PROMPT GAMMA RESPONSE SIMULATIONS FOR NUCLEAR FORENSIC APPLICATIONS

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ABSTRACT

A simulation package has been developed to aid in the characterization and identification of unique nuclear material signatures throughout the nuclear fuel cycle. This work is focused on predicting a sample response to neutron activation analysis (NAA), which will be performed using the mechanical time-of-flight (TOF) system that has been designed for the Pennsylvania State Breazeale Reactor. This TOF system will use epithermal neutrons for NAA and allow for isotopic material analysis with significantly greater sensitivity than conventionally used NAA based on thermal neutrons. This project aims at predicting the prompt gamma response of samples interrogated with epithermal neutrons. A prompt gamma response can be correlated with a delayed gamma response resulting from an irradiation of a sample and will complement the non-destructive analysis capabilities afforded by epithermal NAA. Simulations of prompt gamma responses are performed with DICEBOX, an open-source Monte Carlo code, which simulates gamma cascades from a nucleus using statistical models of level density and photon strength functions. The purpose of the simulation package is to create an easy-to-use interface for a user to input material information of a given sample and assemble a DICEBOX input file. The package extracts necessary information such as internal conversion coefficients from external databases, selects the appropriate level density and photon strength function models, and runs the file to simulate the prompt gamma response. The resulting gamma signatures may be used to make inferences about samples' material processing history or provenance for nuclear forensics characterization. Detection of rare-Earth elements and analysis of radiochronometric signatures are of particular interest and the use of epithermal neutrons in NAA will utilize elevated isotopically sensitive cross sections at resonance energies and allow for detection of trace signatures.

INTRODUCTION

The principal objective of this work is to develop improvements for techniques to identify unique forensic signatures related to the nuclear fuel cycle. For this purpose, a mechanical TOF neutron chopper system has been developed for use at Penn State's research reactor [1]. This chopper system will allow neutrons to be isolated along a beamline within the energy range of 0.5-40 eV and with energy resolution of 2% (0.8 eV at 40 eV). These neutrons can then be used for epithermal neutron activation analysis (ENAA) to perform non-destructive material assay. The precise neutron energy tunability afforded by the system will allow for neutron capture resonances present in many materials in this epithermal energy range to be exploited, significantly increasing the macroscopic neutron capture cross section and subsequently boosting the resulting gamma signal. Typically, NAA is performed to measure the delayed gammas emitted by activated nuclei upon their radioactive decay. In this work, the feasibility of also utilizing the prompt gamma signal

is investigated. Upon neutron capture, a nucleus will immediately deexcite via the emission of prompt gamma rays in a cascade. Similar to the delayed gammas, these prompt gammas are unique for each isotope [2]. It is postulated that analysis of prompt gamma spectra from ENAA may prove to be a useful technique for characterization of nuclear fuel cycle materials and may offer insight into materials' geologic provenance and processing history. Before any experimental trials are performed, simulations of neutron capture events on a variety of nuclei can be performed to estimate prompt gamma yields.

At the core of the simulation efforts is DICEBOX, a Monte Carlo code originally developed by F. Bečvář, and updated by M. Krticka and S. Valenta [3]. This code is open-source and is available through the IAEA's Nuclear Data Services (NDS). DICEBOX is designed to simulate gamma decay from an excited nucleus for which individual levels may not be well-defined. For many isotopes, experimental data only exist for levels up to 2 or 3 MeV. Upon neutron capture, most nuclei reach an initial excited state with energy ranging between 6 and 10 MeV [2]. DICEBOX serves to simulate the prompt gammas, which are not well defined or well resolved, from the upper energies of the known levels to the initial excitation energy upon neutron capture. The simulated prompt gamma response from DICEBOX can be correlated to known experimentally acquired data up to 2 or 3 MeV, such as the data available in the IAEA's Prompt Gamma Activation Analysis Database, to obtain a more complete picture of the expected prompt gamma response. Furthermore, the prompt gamma response can be adjoined to the expected delayed gamma response. Together these two data sets give a more complete representation of the total expected gamma yield when NAA is performed for a given sample.

TECHNICAL APPROACH

Each DICEBOX input file is relatively dense, requiring data from several separate sources, which then must be copied into the file and formatted correctly. Because all the required data are isotope-specific, each prepared DICEBOX input file is also isotope-specific. In order to simulate the prompt gamma response from a hypothetical elemental sample, which is comprised of five separate isotopes, five separate input files would need to be prepared, starting nearly from scratch with each one. A software package has been developed as a (sort of) wrapper around DICEBOX, with one key goal being to aid in the construction and running of these input files by automating as much of the input file assembly as possible. This software package is referred to as the Prompt Gamma Response Simulator (PROGRESS). PROGRESS is essentially divided into two main responsibilities: the assembly of the input file, and the interpretation of the output data file from DICEBOX. The structure of PROGRESS is shown in Figure 1. Data required for the DICEBOX input file, which the assembly package of PROGRESS automatically collects and writes into the file, can be categorized into one of three data blocks: statistical parameters, internal conversion coefficients (ICCs), and known level data.

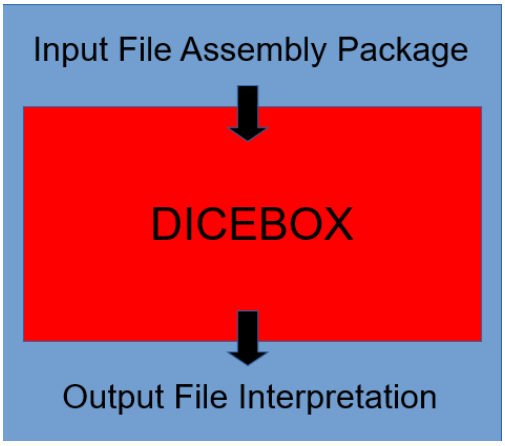

Figure 1: Flowchart structure for PROGRESS, showing how the two main components interface with DICEBOX.

Operation of DICEBOX

Statistical parameters required to run DICEBOX include standard parameters involved with performing any Monte Carlo simulation, such as number of events per cycle (for DICEBOX, this is the number of simulated prompt cascade-inducing neutron captures), the desired number of simulation cycles, etc. Other parameters included in this block include the selections of nuclear level density (NLD) model and photon strength functions (PSFs). DICEBOX comes already equipped with several choices for NLD model and PSFs and allows the user to install and use a custom model if so desired. Separate PSFs may be selected for each of the E1, M1, and E2 transitions. Each of these parameters is not inherently isotope-specific, and so each of these parameters may remain unchanged between multiple different input files. Regardless of these choices, however, any selected NLD model and PSF requires a few isotope-specific coefficients. Whereas a user would normally need to seek out these coefficients and copy the correct ones into the input file on their own, PROGRESS automatically finds the correct values in a published database of experimentally determined coefficients [4]. All user selections and relevant coefficients are then exported and written into the input file with correct formatting.

Internal conversion coefficients (ICCs) give the proportion of the probability of internal conversion electron ejection to the probability of gamma ray emission. These coefficients directly govern the statistical behavior of prompt gamma emission from any nucleus. ICCs are primarily influenced by the density of electron shells surrounding the nucleus as well as the magnitude of the nuclear deexcitation energy. For use with DICEBOX, ICC values will be imported from BrIcc, a software package available through the Australian National University [5]. Specifically, for integrated use within PROGRESS, the standalone executable version of BrIcc, called BrIccS, will be used. The executable file allows a matrix of ICC values to be retrieved and displayed all at the command line. By using this executable, PROGRESS can automatically import and write the necessary ICCs into the DICEBOX input file without needing to interface with an online database. This automatic writing of ICCs into the input file significantly reduces the potential for error versus a user manually copying and formatting each line of data.

The third block of data required for a DICEBOX input file is the block of known excited nuclear level data. A user-determined cutoff energy is defined as the energy below which the nuclear energy levels are well known and defined, and above which the nuclear energy levels are not well known or not well defined, and therefore simulations must be relied upon. Data in this block include the spin and parity of each nuclear level below the cutoff energy, the details of lower energy levels such as their spins and parities, and the associated likelihoods of deexcitation to each of these levels. These data are imported from the IAEA NDS via the Livechart application programming interface (API). By using the API, PROGRESS will directly request up-to-date excited level data each time the assembly package is used to create a DICEBOX input file. To maximize efficiency, PROGRESS only imports the specific data required for the input file, which represents a small slice of all the data which may be pulled using the API. Another script within PROGRESS then reformats the extracted data to match what is required for the DICEBOX input file, then sends the data to be assembled and written.

Preliminary tests of PROGRESS show that the functions described thus far work for several isotopes. Unforeseen issues are being resolved in the code as they manifest, and testing continues as input files are being automatically assembled for an increasingly wider set of isotopes. Once sufficient testing of the assembly package has been completed, focus will shift toward the development of the second part of PROGRESS, the output file interpretation package. This package will be responsible for collecting the simulated prompt gamma emissions output from DICEBOX and reformatting the data for use in data analysis. Currently, the output interpretation package consists of a script which collects the simulated prompt gamma data and plots the counts of prompt gammas as a function of gamma energy. An example of this functionality is shown in Figure 2.

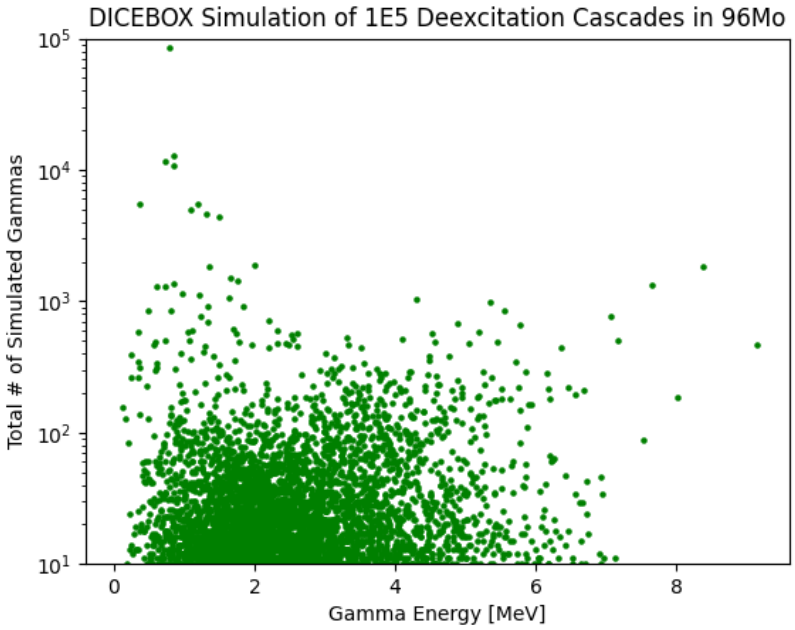

Figure 2: DICEBOX simulation of 1E5 prompt deexcitation cascades from ^{96}Mo, with number of simulated prompt gammas plotted as a function of gamma energy.

Future additions to this package will allow for PROGRESS to be used to construct a library of simulated prompt gamma responses for a wide variety of materials relevant to the nuclear fuel cycle. The prompt gamma response of a given material can be formatted as the intensity of a few of the most prevalent prompt gammas per neutron capture. From this, a user simply needs to know the macroscopic cross section of a material to generate an expected prompt gamma response.

Materials of Interest

Prompt gamma activation analysis (PGAA) offers two distinct benefits for use in nondestructive analysis. The first is that almost all nuclei which undergo neutron capture will emit a prompt gamma deexcitation cascade, regardless of the radioactivity of the resulting nucleus. Traditionally used delayed gamma spectroscopy with NAA is limited in its usefulness to materials that generate radioisotopes upon neutron bombardment; PGAA avoids that limitation. The second benefit is that prompt gammas often have a higher energy than delayed gammas, and that gammas from external sources which may interfere with delayed gammas of interest are less likely to interfere with prompt gammas of interest. Furthermore, due to the truly "prompt" nature of prompt gammas, in that they are produced within a nanosecond of neutron capture, any possibly interfering external gammas may be excludable from a prompt gamma spectrum based on time correlations [2]. Several elements are being investigated as possible candidates to illustrate the usefulness of PGAA as a non-destructive tool for nuclear forensic characterization.

One element with significant application to the field of nuclear forensics is molybdenum. Molybdenum is a naturally occurring trace element which exhibits some geochemical similarities to uranium and can be commonly found in uranium-bearing ore deposits around the world [6]. During the milling stage of the nuclear fuel cycle, uranium is extracted from crushed ore through chemical leaching and formed into a uranium oxide concentrate (UOC). Due to its geochemical similarity to uranium, a significant amount of molybdenum can also end up in the UOC. If the concentration of natural molybdenum in any given UOC sample can be measured, then the original concentration of molybdenum in the uranium-bearing ore can be estimated. From this, inferences into the sample's provenance can be made, as the concentrations of trace elements such as molybdenum can vary significantly on the location and type of ore deposit from which the sample was originally mined. Naturally occurring molybdenum consists of seven isotopes, but does not include ^{99}Mo, which is a strong beta emitter. ^{99}Mo is created either by neutron capture on ^{98}Mo or as a fission product. Several other isotopes of molybdenum are also commonly found in spent nuclear fuel as fission products. It is evident that molybdenum is an element of interest in the nuclear fuel cycle in that it can provide information about both sample provenance and previous neutron fluence through the investigation of its isotopic ratios. However, of the seven isotopes found in natural molybdenum, only three become radioactive upon neutron capture. This means that it is not possible to obtain a complete picture of molybdenum isotopics in a sample with traditional NAA alone, as only some resulting isotopes will generate a delayed gamma signal. Molybdenum isotopic ratio analysis is currently primarily performed using a destructive technique such as inductively coupled plasma mass spectrometry (ICP-MS); however, the use of PGAA might offer an alternative, non-destructive technique with which to perform this analysis.

Several rare-Earth elements (REE) found in uranium-bearing ores can provide similar nuclear forensic insights. Europium, gadolinium, samarium, and dysprosium, to name a few, are all metals which are found in geologic deposits in trace amounts. Both intra-elemental isotopic ratio analysis such as that commonly performed with molybdenum and analysis of ratios of elemental concentrations between REE can provide clues toward the geologic provenance of a sample [7][8]. Many REE have naturally occurring isotopes which remain stable upon neutron capture, minimizing the possibility of measuring their concentrations with traditional NAA. The naturally occurring isotopes of gadolinium, for example, mostly remain stable upon neutron capture. The $^{158}Gd/^{157}Gd$ isotope ratio within a sample provides insight into the neutron fluence, as ^{157}Gd has the greatest known thermal neutron capture cross section of any stable nuclide [9]. The resulting ^{158}Gd, however, remains stable, and therefore cannot be assayed using delayed gammas. Current work is underway with PROGRESS to predict the prompt gamma response from REE to investigate the feasibility of using PGAA for such efforts.

CONCLUSIONS

Prompt gammas present an underutilized source of signals of interest for nuclear forensic characterization. Prompt gammas are already produced during any use of NAA. However, because hundreds of different prompt gamma energies are possible per isotope, and these energies can range from hundreds of keVs to approximately 10 MeV, the spectroscopic presentation of prominent prompt gammas might be less apparent versus the more familiar delayed gammas. The aim of this work is to create a library of expected prompt gamma responses for different materials found throughout the nuclear fuel cycle so that a researcher utilizing NAA for non-destructive material assay knows how and where to look for the prompt gamma signal. DICEBOX is a cornerstone of this effort, allowing for the simulation of prompt gammas which may not be well documented in databases. As PROGRESS continues to be developed, it is hoped that the process of constructing DICEBOX input files, running them, and interpreting the simulation output continues to become a more efficient process. Future additions to PROGRESS may include the support of machine learning models, which could be used to identify a sample by comparing its experimentally obtained prompt gamma spectrum across hundreds of expected prompt gamma spectra within the simulated response library. Use of the novel chopper system at Penn State will allow for NAA to be performed with epithermal neutron energies and boost the system's sensitivity to trace elements by making use of neutron capture resonances in the epithermal energy range.

ACKNOWLEDGEMENTS

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