US Nuclear Data Program
Task Force on Nuclear Data for Homeland Security

Nuclear Data Needs for National Homeland Security Program

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Introduction

One of the tasks addressed by the USNDP Task Force on Nuclear Data for Homeland Security was to develop a needs list for new nuclear data and new database capabilities. Through an informal survey of homeland security technical programs at LLNL and LANL and input from the US Nuclear Data Program community we have developed this document. Only items requested by currently funded research and development projects are listed; no attempt has been made at this point to prioritize these needs.

There appears to be two main program drivers for this needs list:

- Detection of radiological and nuclear materials being transported into or through the US or its concerns.
- Monitoring, detection, and analysis of nuclear explosions and nuclear weapons proliferation through radionuclide monitoring and other detection capabilities.

Some related aspects of these programs drivers, including international treaty negotiations and emergency response, also have a few data needs.

Projects under these two program umbrellas typically encounter nuclear data issues when variant detection schemes or event scenarios are modeled using sophisticated simulation codes such as MCNP(X) or GEANT. These simulations are used to lay the groundwork for proposing and planning new projects and also to optimize the design or analysis of different configurations. Many calculations can be performed quickly, whilst individual experiments involving SNM require extensive authorization and are costly. Simulations of fielded experimental interrogation configurations can be used to interpret the measured data. And very importantly, simulations can extensively explore "what if?" questions.

The simulation capabilities are built upon high-quality fundamental nuclear cross section and decay databases, in the ENDF nuclear data library. These evaluated databases incorporate the detailed information available from experiments and from nuclear models, and allow transport simulations to model the underlying physical phenomena accurately. Several of the projects surveyed had encountered the need for advances in simulation methods and in the underlying ENDF library. These needs range from particle correlations in energy, angle and multiplicity to improved data for photonuclear reactions to improved cross sections for neutron reactions involving unstable or rare isotopes to improved gamma-ray production data.

There were also many projects surveyed where nuclear decay, reaction and structure libraries as well as specific references were consulted to address or explore issues and ideas analytically. The resources used included the ENSDF nuclear structure library and NSR references library, and these resources were typically accessed through a variety of web-based dissemination projects -- the usefulness of these resources depends on the ease of use and the completeness and accuracy of the information present. In many cases the ENSDF and NSR resources were used as input to calculations to generate data for transport simulations or to generate physical constants used to assay a sample. Several of the projects surveyed indicated a need for better decay half-life and branching ratio information in specific cases, e.g. the spontaneous fission half-life of ^{240}Pu, and some had plans to make measurements to acquire the information they were missing, or to benchmark the data that is presently available in ENSDF and ENDF.

Correlated particle information from fissile materials

The timing data stream of neutron counts contains information that can be used to determine properties of the source of neutrons. For materials that support fission chains, a random event that spontaneously creates neutrons, such as a spontaneous fission or an (alpha, n) reaction, is followed by a correlated number of neutrons emitted by the fission chain. The rate of spontaneous fission events is proportional to the amount the spontaneous fission isotope. The length of the fission chain is related to the system multiplication. The number of neutrons emitted in an individual fission satisfies a probability distribution that is approximately Gaussian, with typically about three neutrons emitted on average, but with a reasonable probability that no neutrons are emitted or as many as eight neutrons are emitted. (The nuclear data for the neutron distributions has been tabulated by Holden and Zucker from Brookhaven.) In a multiplying medium this intrinsic fluctuation for each fission is amplified for a fission chain, with a very high probability the chain creates many more than the average number. The large fluctuations in the number of neutrons are a great advantage for detection and for assay. Similar large fluctuations in the number of fission gamma rays emitted are also likely to be advantageous.

Gamma ray timing also carries information about the fission chain. First, when neutrons are absorbed in an (n, gamma) process, the gamma continues the same information about the fission chain as the original neutron out from the absorber, especially as these are penetrating high-energy gammas. The timing of the (n, gamma) neutrons also carries the same information about the moderation process (~ 100 microsecond time scale) as the original neutrons. In addition, each fission in the chain emits a burst of gamma rays, the entire fission chain gamma cascade being prompt (~ 100 ns) compared to the moderation time scale.

Projects involving detection or assay schemes using particle correlations have lead to the following new data needs:

i) The probability distribution for the number of prompt gammas created by an individual fission

While the average number emitted is known it is the actual distribution is needed. Given the large number of decay channels the final distribution is likely nearly Gaussian by the central limit theorem, but the width is not known.

ii) The correlation between the number of emitted neutrons and gamma rays

When very few neutrons are emitted by a fission event, do there tend to be more gamma rays emitted? When there are eight neutrons emitted, is it more likely that there are fewer gammas emitted?

iii) Energy - number correlation for gamma rays

When many gammas are emitted, do there tend to many soft gammas after hard gamma emission? The gamma energy correlation is important because of the energy dependence of penetrability.

iv) Delayed gamma distributions

The fission fragments will beta decay on second time scales, long compared to diffusion time scales, but there will tend to be cascades. There will also tend to be a series of beta decays, each with a different cascade. How do the number distributions of the cascades change in the subsequent beta decays, both in number distribution and energy distribution?

v) Non-fission gamma cascade number and energy distributions

These cascades could, for example, follow (alpha, n gamma), (n, n' gamma) or (n, gamma). All of these processes emit multiple gamma ray bursts, on time scales short compared to time between subsequent scatterings in an event chain. Are there other time scales from metastable states? The gamma rays from these correlated processes are statistically distinguishable from potentially large gamma ray backgrounds, both from the environment and from the alpha decay chain gammas. It is especially the large fission chain fluctuation bursts that contain the most information.

Gamma-ray production information

The gamma-ray lines emitted in radioactive decay or as an excited nucleus decays to its ground state provide a unique, characteristic signature of the decaying element or isotope. Provided that there is an external probe that can broadly induce such radioactive decay, such as a neutron or photon source, this information can be used to detect or assay materials. The rate of photon production is proportional to the amount of material being irradiated folded with the energy-dependent cross section for inducing the radioactive decay signature. The detection probability depends on the attenuation of the characteristic gamma-ray lines in surrounding absorbing material and the rate of background photon production. Detection works best when (1) the absorbing material has a low atomic number, e.g. hydrogenous materials, (2) the characteristic gamma-ray lines are higher in energy, and (3) there is a time or energy dependence to the signal which distinguishes it from background photons. For example, the detection of delayed gamma rays from fission with energies above 3 MeV has been proposed as a detectable signature of fissile materials interrogated with external sources. Assay works best when a detectable signal exists from all elements present. For example, the neutron capture gamma-ray spectrum is starting to gain favor as an assay method in a technique known as Prompt Gamma Ray Activation Analysis.

Projects involving detection or assay schemes using gamma-ray spectroscopy have lead to the following new data needs:

i) Delayed gamma-ray energies and half-lives from fission with half-lives ≈ 0.5 s and $E_\gamma > 3$ MeV

Data in this half-life range is sparse. However, if there is a penetrating radiation with significant yield at times greater than 100 ms, then it would effect design considerations for detection schemes for fissile materials.

ii) High resolution (≈ 1 keV) gamma-ray spectra from neutron capture ($E_n < 50$ keV) on all naturally-occurring materials

High-resolution spectroscopy can be used as an assay tool that is relatively broad based in that most elements except helium have a naturally occurring isotope with significant gamma-ray production following neutron capture.

iii) An event generator to source spectrally correlated gamma rays from decay cascades following neutron capture and other reactions

Investigations of detector response and backgrounds caused by naturally occurring or external neutron sources often require one to conserve energy and spectral shape on an event-by-event basis. This capability is generally not available in traditional transport codes.

iv) Improved representation of neutron scattering and subsequent photon production in germanium detectors, particularly inelastic scattering leading to "neutron bumps"

There are several instances where a promising signature is co-located with neutron bumps in germanium detectors. A better model of the processes involved will allow engineers to consider designs to minimize the interference via simulation.

v) Complete gamma-ray spectra from proton and alpha induced reactions

In some cases, these data were needed in order to model background processes. For example, alpha decay can sometimes result in an alpha-induced reaction from which a photon cascade is emitted. In other cases, the secondary photon spectra are used as a probe. This is discussed in more detail in the next section. It should be noted, that induced photon lines from alpha-induced reactions often have hard-to-model line shapes due to kinematic recoil, which need to be characterized for some applications.

Photonuclear data

Photon sources offer some possible advantages over neutron sources for active interrogation schemes: (1) the source can double as a radiograph source and (2) photons are more penetrating than neutrons for hydrogenous cargos. These advantages have led to some initial work to model photonuclear processes in transport simulation codes. For instance, new capabilities have been developed to model photonuclear and photofission reactions in MCNP(X), with an accompanying development of evaluated photonuclear cross section databases. A first demonstration capability was developed, and some initial comparisons with validation experiments were successful. However additional research is needed to improve the simulation tools. These include:

i) *Development of photonuclear data for $\gamma + {}^{235,238}U$ and ${}^{239}Pu$*

ii) *Photofission delayed-neutron probabilities, energy spectra, and time-dependences for the delayed neutrons*
With photon sources, the compound nucleus that fissions is different from neutron sources. Delayed neutron probabilities and energy spectra can vary widely from isotope to isotope.

iii) *Development of nuclear data to support interrogation methods for conventional and nuclear explosives, using resonant photonuclear absorption on nitrogen and SNM.*
There are two issues here. First, these processes are not included in transport simulation codes because the data is not part of the standard evaluated data files (ENDF) that get incorporated into transport simulation codes. In most cases, however, the known resonances and subsequent decay patterns are tabulated in ENSDF. The second issue is that many of the resonances that would be most useful from the viewpoint of a practical system have never been measured. For example, *(γ,γ')* resonances in ${}^{235}U$ have only been tabulated up to excitation energies of 1 MeV.

There are some data needs in the area of photon source development based resonant processes that should eventually be addressed. One scheme is resonance absorption of approximately 9-MeV photons prom p + ${}^{13}C$ in $1^{4}N$. The other scheme is Argonne FIGARO approach in which 6-7 MeV photons from ${}^{19}F(p,\alpha\gamma)$ are used to interrogate for SNM via (γ,n) and photo-fission processes. In both of these areas, protons are incident on target materials. Data on protons with energies below 5 MeV (higher energies are impractical because the accelerator would be too large) incident on thick stopping targets of various materials are very sparse. There are some thin-target data available, but experience shows that trying to fold incomplete thin-target data with stopping power information to generate effective thick target results is generally quite inaccurate and thus unproductive for applications. A systematic study involving a number of potentially important target materials could be readily accomplished at a small accelerator facility. Furthermore, improved *(γ,n)* data on such materials as deuterium has also been requested since these processes can lead to neutron background problems in some of the proposed interrogation schemes.

Neutron cross sections on unstable and rare isotopes

One national goal is the ability to analyze chemical, biological, and nuclear materials, assemblies and/or debris and identify the origin and user of these materials. Radio-chemical signatures can often facilitate forensics work. Accurate nuclear cross sections are needed in this program, many involving nuclear species off stability that are hard to measure directly. The current focus with relevant nuclear data needs is on the development of new forensics signatures of actinide materials and debris. This is motivating some challenging nuclear theory and evaluation projects, and some new measurement efforts.

Current needs include:

i) *Improved neutron-induced fission cross sections among U, Np, Pu, Am, and Cm; both long-lived and short-lived species are of interest, and these entail measurements and reaction modeling for $E_n < 20$ MeV.*

ii) *Improved neutron-induced capture cross sections on long-lived and short-lived actinides, with similar comments as above.*

iii) *Accurate (<10%) (n,2n) cross sections for ^{235,23,}U, ^{239,240}Pu and ^{241}Am across the energy range of interest, and in particular within 1 MeV of thresholds.*

iv) *Accurate estimates of evaluated cross section uncertainties, including model uncertainties.*

Other data needs

Neptunium is of concern as a proliferant material, as it is created in nuclear reactor waste. There is a need to better determine neptunium critical mass to guide non-proliferation activities. Values reported in the literature for the critical mass vary widely, from 50 – 80 kg. Using different nuclear databases (e.g. ENDF/B-VI, ENDF/B-V in the US, JENDL in Japan, etc) in radiation transport simulations gives calculated critical masses that vary widely, reflecting difference in the underlying evaluated nuclear cross sections.

Feedback from emergency response personnel and the transport security administration indicates that they desire a user-friendly catalog of radioactive decay signatures. The catalog should include their basic properties, e.g. half-life, decay modes, and decay radiations. The method of production and the industry utilizing the source should also be listed. A shorter list of nuclides of common availability with their basic properties should be available in easy to handle formats, e.g. an electronic file, a poster, a wallet card or credit-card sized pocket reference.

In fact, the natural backgrounds on the surface of the earth is of broad concern in the community for two reasons: (1) these interferences need to be understood so as not to be confused with other weak signals, and (2) in one case there is interest in using cosmic muons as a probe for a detection system. About a third of the natural radiation background originates from cosmic rays. Cosmic rays are usually highly clustered in space and time, and much of the natural background is not purely Poisson in its time distribution. The natural time and spatial variation of environmental background radiation is not well understood and is difficult to model. We have little data how fast natural radiation can change due to rain, thunderstorms, wind blown duct etc. In the case of muon-based systems, it was noted that muon-induced fission in heavy elements, and the time and energy distribution of the reaction products is poorly known.