between 20 and 120 Hz. For the experiments analyzed in Section 4.1, the accelerator was operated at 10 MeV with a pulse rate of 50 Hz. The pulse width was 4.5 μsec and the dose rate at 1 meter was approximately 160 R/min. See References 3 and 4 for more details of the photon interrogation methodology. A picture of the accelerator is provided in Figure 1.
2.2 Neutron Interrogation

Los Alamos generally employs 14 MeV neutrons from the d+t reaction as the active source of interrogating neutrons. An MF Physics model CC A-211\textsuperscript{TM} neutron generator is used. The pulse rate is 50 Hz and the pulse width is approximately 10 $\mu$sec. The output is $10^6$ neutrons per pulse. A picture of the neutron generator is shown in Figure 2. See References 3 and 5 for more details.

2.3 Neutron Detection

Various neutron detection systems have been designed and used. All rely upon multiple $^3$He detector tubes that are moderated by polyethylene and shielded by cadmium. Details of individual detection systems are largely driven by the intended application; for example, if portability is required then obviously size becomes a constraint. An example of a large area (4’ by 8’) neutron detection system is shown in Figure 3. This particular system has 4 $^3$He tubes and a 10% detector efficiency. A more portable neutron detection system is partially seen in Figure 4 behind and to the right of the barrel. Further details may be found in Reference 6.
2.4 Data Acquisition and Analysis

The data acquisition employed is the custom Los Alamos-designed PATRM system [Reference 7]. The time history of neutron events is analyzed for correlations. Various implementations of Feynman’s Variance-to-Mean technique are used to analyze the results to determine properties of the multiplying sub-critical system. See Reference 5 for more details of the analysis methods.
3. Simulation

A rich history of simulation / experimental synergy has been demonstrated in support of passive detection schemes. Likewise, simulations also play an important role in the development of active interrogation systems to detect HEU. Simulations are cheap, relative to experiments, and can be used to cover a much larger phase space. They can be used for risk analysis and cost-benefit systems studies. Particular roles of simulation include:

- Enabling experiments (dose rates, criticality safety, authorization basis, …)
- Optimizing system design (active source, detector, …)
- Providing insight into operational protocols (necessary count times, false positives, …)
- Answering “what-if” questions (shielding, background, …)
- Aiding experimentalists in interpreting data (insight into physics)

We consider a simulation tool to be the superset of a radiation transport code and the required nuclear data. At Los Alamos we principally rely on MCNP [Reference 8] as the transport code and ENDF [References 9, 10] for nuclear data. MCNP is a continuous-energy Monte Carlo code that supports generalized three-dimensional geometry for neutral- and charged-particle transport. ENDF provides peer-reviewed and tested evaluations for incident neutrons, photons, and charged particles. For use in MCNP, ENDF evaluations are processed into ACE format using the NJOY processing code system [Reference 11].

Despite the powerful features of ENDF evaluations and the MCNP code, neither of the two active interrogation techniques described in the previous section could be fully modeled by Los Alamos in ~ 2000. Some of the most notable limitations were:

- No photonuclear evaluations in ENDF or transport capability in MCNP
- No detailed modeling of delayed neutrons from fission in MCNP
- No correlations defined among secondary particles from fission in ENDF or MCNP

Many of these limitations have now been removed. We will describe some of the relevant improvements made in our simulation capabilities over the past several years in the remainder of this section.

3.1 Neutron-Induced Fission Delayed Neutron Modeling in MCNP

Versions 4B and earlier of MCNP created the correct number of fission neutrons from neutron-induced fission in a steady-state problem (i.e., using the total \( \nu \)) but did not differentiate prompt and delayed fission neutrons by energy or time spectra. More recent versions of MCNP and its associated data libraries (beginning with 4C) correct this by sampling energy and temporal distributions appropriately (i.e., per specification in evaluations) for delayed fission neutrons. For more details of this capability, including validation calculations, see Reference 12.

3.2 Photonuclear Evaluations in ENDF and Transport in MCNP

Substantial efforts at Los Alamos have been expended toward implementing a photonuclear simulation capability in MCNP. First, Los Alamos chaired an IAEA Coordinated Research
Project [Reference 13] that produced 160 ENDF-format photonuclear evaluations, based on experimental data and GNASH model calculations. Particularly important to note is that these evaluations included not only integrated cross sections for the various photonuclear reaction channels, but also secondary energy and angular distributions for neutrons and other particles [Reference 14]. The latter data, of course, are required by a transport code. MCNP was also extended to use data derived from these photonuclear evaluations. A complete description of this capability may be found in Reference 15. Sample validation results are shown in Figure 5.

![Figure 5. Calculation vs. experiment of neutrons per electron on Pb as a function of energy and sample thickness.](image)

### 3.3 Photofission Delayed Neutron Data

The initial implementation of photofission data in ENDF and MCNP considered prompt fission neutrons only. As discussed previously, active detection schemes often rely upon re-interrogation of the material by delayed fission neutrons. To provide this functionality in our simulations, we first added photofission delayed neutron multiplicity ($\nu$) and energy distribution data to the ENDF evaluations described in the last paragraph. A private version of MCNP was created to utilize these data. See Reference 16 for more details. Results from simulations of benchmark experiments using this capability are described in Section 4.1.

### 3.4 Fission Multiplicity Distributions in MCNP

Sophisticated neutron detection systems often rely upon correlations in signals. For example, the analysis of a multi-detector system might “score” counts only if more than one individual detector had a signal within a certain time of each other. Simulation of such sophisticated detection schemes requires careful consideration of correlations among secondary particles emitted by physical processes such as fission.

Previous production versions of MCNP emitted a bounding integer number of neutrons from fission. For example, if $\nu$ was 2.6, then 60% of the time 3 neutrons were emitted and 40% of the time 2 neutrons were emitted. In reality, the number of neutrons emitted from such a fission event can range from 0 to many (perhaps 8 or 9).

Recently, the capability was incorporated in MCNP Version 5.1.40 to sample from a
probability distribution for the number of secondary neutrons from fission, the so-called p(v) distribution. The average number of neutrons is preserved. See Reference 17 for more details of this capability.

3.5 Calculations of Correlations Among Secondary Particles from Fission

In the future we would like to include additional correlated data in our simulations. As a first step, Monte Carlo simulations of fission-fragment statistical decay by sequential neutron emission followed by $\gamma$-ray emission have been carried out. Average neutron and $\gamma$ multiplicities and energy distributions are obtained from this work. In addition, however, correlations among neutron and photon energies are also calculated. An example showing the resulting neutron energy spectra as a function of the number of neutrons emitted from fission is shown in Figure 6. Full details of the theory and calculations may be found in References 18 and 19.

![Figure 6. Calculated neutron energy spectra as a function of number of neutrons emitted from fission (Ref 18).](image)

3.6 Evaluated Photonuclear Data for Actinides

In the IAEA Coordinated Research Program, the original actinide cross-section evaluations came from Obninsk, Russia. Because these evaluations did not include delayed neutron information from photofission, delayed neutron data were added to the Obninsk evaluations by Los Alamos (see Section 3.3).

Recently at Los Alamos, in collaboration with CEA Saclay, we have evaluated the actinide photonuclear cross sections for $^{235,238}\text{U}$, $^{239,240}\text{Pu}$, $^{237}\text{Np}$, and $^{241}\text{Am}$. The GNASH modeling code was extended to model the photofission process. We were able to make extensive use of GNASH nuclear reaction modeling parameters already developed for our work on neutron reactions (e.g., fission barriers, level densities, etc.). This enabled us to use our more advanced computational tools for predicting exclusive cross sections, spectra, and angular distributions for the emitted neutrons [Reference 20].

Our evaluations for the prompt fission neutron multiplicity were based on measured data from Livermore. See for example, in Figure 7, the data and evaluation for photofission of $^{235}\text{U}$. Also shown in the figure are the results that would have been obtained if one took the ENDF neutron evaluation for the A-1 system shifted by the neutron separation energy, since one would expect these results to be similar. It is reassuring that indeed the two approaches lead to similar results.

For the photofission delayed neutron multiplicities, our approach has been to utilize the
equivalent neutron induced evaluations for the A-1 system (shifted by the neutron separation energy), but then to renormalize these values so as to better match the measured data from Caldwell. See Figure 8 for the results for $^{235}\text{U}$. Note that a measurement program has been initiated at CEA/Saclay to obtain new data for delayed neutron multiplicities [Reference 21].

Figure 7. Evaluated prompt neutron multiplicity from $^{235}\text{U}$ photofission.

![Prompt Nubar - $^{235}\text{U}+\gamma$](image)

Figure 8. Evaluated delayed neutron multiplicity from $^{235}\text{U}$ photofission.

4. Benchmarks

4.1 Bremsstrahlung Photon Interrogation

Experiments using the bremsstrahlung photon active source interrogation technique described in Section 2.1 were performed at the Los Alamos Critical Experiments Facility (LACEF). HEU shells (the so-called Rocky Flats shells) were arranged in two configurations. The first had a mass of 4.8 kg and the second had a mass of 21.5 kg. The accelerator was pulsed at 50 Hz with a 10 MeV endpoint energy. Approximately 100 counts per second were observed with the small-mass HEU configuration, and approximately 1000 counts per second were observed with the large-mass HEU configuration. The count rates were well over background rates. Singles and
doubles were clearly observed.

The capabilities described in Sections 3.2 and 3.3 were used to simulate the experiments. While absolute comparisons with experiment were not practical, the relative agreement was encouraging. The experimental count rate ratio (large object / small object) was 10.0 and the simulated value was 8.7. See Reference 16 for more details.

4.2 14-MeV Neutron Interrogation

Also at LACEF, the techniques and apparatus described in Section 2.2 were used to interrogate bare configurations of HEU with 14 MeV neutrons, again using the Rocky Flats hemispheres. Four configurations, ranging from 13.7 to 27.4 kg of HEU were measured. The neutron multiplication of the configurations was determined using the analysis methods described in Section 2.4. The neutron multiplication was also calculated with MCNP. Results are shown in Table I. Agreement between experiment and simulation is quite good.

Table I. Experimental and simulated neutron multiplication from 14-MeV neutron interrogation of 4 configurations of HEU.

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<tr>
<td>RF1-30 [22.4 kg]</td>
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<td>0.05368</td>
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5. Summary

Detection and characterization of SNM, in particular, HEU, often can be enhanced by using active interrogation. We have described 2 active interrogation techniques being studied at Los Alamos, using either bremsstrahlung photons or 14 MeV neutrons. Both techniques rely on delayed neutrons from fission to create an artificial steady-state intrinsic neutron source in the object being analyzed. This neutron source is then used to re-interrogate the object. Noise-analysis techniques that have been previously employed for passive plutonium systems are then used to analyze the response.

Simulation of active interrogation techniques places new requirements on nuclear data evaluations and radiation transport codes. Several enhancements to data and code physics have been described that allow us to model aspects of these technologies with reasonable agreement compared to benchmark experiments.
Additional work is required for full fidelity simulation capability. For example, theoretical work to determine detailed correlations among secondary particles from fission has been carried out and we need to incorporate results from this work into our data bases and transport codes. In addition, initial focus at Los Alamos has been on delayed neutrons from fission; we would like to extend this to include delayed photons from fission as well. Finally, improvements in the accuracy of integrated and differential cross sections based on the forthcoming release of ENDF/B-VII [Reference 22] need to be made available to the MCNP user community.

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